Nevada Test Site–Directed Research and Development

FY 2008 Report

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Report Date: April 2009

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SDRD FY 2008 Program Summary

The Site-Directed Research and Development (SDRD) program completed a highly successful year of innovation and technology advancement in support of the Nevada Test Site (NTS) mission and in meeting the needs of the National Nuclear Security Administration (NNSA). Highlights this year include numerous accomplishments that directly showcase the impact of our research and development investment. Eight SDRD-developed technologies were integrated into programmatic efforts, three of which represented new nonproliferation efforts. Eleven new invention disclosures were filed, six journal publications were generated, and two R&D 100 Award submissions were put forth this year. Furthermore, our participation in the Laboratory Directed Research and Development (LDRD) and SDRD Symposium in Washington, D.C., highlighted two national security projects that garnered attention from a broad audience including attendees from NTS, NNSA, National Weapons Laboratories (NWL), the Department of Homeland Security (DHS), and the Department of Defense (DoD).

The SDRD program has seen a dramatic increase in the yearly total of submitted proposals—from 69 in FY 2002 to 187 in FY 2008—while the number of projects funded has actually decreased from a program high of 57 in FY 2004 to the 21 newly selected projects for FY 2009. The overall effect of this trend has helped ensure an increasingly competitive program. Proposals were evaluated for technical merit, including such factors as innovation, probability of success, potential benefit, and mission applicability. Authors and reviewers benefited from the use of an R&D guidance document entitled the “NTS Technology Needs Assessment,” compiled from NTS, NWL, and NNSA sources. This tool continues to provide significant value in aligning the SDRD program with mission priorities.

The reports contained herein cover SDRD project activities that occurred from October 2007 through September 2008. The technical achievements described are a tribute to the skill and enthusiasm principal investigators brought to their individual projects. While many of the R&D efforts drew to successful and natural conclusions, some spawned follow-on work that may lead to further research. The desired result of all SDRD activities is to develop and/or refine technologies that are ultimately implemented by programs. Some of the following project reports clearly identify R&D efforts with those kinds of results. Others resulted in negative findings—the entirely valid conclusion often reached in the pursuit of “high-risk” research that a particular technology is currently impractical. Both types of results help make National Security Technologies, LLC, more agile by identifying solutions that can be directly applied to our programmatic mission.

The SDRD program is a multi-team effort, and my sincere gratitude goes out to Katharine Kelly Streeton and Michele Vochosky for compiling, editing, and publishing this report; to Nancie Nickels for her support with cover and divider graphics; to Project Controls Engineer Dennis Wai for his valuable contributions in tracking costs on the array of FY 2008 projects; to Casey Hulet and his team for compiling the financial data for congressionally mandated reporting requirements; and to

SDRD site representatives Rob Buckles, Paul Guss, Chris Hagen, Steve Iversen, Rick Maurer, Al Meidinger, Joe Meachum, Bill Nishimura, and Carson Riland, for helping implement and manage a very productive year of R&D. Also, program support by Lynn Veaser and Larry Franks is gratefully acknowledged.

And finally, this year marked the departure of Wil Lewis as the SDRD Program Manager. His record of achievement from the birth of the program to where it stands today speaks for itself.

Howard Bender, Acting SDRD Program Manager

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A

AC	alternating current
ACN	acetonitrile
ADC	analog-to-digital converter
AGC	automatic gain control
A-O	acousto-optical
AP	access point
API	application programming interface(s)
ASE	amplified stimulated emission
ASIC	application-specific integrated circuit

B

BAT	Burst Alert Telescope
BCC	body-centered cubic
BNL	Brookhaven National Laboratory

C

CAD	computer-aided design
CBE	chemical, biological, and explosive
CBM	coherency-based multiplexing
CCD	charge-coupled device
CCX	Cisco compatible extensions
CDD	concealed directional detector
CeBr ₃	cerium bromide
CLYC	cesium lithium yttrium chloride
CM	consequence management
COTS	commercial off-the-shelf
CP-SH	conjugated polymer with thiol endcaps
CW	continuous wave
CZT	Cd _{1-x} Zn _x Te [ternary compound]

D

DAC	digital-to-analog converter
DARHT	dual-axis radiographic hydrodynamic test (facility)
DC	direct current
D-D	deuterium-deuterium
DHS	U.S. Department of Homeland Security
DoD	U.S. Department of Defense

DOE	U.S. Department of Energy
DOP	degree of polarization
DPF	dense plasma focus
DPF-FPA	dense plasma focus fast-pulsed assembly
DPF-NRS	dense plasma focus neutron resonance spectroscopy
D-T	deuterium-tritium
DU	depleted uranium

E

EAP-TLS	Extensible Authentication Protocol-Transport Layer Security
EDA	electronic design automation
EDFA	erbium-doped fiber amplifiers
e-h	electron-hole (pair)
EMDS	Electromagnetic Design System [Agilent]
EOS	equation of state
EPM	embedded piezoresistive microcantilever
ESSID	extended service set ID
EVDO	Evolution Data Optimized [cellular modem]

F

FBG	fiber Bragg grating
fcc	face-centered cubic
FCC	Federal Code of Communications
FFT	fast Fourier transform
FOM	figure of merit
FOV	field of view
FPA	fast-pulsed assembly
FPGA	field programmable gate array
FWHM	full-width at half-maximum
FY	fiscal year

G

GPS	global positioning system
GRIS	gamma-ray imaging spectrometer
GRSI	Goddard Rad Source Imager
GSFC	Goddard Space Flight Center
GUI	graphical user interface

H

HCP	hexagonal close packed
HE	high explosive
HEDP	high-energy density physics
HPGe	high-purity germanium
HV	high voltage

I

IED	improvised explosive device
IR	infrared

J

JFET	junction field-effect transistor
------	----------------------------------

L

LaBr ₃	lanthanum bromide
LANL	Los Alamos National Laboratory
LANSCE	Los Alamos Neutron Science Center
LAO	Los Alamos Operations (NSTec)
LBNL	Lawrence Berkeley National Laboratory
LDRD	Laboratory Directed Research and Development
LED	light-emitting diode
LGB	lithium gadolinium borate
LIBS	laser-induced breakdown spectroscopy
LIFI	laser-induced fluorescence imaging
LINAC	linear accelerator
LLNL	Lawrence Livermore National Laboratory
LO	Livermore Operations (NSTec)
LOD	limit of detection
LOS	liquid organic scintillator(s)
LVDS	low-voltage differential signaling

M

MCNP	Monte Carlo N-Particle [transport code]
MCNP _x	Monte Carlo N-Particle Extended version [transport code]
MCP	microchannel plate
MEF	metal-enhanced fluorescence

MFP	mean free path
MHD	magnetohydrodynamic
MHRDR	magnetohydroradiative dynamic research [code]
MIRA	microwave infrared and acoustic [instrument]
MPDV	many-point photon Doppler velocimetry
MS	mass spectrometer

N

NaI(Tl)	sodium iodide doped with thallium
NASA	National Aeronautics and Space Administration
Nd:YAG	neodymium:yttrium-aluminum-garnet (laser)
NESTT	Non-explosive Security Testing and Training
NIF	National Ignition Facility
NIST	National Institute of Standards and Technology
NLV	North Las Vegas (NSTec)
NNSA	U.S. Department of Energy, National Nuclear Security Administration
NP	nanoparticles
NPBS	non-polarizing beam splitter
NPTEC	Nonproliferation Test and Evaluation Complex
NRCC	Nevada Radiological Computational Center
NRS	neutron resonance spectroscopy
NSLS	National Synchrotron Light Source
NSPD	nanosecond single-photon detector
NSTec	National Security Technologies, LLC
NTOF	neutron time of flight
NTS	Nevada Test Site
NWL	National Weapons Laboratory/Laboratories

O

OS	operating system
----	------------------

P

PBS	polarizing beam splitter
PC	principal component (band)
PCB	printed circuit board
PD	photodiode
PDA	personal digital assistant
PDV	photonic Doppler velocimetry
PL	photoluminescence

PM	polarization multiplexing
PMT	photomultiplier tube
POSS	polyhedral oligomeric silsesquioxanes
PSD	pulse-shape discrimination
PVT	polyvinyl toluene

R

R&D	research and development
REDAR	Radiation and Environmental Acquisition Recorder
RF	radio frequency
RITS	radiographic integrated test stand
RMS	root mean square
ROM	read-only memory
RSL	Remote Sensing Laboratory (NSTec)
RSL-A	Remote Sensing Laboratory – Andrews Operations
RSL-N	Remote Sensing Laboratory – Nellis Operations

S

SDRD	(Nevada Test) Site-Directed Research and Development
SNL	Sandia National Laboratories
SNM	special nuclear material
SNR	signal-to-noise ratio
SOA	semiconductor optical amplifier
SP	surface plasmon
SPL	Short-Pulse Laser (Laboratory)
STL	Special Technologies Laboratory (NSTec)

T

2-D	two-dimensional
3-D	three-dimensional
T&E	Test & Evaluation
TEM	transmission electron microscope
THF	tetrahydrofuran
THz	terahertz
THz-TDS	terahertz time-domain spectroscopy (system)
TOF	time of flight [measurements]

U

UCSB	University of California, Santa Barbara
UMPC	ultra mobile personal computer
UNLV	University of Nevada, Las Vegas
USB	universal serial bus
UV	ultraviolet

V

VCD	vacuum Compton detector
VISAR	velocity interferometer system for any reflector
VoIP	Voice over Internet Protocol

W

WDM	wavelength division multiplexing
WEP	wired equivalent privacy
WPA	Wi-Fi Protected Access

X

XRD	x-ray diffraction
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ACCELERATORS AND PULSED POWER

PICOSECOND TIME-RESOLVED ELECTRON DIFFRACTION OF PHASE TRANSITIONS

Howard Bender,^{1,a} Dave Schwellenbach,^a Ron Sturges,^a Nick Wilcox,^a Rusty Trainham,^b and James Potter^c

The purpose of this research was to explore using megavolt relativistic electron beams and electron diffraction to do pump-probe studies of phase transitions with picosecond time resolution. The motivation here was to provide a new capability, and tool, for advanced material studies to support hydrodynamics research in weapon physics. Like x-ray diffraction, electron diffraction provides a direct method to interrogate solid-state structures and until recently has been performed mainly with lower-energy kilovolt beams from transmission electron microscopes (TEM). However, using relativistic beams offers potential advantages, such as reduced effect of space charge and the ability to penetrate thicker samples. Furthermore, linacs can easily produce the pulse bunches that will enable resolution studies in the picosecond range. A new 2-MeV short-pulse linac located at Los Alamos Operations (LAO) was used to record static diffraction patterns in polycrystalline metals. This report describes our first attempts at diffraction experiments and focuses on the performance demands required of the accelerator to record high-fidelity diffraction patterns.

Background

A fundamental understanding of material properties in the high temperature and pressure regime is critical to the weapon physics community. Experimental studies in these extreme conditions were previously the purview of underground nuclear testing, and more recently were, and continue to be, the focus of predictive computer-based modeling. In order to perform accurate simulations, it is extremely important to know a material's equation of state (EOS). Experimental data are needed to verify models and refine EOS to obtain accurate simulations. Also essential are the details of phase transition and melt properties, but these are not generally well-described by current models. Thus, a need exists to provide experimental tools to diagnose phase transitions. Many techniques have been proposed and implemented, such as x-ray diffraction (Morgan, 2007) and THz spectroscopy (Kruschwitz, 2008).

Our work was motivated by a new potential phase-transition diagnostic based on recent results by the Stanford Linear Accelerator Center (Hastings, 2006) that demonstrated single-shot diffraction patterns could be produced with 5-MeV electron beams in thin samples of polycrystalline metals. This relatively straightforward technique transmits electrons through the sample to produce a diffraction pattern onto a phosphor that is then imaged with CCD cameras. The reduced space-charge

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effect of megavolt beams, the ability to penetrate thicker samples, and the ease of producing short pulses in linacs made this an attractive option for hydrodynamic materials studies. These experiments can be run in single-shot mode or with a pulse train that would provide a continuous sampling of the phase-transition dynamics. In fact, as mentioned in Hastings et al. (2006), this pulse train approach can create “movies” of phase transitions and structural dynamics.

Electron diffraction, which has been widely used by the analytical sciences community for nearly 100 years, is a complementary technique to x-ray diffraction, yet it has some distinct advantages relevant to material dynamics studies. For instance, the radius of the Ewald sphere, which gives the range of constructive interference of the reciprocal lattice, is proportional to $1/\text{wavelength}$. The de Broglie wavelength of an electron at 1 MeV is ~ 0.001 nm, as compared to a characteristic x-ray photon with a wavelength of ~ 1 nm. The shorter electron wavelength ensures that electrons will scatter off multiple lattice planes, thereby offering a two-dimensional look into the structure. In addition, electron diffraction is very sensitive to subtleties in the lattice structure, such as defects, dislocations, and the like. It has also been shown that electron diffraction using a TEM can clearly distinguish between solid transitions, such as the HCP (hexagonal close packed) $\alpha \rightarrow$ BCC (body-centered cubic) β in Ti films.

The goal of this investigation was to extend electron diffraction at megavolt energies with picosecond resolution as a phase-transition diagnostic with high temporal resolution. We expected to explore both solid-liquid and solid-solid phase transitions. Initial experiments focused on recording static diffraction patterns with picosecond electron bunches generated by the linac. However, we quickly determined that important characteristics of the linac, such as beam control, were not adequate for high-fidelity diffraction imaging. Thus, much of the effort was directed at improving accelerator performance and refining our experimental setup.

Project

LINAC Design and Operation

The LAO linac was recently commissioned and provided the platform for which to pursue our diffraction studies. This system was built around a 100-kV injector, a spare injector to the 20-MeV linac once located at Special Technologies Laboratory (STL) and later decommissioned in the early 1990s and sent to Idaho State University. We have since upgraded the machine by adding an S-band RF accelerating section to boost the energy to 2 MeV. The performance of the injector itself was significantly enhanced while executing an FY 2007 SDRD project (Sturges, 2008) and provided a sub-nanosecond injection pulse. The klystron and modulator were tested to full power in January 2008, and the S-band accelerator sections were installed and vacuum-conditioned in February. The RF waveguides were installed and power-conditioned in March and the first accelerated beam was produced in April. A full description of the accelerator and RF systems can be found in a recent paper by Bender (2008).

Beam Line Layout and Operation

Our initial concept for electron diffraction, borrowed from techniques used in TEM systems, was based on simple transmission geometry with focusing electron optics. The first-generation beam line is shown in Figure 1. Basic solenoid transport is used throughout the length of the beam line up to the downstream section at the exit of the accelerator cavities. The downstream section uses quadrupole triplet focusing magnets to provide small collimated spots on the target and also images the diffraction patterns. The target chamber houses thin foils for electron diffraction and the imaging chamber houses a phosphor screen.

Figure 1. Schematic layout of the linac used for the initial electron diffraction experiments

As part of this project, we performed the first energy and current measurements on the fully assembled accelerator. Beam energy in short-pulse mode (1 ns) was measured with a 60-degree-bend magnet spectrometer that was previously calibrated using a negative ion source (Trainham, 2005) for use at ~20 MeV. For this measurement, the magnet current was reduced to a few amps; reliable energy measurements obtained in the 0.3 to 2.0 MeV range are shown versus input RF power in Figure 2a. Energy resolution or spread was nominally <5%.

Figure 2. Fundamental beam characteristics of the linac: (a) beam energy vs input RF power; (b) beam pulse-width measurement

A plot of beam current as a function of time as measured with a fast Faraday cup is shown in Figure 2b. The injector pulse at approximately 1 ns and the RF modulation at 2.8 GHz are clearly seen. Individual pulses in the train are limited by the 80-ps Faraday cup response.

Electron Diffraction Studies

Simulations and Experimental Considerations

Electron diffraction using megavolt beams presents some immediate challenges that must be understood and addressed. First, the scattering angle, which is highly energy-dependent, becomes fairly small as energy increases (Figure 3). Also, the intrinsic beam divergence, which convolves with the diffraction pattern, may blur and wash out the pattern; this has been described by Wang (2006). Energy spread in the beam, ΔE , further contributes to smearing of diffraction patterns because the image is a summation of all the wavelengths in the energy band. Wang (2006) also points out that x-rays with $\Delta E = 2\text{--}5\%$ did not show a significant difference in modulation over monochromatic x-rays. Other references suggest that an energy spread of $<1\%$ is sufficient for good diffraction imaging. Beam divergence is most often controlled by using smaller pinholes and collimating the beam with long apertures.

Figure 3. Scattering angle vs energy for Al major diffraction peaks

To further understand these effects, we performed preliminary simulations to understand imaging diffraction rings in simple polycrystalline metals. For the diffraction simulations we used the Amaze series of codes from Field Precision (Humphries, 2008). Beam current distributions and emittances were produced by Gendist, and a separate C program used the output from Gendist to generate the Bragg angle diffraction components. Finally, Omnitrack was used to calculate the beam transport and emittance at the detector plane. Figure 4 shows the simulations for polycrystalline Al. With idealized zero emittance, the modeled four indices (111), (200), (220), and (311) are clearly resolved. However, adding a reasonable emittance estimate of 0.3π mm-rad causes broadening and smearing of the rings. Figure 4 also shows the image distortions introduced by the quadrupole lenses.

Figure 4. Electron diffraction simulations for polycrystalline Al. Modeled diffraction indices are (111), (200), (220), and (311).

Experimental Results

Based on simulations and experience with our first-generation beam line, we constructed a new beam line with improved collimation for better beam control. In addition, we removed the quadrupole triplet focusing magnets, as they made operation more complex and induced distortion as just described. A photo of the second-generation setup is shown in Figure 5. Two target ladders, each with graphite collimators ranging in size from 0.5 to 2 mm, were used. The upstream collimator was 25 mm long. The downstream collimator, which held the metal foil sample, was 12 mm long. A microchannel plate (MCP) was used with a lens-coupled CCD to image either a pure beam or transmitted beam through foil.

We obtained transported beam images using a variety of collimator diameters; these images are shown in Figure 6a and b. Lineouts of the images are also shown with the FWHM spot size. As can be seen, the 0.5-mm collimator produced the best spot at 2.2 mm. The latter spot implies a divergence of approximately 3 mRad.

Figure 5. Second-generation beam line setup for diffraction experiments

A variety of metal foil targets were then evaluated to obtain diffraction patterns. Our attempts at using very thin Al TEM standards (~ 50 nm) to image diffraction patterns were inconclusive. A thicker $0.5\text{-}\mu\text{m}$ Sn foil was also tested and the image we obtained is shown in Figure 7. No discernible diffraction rings were observed here either.

Discussion

Multiple issues became apparent in our endeavor to produce electron diffraction patterns. Beam divergence is crucial, and additional collimation is needed as well as implementing smaller pinhole diameters to improve lateral coherence. Emittance has not been measured; however, previous data from the STL linac suggest that the injector alone is about 0.1π mm-rad. Emittance growth in the cavities is a concern and should be explored in future work. Our second-generation beam line had no means for precise energy selection and future work will also require use of the spectrometer as a monochromator.

Figure 6. Images and lineouts of transported beam (no foil target) with (a) 1-mm collimators and (b) 0.5-mm collimators (grid seen in the images is a 1-mm gap between MCP detectors)

Figure 7. Image of beam transmitted through 0.5-μm Sn foil

A further issue with sample thickness has also been explored by experimental means, yet optimum thickness was only recently addressed more analytically. The optimum thickness can be determined by examining the scattering function. The differential cross section increases with energy as expected; however, the total elastic scattering cross section, as given by

$$\sigma_{TOTAL} = 2\pi \int_0^{2\pi} \frac{d\sigma}{d\Omega} \sin(\theta) d\theta \quad (1)$$

reaches a constant due to the fact the smaller scattering angles and reduced total area compensate for this increase (Rudakov, 2006). The mean free path describes the average distance between scattering events, and is given by

$$1/\sigma_{TOTAL} N. \quad (2)$$

This equates to ~200 nm for Al at 1 MeV. A thickness of one mean free path (MFP) ensures the highest probability of single scattering and, based on Poisson statistics (Rudakov, 2006), yields a probability of 0.37. We have recently obtained a foil that is exactly 200 nm thick that can be used for additional experiments.

Conclusion

We proposed using megavolt relativistic electron beams and electron diffraction to do pump-probe studies of phase transitions with picosecond resolution. We have performed a proof-of-principle experiment and examined performance issues and improvements to the linac for diffraction studies. This was a sizeable effort, and very successful, with the linac now operating at nominal 0.5–2.0 MeV energy with sub-100-ps bunched pulses. Multiple samples were tested, and our experiments revealed that beam control is a critical issue when trying to obtain high-fidelity diffraction pattern images. Methods to improve beam divergence and control emittance were established. It is expected that once these are achieved, the desired diffraction pattern should be forthcoming and dynamic experiments with a laser-driven shock or heating to induce solid-solid or solid-liquid phase transitions should be possible.

Acknowledgments

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TIME-DEPENDENT NEUTRON IMAGING ON A DENSE PLASMA FOCUS

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A system was designed to image fusion-produced neutrons from a dense plasma focus (DPF). Images were formed using a tungsten occluder with a slit aperture. The detector was an array of scintillators that were aluminized on the outside and shaped to focus the volume scintillation source onto optical fibers that carried the signals to a streak camera. The system was designed for a yield of 3×10^{12} neutrons at an energy of 14.1 MeV (deuterium-tritium [D-T] fusion neutrons). Data were taken on a DPF burning deuterium only (D-D neutrons at 2.45 MeV) at a yield of around 2×10^{10} . Even at this low yield, important physics results were obtained. It was seen that the temporal history of the neutron emission was sometimes double-peaked. The neutron-emitting region was seen to move vertically off-center, and then return to the centered position.

Background

The long-term goal of every fusion-generating device is to increase yield. Using fusion products to diagnose these devices is the only reliable way to understand the fusion process. Total yield, time history, and neutron imaging are the diagnostics most important to this goal.

Previous neutron imaging efforts have mostly used “pinhole” occluders to produce two-dimensional (2-D) images. In order to improve resolution, the aperture has been narrowed until very few neutrons are detected on present-day laboratory fusion sources. This project pursued a very different design philosophy: getting adequate signal was the first priority, and everything was built around that.

Project

Our first design featured an occluder with eight slits. This design gives orders of magnitude more throughput than neutron imaging systems now being fielded on inertial confinement fusion experiments, for example. A mathematical reconstruction of the data provides time-dependent 2-D images. Eight scintillator arrays detect the neutrons and send their signals to streak cameras via optical fiber. The scintillators are aluminized on the outside, and shaped so as to optimize light gathering from the volume (scintillation) source. (Each scintillator is referred to as a “paddle,” for reasons evident from Figure 2.) This eight-slit design was judged to be too ambitious, so time-dependent imaging in one spatial dimension has been pursued. One-dimensional imaging can be accomplished with a single slit. The axial direction of the discharge was chosen, because it was expected to show the most interesting physics.

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An optical code was written to calculate the light-gathering ability of a scintillator paddle machined from a sheet with a shaped tool bit. Such a design gives focusing in two orthogonal planes, each containing the optical fiber axis. This code was written and very close to success when it was decided that a back-of-the-envelope design would probably gather at least half as much light as an optimized design, so further development of the code was suspended in the interests of project schedule.

We were given the opportunity to “ride along” on two DPF experiments: a series of shots on the D-D machine in North Las Vegas (NLV), and another series on the D-T machine located at the Nevada Test Site (NTS) scheduled for the last two weeks of September. We planned the D-D shots to allow testing of the streak camera in the presence of electromagnetic pulse and neutron noise, and to practice aligning the system. We did not know whether to expect any useful data at this low yield and at the lower D-D neutron energy. (Scintillator light yield scales linearly with deposited energy.) We were delighted when we got several good data sets, which are shown below. Because the D-T shots were not completed by the end of this SDRD project, the results are not included in this report.

The basics of neutron imaging are shown in Figure 1, where o is the object distance, i is the image distance, and A is the opening angle.

Figure 1. Basic schema of neutron imaging

The design of the system involves choosing values for o , i , and A to match the characteristics of the source, detectors, and streak camera and to maximize some combination of resolution and signal-to-noise ratio. In order to produce an optimized design, at least approximate knowledge of the source and detectors is needed at the outset. At the time of the design of the system, this knowledge was lacking. In addition, it was not known how to treat the problem of nuclear cross talk (explained below). These experiments should be regarded as the first step in an iterative process of optimization.

The occluder, made of machinable tungsten that is 80% W and 20% Cu by weight, is 20 cm long, and the opening angle is 0.169 degrees. It was designed for D-T neutrons, which have a range of 3.2 cm in machinable tungsten, versus 2.6 cm for D-D neutrons. It was also designed for different image and

object distances than those used to take the data shown in this report. In the experimental situation, the occluder and detector were moved closer to the source to get more signal at this low D-D yield. The result was a very small field of view.

The scintillator paddle design is shown in Figure 2. The paddles are made of St. Gobain BC-408 plastic scintillator, which responds to neutrons by emitting blue-violet light. The paddle thickness is 1 mm.

Figure 2. Scintillator paddle design (dimensions are in inches and [millimeters])

The slot at the bottom of the paddle receives the optical fiber, which is glued in with index-matching epoxy. The optical fibers are type JTFSH from Polymicro. The silica core is 1000 μm in diameter, and has a hard polymer cladding. The numerical aperture of 0.37 is higher than that of most optical fibers. Because the fiber cladding is actually thicker than the paddle, the stack was staggered, so that every paddle was displaced laterally from those above and below it.

The shape of the paddles was obtained by treating the top surface as if it were a spherical mirror, and choosing the radius of curvature to focus the centroid of the area onto the tip of the fiber. This quick design is probably at least half as efficient as one that was properly optimized.

Previous effort suggested that total internal reflection is the most efficient means of gathering light, and that the design would be more effective if the aluminized surface were replaced with a dielectric cladding (Lewis, 2008). This hypothesis could easily be tested in this context by the optical code that was written. The expectation of the author is that the most efficient collector would be aluminized, but have a greater ratio of thickness to length and width than the present design. The thickness was forced to be less than desirable, for the following reason. The NTS DPF is situated in a pit, so the line of sight is angled down at 25 degrees. Thicker paddles would have given greater magnification, necessitating a longer image distance and putting the detectors above the roof of the building. These thin detectors require many bounces before a photon is collected. At each bounce, there is some absorption, so in the end, the light gathering is not very efficient, and these thin paddles might well be more efficient if they relied on total internal reflection. A follow-on effort to this project would certainly revisit the issue.

A total of nine shots showed some signal, of which three gave usable data. Figure 3 shows the data from shots 2, 8, and 9, in order from left to right. Time in nanoseconds is along the x axis, and the 29 fibers are spread out along the y axis. The width of each plot corresponds to 830 nsec.

Each data set first had the minimum value removed, then the stars. (Stars are randomly situated groups of pixels with anomalously high signal values. They are believed to be caused by direct interactions of stray neutrons with the camera.) Each plot was scaled individually, so that the smallest datum in the set is represented by black, and the largest by red. Because the signal was weaker for shot 2, the plot has lower contrast.

Shot 2 had a yield of 1.96×10^{10} , with an image distance (i) of 1.94 m, and an object distance (o) of 0.54 m. These are not the parameters for which the system was designed; rather, the distances were diminished to get more signal on these low-yield shots, although the result was a small field of view. Shot 8 had a yield of 1.90×10^{10} , an object distance of 0.54 m, and an image distance of 0.17 m. Shot 9 yielded 2.5×10^{10} , there was no occluder, and the distance between source and detector was 0.71 m.

At the NTS, the discharge axis is vertical, so the imaging system was designed with the slit and scintillators horizontally oriented. At NLV, the axis of the discharge is horizontal. Because the D-D shots were done to validate the hardware and practice alignment, the orientation used was what we planned to use for the D-T shots, so it was actually the direction perpendicular to the DPF axis that was imaged during the D-D shots.

It was originally planned that the point-spread function would be deconvolved from the data, but deconvolution has a strong tendency to magnify noise; therefore, we did not apply the process to this already noisy data set.

Figure 3. Data from shots 2, 8, and 9

Figure 4 shows the signals summed over all pixels in the y direction of Figure 3, which gives the entire signal gathered by the system for each shot, as a function of time. The triggering of the scope was chosen as the zero time for display of each trace. The relationship to the beginning of the electrical discharge cannot be inferred from the plot.

During the experiment, 2-in.-thick lead bricks surrounded the detectors, and half-inch lead shielding was placed in front (between source and detectors). Even so, it was at first believed that the first peak was due to gamma rays and that the second was caused by neutrons. If this were the case, the time between the two peaks should have been 107 ns for shot 2, and 30.6 ns for shots 8 and 9, for which the detector is closer. One would also expect that the areas under the two peaks would bear a constant ratio for all shots, because it would reflect only the relative sensitivity of the detectors to gamma rays versus neutrons. These expectations were not fulfilled. For each shot an eleven-parameter function

Figure 4. Total signal as a function of time

comprising two peaks gave an excellent fit to the data. The delays between the two peaks for shots 2, 8, and 9 are 161, 147, and 186 ns, respectively. The ratios of the areas of the two peaks are 0.35, 0.30, and 0.12, respectively. Not all shots produced dual peaks. Shots 1, 3, and 4 appear to be single peaks, although the signals are faint. Dual peaks are sometimes, but not always, recorded by another diagnostic used at the DPF. We must thus reject the hypothesis that one peak is gamma rays and the other neutrons. Other possible mechanisms were considered. The first peak could be direct neutrons, and the second could be neutrons scattered off the floor or some other object, neutron-induced secondary emission, neutrons down-shifted in energy passing through the occluder, or interactions of the direct neutrons with the detection system (for example, scintillation in the optical fibers). None of these mechanisms yields predictions for ordering among the shots of delays and ratios of areas that are consistent with the data. It can be concluded that the recorded time signals are due to the time histories of the neutron emission. (Alternate mechanisms might still contribute on a lower level.)

The data in Figure 3 for shots 2 and 8 were summed over the width of each fiber, and then the 29 time-dependent signals were flat-fielded by the data from shot 9 (no occluder). Figure 5 shows the result for shot 2.

Figure 5. Data from shot 2, summed over each optical fiber

The y axis gives position in the object plane. In the configuration used, the field of view of the imaging system is 3.2 mm. (The field of view is determined by the imaging geometry, and should not be confused with the width of the plot.) It can be seen that there is a bright region in the center position, and also brighter portions at the top and bottom of the plot, but that the image is fairly flat in the y direction, with the time history shown in Figure 4. If neutron imaging were working according to design, only the central brighter portion would appear, and the parts of the image above and below would have a low signal. There are three problems that cause cross talk between scintillator paddles and contribute to a flat distribution. (Figure 3 seems not to show evidence of cross talk, at first glance. Signal leaking between channels on the camera would show cross talk between *fibers*. Cross talk between *paddles* shows up in the somewhat flat shape of the distribution in Figure 5.) Another problem arises from the departure of the image and object distances from their design values, and is explained below.

One source of cross talk is scattering of neutrons in the occluder and its holder. Neutron imaging works best when the detectors are far from the occluder, because the occluder does not actually stop neutrons; it merely scatters them. A good image can be formed only when most scattered neutrons miss the far-away detector. Both of the detector locations used for these shots were too close to the occluder, thus intercepting the scattered neutrons, which resulted in cross talk. The second source of cross talk was optical leakage between paddles. Each was aluminized for reflection, and then painted with white spray paint. Before they were sent out for the experiment, a bright light was shone onto the paddle end, and light was seen coming from the ends of the fibers. The solution to this light leakage problem was to place an aluminum shield between every pair of adjacent paddles, but there was not enough time to do this before the D-D experiment. The shields were put in place between the D-D and D-T experiment dates. The third source of cross talk is due to recoil of neutrons in the plastic. When a neutron hits a nucleus in one paddle, it can recoil into adjacent paddles and contribute to signal in them by subsequent collision. This source of cross talk is unavoidable. It should be calculated and measured, then deconvolved from the resulting signal.

Figure 6. Shot 8 showing movement of the source in time

Besides these three sources of cross talk, there is a further complication. The detector is wider than the occluder, and in this case, the two were so close together that the detector was not properly shadowed from the source. Neutrons could travel in straight paths around the occluder to reach the detector. The paddles on the top and bottom of the stack were shielded only by the aluminum holder, and all the paddles were shielded only by aluminum on their right and left edges. The expected result is a bright spot in the center caused by neutrons traveling through the slit as desired, but also bright areas on the top and bottom caused by neutron flux attenuated only by the aluminum, on top of a rather flat background caused by neutron flux hitting the right and left edges of the paddles. Cross talk also contributes to this flat background. This is just what Figure 5 shows, so we conclude that the central bright portion is an image of the neutron-emitting region with a width of about 1.5 mm. Because the point-spread function has not been deconvolved from the data, this is recognized to be an over-estimate of the true size. The expected width is about 1 mm.

Figure 6 shows shot 8, binned in time, as a function of position on the detector. A salient feature is that for the earliest and latest time intervals the distribution is quite flat, while for the intermediate time interval there is an unmistakable tilt up towards the left. This movement in time of the neutron-emitting region is not dramatic, but it is unambiguous. Because the field of view is too narrow, it is difficult to say how far the source moved, but it is clear that it did move. Previous x-ray imaging of the DPF at the NTS also showed lateral movement of the plasma (Berninger, 2007).

Conclusion

Imaging of D-D neutrons has been accomplished for the first time. Motion has been detected for the first time in an unclassified context. The time history was found to have two peaks for some shots. The imaging concept and hardware were validated in this low-yield, low-energy D-D regime. Plans have been laid for a neutron imaging system that would give images in two spatial dimensions and time.

To the best of our knowledge, D-D neutron imaging has never been seriously pursued, but we have achieved it. The use of deuterium only has enormous advantages in convenience and cost. D-D fusion produces a yield of only about 1% that of D-T fusion under the same conditions. However, the NLV DPF (D-D) has higher current than the NTS DPF (D-T), so it produces closer to 10% the yield of the D-T machine. In addition, the NLV DPF machine is due for an upgrade to increase its yield. The sensitivity of the neutron imaging system could be increased in a number of ways. There is a real possibility that a system could be designed for the D-D machine that would have significantly better resolution than this serendipitous first attempt.

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FEASIBILITY STUDY FOR DENSE PLASMA FOCUS ON ATLAS

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The Atlas machine at the Nevada Test Site (NTS) is a very large capacitive energy storage machine, designed for liner implosion experiments. These types of experiments bear many similarities to dense plasma focus (DPF) experiments, and the electrical characteristics of the Atlas machine are very similar to our existing smaller-scale plasma foci. This project revealed that doing plasma focus experiments at Atlas would be feasible. The Atlas facility appears to offer one of the most cost-effective means of investigating high-powered plasma focus research available at the NTS, and would allow for safely elevating the yields. The modification of the Atlas machine would be concentrated in the load region.

Background

The SDRD program has catalyzed progress in dense plasma focus research and development at National Security Technologies (NSTec), advancing neutron detection science, improving overall source performance, and increasing source output. The progression of sources from a small 1.5-kJ (0.0015 MJ) source through the present 0.133-MJ deuterium-tritium (D-T) fusion device at the NTS, and the use of deuterium fusion 0.5-MJ deuterium-deuterium (D-D) sources to the soon-to-be-operating 1-MJ source with the superheader, have been made possible with SDRD advances (Hagen, 2002; Pigg, 2003; Ziegler, “Enhanced,” 2004; Ziegler, “Particle-discriminating,” 2004; Hagen, “High-output,” 2005; Hagen, “Optimized,” 2005; Hagen, “Two-bank,” 2005; Ziegler, 2005; Meehan, “Design,” 2006; Meehan, “Novel,” 2006; Ziegler, 2006; Meehan, “Anode,” 2007; Meehan, “Dense,” 2007) It must be noted that all of these sources have been designed around a pool of existing capacitors and were constructed of switches and other pulsed power components from completed projects. There has been a steady progression of machine performance, as shown in Figure 1. The Gemini bank is presently under construction; it represents the highest voltage, and near to the highest power bank, that can be constructed with our existing components.

The Atlas machine, a very large capacitive energy storage machine, was designed for a rather different purpose than powering plasma foci. Even though the original mission of Atlas was liner implosion, this type of experiment shares many physical similarities with Z-pinching hydrogen isotopes in a DPF, as shown in Figure 2. On the left, a metal conductor is electromagnetically imploded into an implosion target. On the right, a highly ionized plasma shockwave implodes the trapped hydrogen isotopes on the z-axis.

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Figure 1. DPF progress and possible future development under SDRD

Atlas stores energy at high voltage in capacitors, then discharges this energy into a load through high-speed, high-current switches and high-power transmission assemblies. It can store 24 MJ at full voltage (240 kV) and is capable of delivering 20 MA to a DPF load. Because it can operate over a wide range of stored energies, Atlas is an ideal platform for running high-power plasma focus experiments.

This SDRD project investigated the feasibility and practicality of modifying the Atlas machine for use with a DPF load. Issues we addressed were:

- the degree of modifications needed for the 12 energy storage modules,
- if and how the tri-plate transmission lines should be modified, and
- how to scientifically design and engineer a DPF load to operate within the Atlas performance envelope.

Figure 2. Comparison of Atlas-type liner implosion (left) and DPF (right)

In addition, the performance of dense plasma focus tubes on Atlas was estimated, as neutron output and high energy density plasma physics were the driving focus in this project.

Project

The project proceeded through phases: facility investigation, development of magnetohydrodynamic (MHD) computer models of Atlas, and engineering design of Atlas-compatible DPF electrodes.

Facility Inspection

Atlas was visited many times to understand the construction of the bank and how the power flows from the capacitor banks through the transmission assemblies to the load station. An inspection of Atlas revealed that the equipment we would require was present.

The capacitor banks, designed to have a quarter-cycle time of $5\ \mu\text{s}$, are appropriate for operating a DPF without further modification. Two other important features of Atlas are that it can be operated over a range of bank charging voltages (and therefore a range of stored energies) and with as few as 4 of the 12 modules. Thus, it seems to be an ideal platform for a DPF development program, as it would allow for small-step growth with no modification of the machine, a great cost and time savings. The Atlas-era tri-plate transmission lines functioned well; no modifications were perceived as necessary. We had the opportunity to inspect a few Atlas load components and later used them when developing

the electrical interfaces for the DPF electrodes. Existing construction and engineering drawings of the Atlas facility were used as reference during interviews with Atlas personnel. These experienced staffers indicated that the central region of the components below the former liner assembly that originally housed Atlas liner packages could be modified to fit a DPF anode and cathode assembly. Also important is the fact that no modifications of the interface between the tri-plate transmission lines and the central assembly would be needed.

Atlas Computer Model Development

In order to design an anode-cathode assembly, it was necessary to estimate the current delivered to the DPF load, and to design the load to have the plasma sheath arrive at the end of the anode at the time of maximum current. Two DPF codes, MACH2 and an updated version of the magnetohydroradiative dynamic research (MHRDR) code (Makhin, 2001), were used in addition to an empirical circuit modeling code developed by Los Alamos National Laboratory, Scat95 (Goforth, 1998). Work with the MHRDR code was performed by staff at the University of Nevada, Reno, who modeled the performance of DPF tube designs coupled to Atlas. Based on experimental observations at a lower stored energy, a range of pressures required for proper plasma sheath operation was established (Gratton, 1977; Bruzzone, 1993). With the operating pressure restricted to less than 25 torr, anode-cathode physical parameters were then varied to achieve maximum yield in the codes.

The modeling results suggested that, for the cases studied, yields far beyond those presently observed with existing DPF machines could be expected. Both MHRDR and MACH2 are two-dimensional MHD codes. The plasma dynamic that generates the neutrons is a violent shock implosion and is not well simulated in two dimensions, so the neutron yield modeling mechanism is “calibrated” to match the yield of a given configuration in order to explore designs that are geometrically close to the “calibrated” model. The MHD models were calibrated using a well-known current scaling relationship (Freeman, 2002) that is based on the experimental results of dozens of DPF researchers around the world. The graphic in Figure 3 shows yield extrapolated to the predicted peak current levels delivered by Atlas.

It is not known how well this empirical relationship will extrapolate to larger stored energies or higher-current DPF designs. The MHD modeling done during this project did not reveal any evidence of yield saturation reported by other DPF researchers (Herold, 1989; Zakullah, 1995), and we note that yet other researchers report no saturation in yield using their MHD simulations (Coverdale, 2007). Experiments with stored energies as high as Atlas have been performed, and researchers have reported gas-puff yields of at least 10^{13} neutrons (Coverdale, 2007). Should DPF experiments be performed on Atlas, experimenters will search for this saturation effect. Atlas would provide the ideal platform for researching this phenomenon, since it is capable of storing energies well beyond the reported yield saturation limit.

Figure 3. Yield vs stored energy

Both MHD codes calculated the rundown time of the chosen geometry to be close to $5 \mu\text{s}$ (Makhin, unpublished). The calculated yield also scales with the same power law that is observed experimentally, and when used with the calibration factor, it predicts experimental yields quite well. The current and neutron production curves from MHRDR models are shown in Figure 4.

MACH2 computes a neutron production and bank current curve similar to that of MHRDR, and MACH2 is capable of modeling more complicated geometries. This capability allowed us to include actual installed shapes into the MACH2 model and avoid some numerical singularities associated with sharp corners in the MHD simulation. The MACH2 results are shown in Figure 5.

Design

After the proper electrode configuration was determined, we formally designed the DPF tube and electromechanical interface for Atlas. Engineering drawings that would be suitable for fabrication and installation of an actual DPF electrode in the Atlas load station were produced. Three-dimensional models of the design are shown in Figure 6.

Figure 4. MHRDR simulation of neutron production and bank current

Figure 5. MACH2 simulation of current and pressure data

Figure 6. Engineering drawings for Atlas DPF tube (left) and interface (right)

Conclusion

This work has shown that it is very feasible to use the Atlas facility for DPF research. The Atlas facility appears to offer one of the most cost-effective means of investigating high-powered plasma focus research available at the NTS, and would allow for safely elevating the yields in place from 1 to 24 MJ. The modification of the Atlas machine would be concentrated in the load region, while the remainder of the facility could be used as is.

The project also extended the MHD modeling capabilities, engineered and designed a high-current Atlas-appropriate load, and led to the definition of a technological growth path for DPF.

The promise of higher yields could lead to many new DPF applications, including neutron resonance spectroscopy, neutron imaging in support of the National Ignition Facility, and other uses. The Atlas DPF could open the possibilities of supporting nuclear forensics and fusion energy research.

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DESIGN OF A NEUTRON RESONANCE SPECTROSCOPY EXPERIMENT

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Neutron resonance spectroscopy (NRS) is a research tool that infers an element's temperature from measuring the change in the width of a neutron resonance absorption peak. NRS is usually performed at particle accelerator facilities capable of producing short, intense neutron pulses. If it were possible to use a dense plasma focus (DPF) as the source of the neutrons, an inexpensive NRS diagnostic could be added to many existing experimental facilities. This research explores the possibility of using the DPF for NRS.

Background

Most NRS experiments that have been carried out have used sources similar to the high-current, 800-MeV proton storage ring at the Los Alamos Neutron Science Center (LANSCE) (Yuan, 2005). NRS scans the sample with a range of neutron energies; the scanning occurs in a few microseconds and depends on the fact that lower-energy neutrons travel more slowly than higher-energy neutrons. In the preferred configuration, a short neutron pulse is generated in order to pass through the sample at the time of interest, and then is allowed to time-disperse over the flight path to the detector. The NRS spectrum is then recorded as a time-of-flight signal, and then analyzed using customized software. The width in energy of the resonance absorption line is measured, and the temperature of the sample at the instant the neutrons passed through the shocked sample is inferred.

If it were possible to use the DPF as the pulsed neutron source, an NRS facility that allowed each experiment to be optimized for its unique parameters could be conceived. NRS experiments could be done rather cheaply and on the experimenter's schedule. The primary benefit in using DPF-NRS would be lower facility costs, which would mean that experiments could be done more often and have more optimal experimental setups, as indicated by shortening the source-moderator distance and target-detector distance from Figure 1 to that of Figure 2.

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Figure 1. Existing LANSCE NRS setup (Yuan, 2005)

Figure 2. Proposed DPF NRS setup

Project

The primary goals of this project were to:

- Determine at what DPF fusion neutron output level the DPF could deliver epithermal neutron flux comparable to NRS experiments that have already achieved success.
- Determine if the time-resolution of the DPF-produced neutron pulse was sufficient to allow resolution of resonance absorption peaks in the sample under test for various ranges of resonance energies.
- Design software tools to perform the analysis of the performance of the DPF-driven NRS experiment, to allow for quantitative measures of experimental performance, and to aid in experimental design of potential future experimental series.
- Determine improvements, if any, in the experimental configuration that substituting a DPF for LANSCE would enable.

Important Experimental Factors

The experimental factors that affect the accuracy to which one can measure the width of a resonance include the rest-state width itself and the quality of the epithermal beam when it arrives at the detector (this quality is a function of the magnitude of the output neutron source pulse, the pulse width of the source, the effect of the moderator on the beam quality, the distance between the moderator and the element under test, collimation, and the flight-path length). The data are also affected by artifacts in the detector system and by detector solid angle. Changing the setup to optimize one of these parameters may affect several others.

Simulated Epithermal Flux on Target

The most important question to answer was whether NRS would be possible with the 1-MJ “TallBoy” DPF, loaded with a deuterium-tritium (D-T) mixture. Based on comparisons used by Yuan et al. (2005), we determined it was plausible to do NRS with the 1-MJ D-T DPF, as well as with a fast-pulsed assembly (FPA) driven by the same. Due to the complexity of the problem, this was not obvious from a cursory inspection of the setup. Based on a comparison of the LANSCE proton storage ring driving a neutron spallation target close-coupled with a moderator, the relevant experimental variables were the relative neutron flux at epithermal energies, the solid angle subtended by the detector system as viewed by the shocked target, the detector efficiency, and the moderator brightness (Figure 3).

Figure 3. Simulated neutron time of flight (NTOF) signal at 1 m from moderator

The simplest way to compare the epithermal flux from the LANSCE proton storage ring to the moderated pulse from the 1-MJ DT DPF was to model the mock-up experiment using Monte Carlo N-Particle extended transport code (MCNPx). This work was carried out at the University of Nevada, Las Vegas (UNLV), Nevada Radiological Computational Center (NRCC) for a number of different configurations, including deuterium-deuterium (D-D) DPF pulse, D-T DPF pulse, and DPF-FPA pulse, and several different moderator/reflector constructions. The simulations were designed to determine how many of the original neutrons from the DPF source pulse left the moderator from 19 to 23 eV. This number would give a good estimate of the number of energies at which the resonance absorption line would be sampled, which would determine if the resonance absorption line could be resolved. Some of the results are shown in Figure 4. Integrating under the curves in Figure 4 from 19 to 23 eV would give the number of source neutrons that were moderated to the resonance absorption line energy and, therefore, the number of sample points for the resonance absorption line. For LANSCE, 1.6×10^7 neutrons are generated in the 19 to 23 eV range for every source proton. For the 14 MeV DPF, 2.5×10^9 neutrons are generated in the 19 to 23 eV range for every source neutron. With a D-T DPF yield of 10^{14} 14.1 MeV neutrons, and a detector array located 5.75 m away from the target, there would be an estimated 180 counts in the 19 to 23 eV region (purple highlights). The same code predicts that the LANSCE NRS experimental setup would put about 1000 neutrons in the same energy range (conservative estimate). In short, *the simulation indicated the possibility of making this measurement.*

Figure 4. Monte Carlo simulation of moderated neutron spectra

System Energy Resolution and Sources of Error

Source-Moderator Pulse Broadening

The simplest way to think of moderator broadening is to think of it as a signal-processing problem. There are two contributing factors to this source of broadening, one of which is the width of the source neutron pulse, while the other is the relative degree of moderation (the more moderation performed, the longer the resulting neutron pulse will be). The only factor that affects the resolution is the width of the source neutron pulse. Given the same moderator, the degree of moderation only affects how many neutrons end up in the epithermal range. If the source neutron pulse is infinitely fast, you have the delta function response for the moderator. For any other distribution of source pulse, the resultant moderated neutron pulse would be the convolution in time of the source neutron pulse and the delta function response of the moderator. The resulting neutron resonance absorption spectrum would be broadened only by the natural line shape and the temperature of the material, assuming a perfect detector system. Given the above, significant modeling effort was expended using UNLV's NRCC in finding the most efficient moderator for NRS experiments, and the most efficient moderator for NRS experiments found was beryllium hydride (BeH_2), with normal polyethylene (CH_2) following closely behind (Figure 5).

Figure 5. Performance of various moderators for NRS

Geometric Broadening

Geometric broadening comes from two sources, one due to detector-target solid angle, and the other due to the difference in energies of neutrons resonantly absorbed as seen from different angles

from the direction of the epithermal neutron beam. The component of geometric broadening due to detector-target solid angle is significant only when the detector is located close to the target (which is almost never possible in actual NRS experiments). The primary component of geometric broadening is caused by the interaction of the epithermal neutrons with the shock in the material. Relative to the shocked material, the neutrons passing through the material are found to have a kinetic energy that depends on the angle from the normal of the shock front, as shown in Equation 1. The relativistic form of the kinetic energy is listed, although for low-energy resonance absorption the correction is small (Taylor, 1992). It can be shown that a series approximation yields the non-relativistic form of the equation as a special case.

$$\text{KE}_{\text{neutron}} = m_{\text{neutron}} \left(\frac{1}{\sqrt{1 - \frac{(\cos(\theta) \beta_{\text{neutron}} + \beta_{\text{shock}})^2}{(\cos(\theta) \beta_{\text{neutron}} \beta_{\text{shock}} + 1)^2}}} - 1 \right) \quad (1)$$

$$\text{KE}_{\text{neutron}} \approx \frac{1}{2} m (\beta_n + \cos(\theta) \beta_s)^2$$

This kinetic energy can then be used to determine the shape of the received absorption lines in the detector system. Neutrons arriving at the detector system, after having passed through the shock front at different angles, broaden the absorption lines. For higher energy lines, this effect will be more pronounced. For the sake of illustration, consider a detector array that subtends a 45-degree cone at the target, or a solid angle of 1.8403 steradians (which would be either an extremely large detector array or an array that is very close to the target). The broadening due to the arrival angle and energy of the neutrons would look like Figure 6.

Figure 6. Broadening due to arrival angle at detector array

To determine the net effect on the line shape, an integration over the solid angle subtended by the detector system would have to be performed. Specialized software was written during this project to perform the correct analysis.

Minimum and Maximum Detectable Resonance Energies

There are many factors that come into play when trying to determine if your experimental setup will allow the observation of an absorption line in an experiment. A simple way to break it up is to study what factors affect the observability of high- and low-energy resonances. This is instructive, but in reality, these factors cannot be considered separately. The modeling of our experiment consisted of a Monte Carlo calculation of the flux from the source through the moderator and target assembly. Once we had the neutron flux originating at the target, we had to obtain line shapes for the resonances in our target from the published literature (ENSDF, 2008). Once we had the absorption lines, we had to perform broadening on the absorption lines, based on the assumed temperature of the target and the various other sources of broadening discussed in the previous section, in addition to other more esoteric sources of broadening listed in the references (Stone, 2005).

Minimum Detectable Resonance Energy

The factors affecting *minimum* detectable resonance energy also affect the *maximum* detector distance. There is a minimum absorption energy width that can be detected with a particular DPF system configuration. In order to determine the temperature of the shocked material from the low-energy resonance absorption line, you must be able to accurately measure the width of the resonance. As noted in the sections above, broadening of the low-energy resonance absorptions would probably not be an issue, but the noise floor will be. The problem with the low-energy absorption lines is that even with large degrees of moderation relatively few neutrons end up in the energy range of interest. The minimum resonance absorption energy becomes a question of what signal-to-noise ratio is necessary to measure an absorption line over the noise floor. An example of simulated low-energy resonance is shown in Figure 7.

Figure 7. Simulated time signal for low-energy resonance

Figure 8 shows a simulated time signal that would be recorded on an NRS experiment. The plot on the left is a detail of the resonance region where the resonance lies. This illustration shows how low-energy resonances can be lost in the noise. In both plots, the blue curve has a higher degree of moderation than the magenta curve. The purpose of the increased degree of moderation would be to put more neutrons into the resonance window.

Figure 8. Simulated absorbance of neutron signal in the energy domain

Maximum Detectable Resonance Energy

The factors affecting *maximum* detectable resonance energy also affect the *minimum* detector distance. The maximum energy that a system can record is also limited by the detector array; however, the limiting factor is almost certainly not the noise floor. The neutron flux for the high-energy portion of the moderated pulse (i.e., ≥ 100 eV) is significantly higher than the background, but is still affected by fluorescence in the scintillator and by bremsstrahlung in the collimating shield. Shown in Figure 9 is the simulated relative signal absorption for a 100-eV resonance with different separations of a circular emitter, 1 meter in diameter, and a circular absorber, 1 meter in diameter, located at several different normal separations.

As is apparent in Figure 8, the closer the detector is to the emitter, the larger the width (in energy) of the absorbed line. Shown in Figure 9, we see the same absorbance signals in the time domain, and begin to understand the real limiting factor for measuring the width of relatively high-energy resonance absorption lines.

Figure 9. Time domain neutron absorbance at 100 eV

However, the resonance absorption line width (in time) is so short that the detector and the recording system become the limiting factors. If you make simple assumptions about how many time samples you would need to determine the presence of a resonance absorption line for any given resonance energy and flight path, you could estimate the upper bound for the resonance absorption lines that you could expect to see. If you assume that you require at least four points to estimate the FWHM of the resonance absorption line, irrespective of the actual functional form of the absorption line, you would see that 100 eV is near the practical limits for NRS using DPF, as shown in Figure 10.

The best way to interpret the data in Figure 10 is to read the x-intercept. If the energy of the resonance absorption line is higher than that of the x-intercept, the digitizer would not be able to record it due to the sharpness of the line. For instance, measuring an absorption line with a 0.1 eV FWHM you could expect to resolve absorption lines up to about 140 eV. Given a detector at 1 m, measuring an absorption line with a 0.2 eV FWHM, you would expect to see resonances up to about 230 eV. Since the 0.2-eV FWHM case is wider in energy than the 0.1-eV FWHM case, the line would also be wider in arrival times at the detector system; so, it makes sense that you would be able to resolve “fatter” lines higher up in absorption energies.

In actual practice, you would want to minimize the effect the scintillator fluorescence and shield bremsstrahlung have on the time-of-flight signal; this would require experimental tuning to determine the proper distance for the resonance of interest. Your moderator would limit the highest observable neutron resonance energy. If the resonance energy were higher in energy than the peak in

the moderated neutron spectrum, it would be unobservable due to the rapid fall-off of the moderated neutron energy spectrum as energy increased above the peak (Figure 4). To measure higher neutron resonance energies, you would need to redesign the moderator-reflector assembly, yet you would then run into the problems discussed above.

Figure 10. NRS-required detector system resolution

Conclusion

Based on comparisons to previous NRS experiments and MCNPx simulations of the experimental setup for the 1-MJ D-T DPF-NRS experiment, we concluded that NRS using DPF as a driver is possible using the simulated configurations. The experiment should deliver enough neutrons in the resonance energy range given losses from solid angle and detector efficiency to properly determine the shape of the line and, therefore, the temperature of the shocked sample. There are several aspects of the experiment that could also be improved should additional effort be made, such as improving the efficiency of the reflector, moderator, and collimator assembly. It may be possible to integrate some of these components with a fast-pulsed assembly for an even larger range of possible observable neutron resonances.

The research conducted this year indicates that it might even be possible to perform NRS experiments with D-T DPF operating at 10^{14} neutrons per shot and, with higher yields from a DPF-FPA, a broader range of neutron resonance energies could be seen.

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COMPUTER SCIENCES

WAVELET OPTIMIZATION FOR DETECTION AND SPECTRAL IDENTIFICATION

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Wavelet analysis, an emerging, powerful analytical tool that is an inherently multiscale technique, was applied to radiation detector time-series data and also to associated spectral data in order to improve anomaly detection performance. The motivating framework for this study was to focus on operational implementations for these novel analytical techniques. Based on this study, the wavelet analysis technique appears to be a fast and reliable filtering technique for these data types, provided the targeted features are roughly characterized. Some empirical limitations are also detailed. As a first filter for spectral data, wavelets can greatly concentrate the actionable information in a spectrum to be used by an analyst or an intelligent identification algorithm. The selection advantages of wavelets as an anomaly technique can be made even more powerful when coupled to an adaptive learning algorithm.

Background

Wavelets are a successful and highly implemented signal processing tool in such diverse applications as stock market futures (Turiel, 2003), medical diagnostics (Ivanov, 1996; Ivanov, 1999), astrophysics (Argüeso, 2006; Sanz, 1999), psychology (Ashenfelter, 2009), and even gamma spectroscopy (Sullivan, 2006). The nearly universal applicability of wavelets comes from its ability to both detect and analyze *changes* in any data, giving strong clues to the underlying cause of those changes.

The goal was to apply these emerging wavelet analysis tools to several vital deficiencies in current operational radiation search and identification approaches. It has been previously noted that improvements to the detection algorithms have significantly trailed behind the technological improvements in the detector hardware for national security applications (Vojtech, 2008). Thus, the most pronounced impacts in detection applications can be made through algorithm enhancements. Implementation of algorithm improvements can be done with little investment in hardware, can be done in parallel with existing algorithms, and can provide dramatic improvements in signal detection.

Wavelet analyses were derived from Fourier analysis; therefore, they operate very similarly. Unlike the more common Fourier transform analyses, which are ideal at quantifying signals encompassing the entire data set, wavelet analysis adjusts the scale/resolution of a single-wave oscillation signal by stretching it to different scales. Wavelets, which have a strong track record in denoising, can also be applied to noise filtering of this time series or spectral data. *The Illustrated Wavelet Transform Handbook* by Paul Addison (2002) provides an excellent empirical overview of wavelet analysis approach and provides some specific examples.

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Project

This study demonstrated the efficacy of wavelets in both enhancing signal detection and minimizing background, focusing on the problem that most operational data are generally dominated by noise from one data point to the next. The true signature involves a persistent trend across multiple points. A local feature will increase and then decrease through the data. Because the pervasive noise throughout this signature poses a challenge for detection, legacy methods have relied on either high signal-to-noise ratios to mitigate the noise-induced false alarm rate or laborious manual examination.

Figure 1. Comparison of a reverse-chirp signal and a NaI(Tl) spectrum. The raw chirp (a) shows the decrease in frequency (increase in scale) as the channels increase. The same trend occurs in gamma spectra, such as the one shown in (b) for Rn²²². The corresponding wavelet scale diagram is shown for the chirp (c), and the spectrum (d). The same trend toward higher scales (black arrows) is exhibited by both data.

This project applied wavelets in spectral analysis, with an emphasis in optimized peak detection and quantification in low-resolution/low-statistics data. Wavelet analysis also was applied to photopeak detection in sodium iodide doped with thallium (NaI(Tl)), high-purity germanium (HPGe), and lanthanum bromide (LaBr₃). One of the key issues in gamma spectroscopy is the change of peak resolution as the energy increases. An optimized wavelet scale at lower energies is not effective at characterizing peaks at higher energies. In effect, gamma spectra exhibit reverse chirp signatures. Wavelet analysis is certainly effective at capturing the features of a chirp signal, and it would be operationally straightforward to apply that method toward gamma spectra. Figure 1 shows the wavelet features compared to a reverse chirp. The similarities illustrate the effectiveness of implementing the multiscale analysis capability in wavelet analysis. Figure 1(d) also illustrates some of the advantageous features of wavelets. The location of a centroid of a photopeak across a large range of scales is the same. The value of the wavelet coefficient, which translates into peak area, varies slowly in a moderate range of scales.

Anomaly Detection in Time-series Data

Wavelet analysis was applied to time-series data from aerial systems. Others have successfully applied wavelet analysis on mobile detector systems (Campbell, 2007) to show that desired features can be effectively keyed into the algorithm based on scale. An example of a mobile feature would be a stationary point source being detected from a vehicle traveling at 30 mph. This signal would need to be elevated over nuisance alarms caused by changes in the background. Multiple wavelet filters, which have been previously characterized to match either point source, extended source, or change in background, have been applied to improve the detection levels over the standard alarm thresholds in post-processing. The results for aerial systems show that wavelet distance scales improve the lower limit of signal-to-noise detection without significantly reducing the false-alarm rate. These techniques provide a way to improve the system sensitivity for moving detection systems in aerial, vehicular, or pedestrian platforms.

Wavelet Analysis of HPGe Spectra

Spectral-peak detection and quantification of HPGe are generally straightforward due to HPGe's high resolution. Wavelets would require significant tuning to differentiate low-statistic photopeaks from noise because the scales of both are only a few channels. Nevertheless, some areas, especially in the mid-600-keV range, have overlapping plutonium isotopic peaks that must be differentiated effectively. Wavelet approaches may even provide an improved capability in the analysis without having to improve detector system performance. Also, wavelets can be used to detect peaks below the threshold of visual detection (at the expense of false-alarm rates), thereby providing more actionable information. Without time-consuming screening, these peaks are often missed because the statistical noise effectively masks their detection. Wavelet analysis of HPGe can be beneficial, particularly when coupled to a strong and reliable isotopic search routine.

In Figure 2, the blue line shows the second derivative of the counts per channel, which is generally attractive for enhancing local features. In this case, the channel-to-channel statistical fluctuations dominate the blue line, with the exception of the large peak around channel 1800. Normal smoothing would diminish the source peaks significantly. The red line shows Mexican hat wavelet analysis of a scale width of three channels. These effectively smooth over the statistical noise peaks, while amplifying the source peaks.

Figure 2. Comparison of wavelet analysis to the second derivative of a ^{137}Cs HPGe spectrum. The plot on the left illustrates the denoising potential of wavelets; the blue line shows the second derivative of the spectrum. A small, noise-dominated peak is shown at top right, while a large 661-keV peak is shown at bottom right. The signal of each peak was amplified (lower right), while the small-scale noise peaks were smoothed.

Figure 3 shows the success of the automated peak detection system for HPGe. It correctly identified and marked the large and essential peaks needed for proper isotopic identification (shown in the

embedded table). A thorough visual scan revealed that it missed five peaks, identified nine of the visually detected peaks, and detected four previously missed peaks. The missing peaks were likely a result of the high threshold set in the algorithm described below.

Figure 3. Automated wavelet analysis of unknown HPGe spectrum. It was later determined to be ^{252}Cf , shielded ^{226}Ra , ^{40}K , Fe capture, and H capture. The blue line represents the spectrum, and the diamonds are the peaks and strengths (shown in the embedded table) determined by wavelet analysis.

After algorithm test and evaluation, the most reliable and simplest (i.e., closest to detecting the same peaks as a visual search) approach was to select the wavelet scale based on a 4-channel width. The raw data were rescaled by the local variance through a floating window of 100 channels. This approach was selected over a larger variance window, square root of local mean, no rescaling, and local mean. Based on the local variance, wavelet peaks were selected if they had a wavelet peak ratio greater than $4x$ the local variance. It should be noted that wavelet peaks amplify the photopeak signal above the peak maxima, as shown in Figure 2. A local window data normalization is greatly preferred over a

large or full window, because two orders of magnitude change in statistics occurs from the low to high energy as is shown in Figure 3. This local window normalization allows the use of simple statistical detection threshold across the full energy range.

Wavelet Analysis of LaBr_3 Spectra

One of the goals of this work was to apply wavelet spectral analysis to LaBr_3 spectra. The self-activity of LaBr_3 is well-characterized, but is proportional to the integration time. The self-activity currently limits the effective use of this otherwise ideal crystal for scintillation. Various wavelets were analyzed on a collection of nine isotopes and six wavelet types (Figure 4). The study included a background characterization that effectively corrected the self-activity. The results are shown in Figure 4.

Figure 4. Performance study of wavelets on LaBr_3 spectra: (a) Statistical results of multiple wavelet applications on LaBr_3 spectra; the Mexican hat wavelet had the best performance, while the Morlet wavelet was the least effective. Also shown are comparisons of wavelet-transformed spectra ([b] Mexican hat, [c] Morlet) with LaBr_3 raw spectra.

Because the Morlet wavelet is designed for multippeak features such as those arising from diffraction in optics, it was a poor choice for detecting a single photopeak. The Mexican hat wavelet, which is the negative of a Gaussian second derivative, captured the general shape governing large-statistics photopeaks. It should be noted that the choice of wavelet type is not crucial to the success of the wavelet method, because the weighting of the filter is not as dependent on the chosen weighting function as it is on the width of the filter. This is analogous to traditional smoothing filters in many respects. Wavelets do indeed have an effect on the optimization (second-order effects) of the wavelet filtering techniques.

Wavelet Analysis of Sodium Iodide Spectra (NaI(Tl))

An automated, statistical wavelet study of an extensive collection of $3'' \times 3''$ NaI(Tl) spectra was performed in order to determine the performance of the custom-designed wavelet algorithms. It should be noted that the quality of NaI(Tl) varies significantly depending on crystals, electronics, and environment. There were 43 spectra of different single isotopes, although many of these contained decay products mostly in equilibrium, as well as several background lines. Our goal here was to develop and test a simple but automated software tool that could pull a spectrum in and return the location and area of each peak accurately. A plot of the location and strength were overlaid onto the gross data as a qualitative tool. Thus, there was a qualitative and quantitative method to validate the accuracy of the automated method.

Figure 5 shows a cross section of these plotted results. Subplots (a) and (c) were picked to show some of the successes of the wavelet algorithm, while (b) and (d) illustrate several limitations.

Overall, the post-processed validation of the data showed that there were 495 out of 547 correctly detected peaks, 52 missed peaks, and 22 false peaks. The 547 total peaks, which included visually blended peaks, were determined visually from representative blue spectra listed in Figure 4. The Type II error (false negative) was 10%, and the Type I error rate (false positive) was 0.7% (based on the algorithm discussed below). The primary cause of the false negatives was proximity of a smaller peak to a larger one. In those cases, the offset decay from the dominant peak's wavelet coefficient overshadowed and caused interference with the secondary value. One way to resolve this primary failure mode would be to apply smaller scale wavelets near the large wavelet (would reduce the range of effect from the dominant peak), filter out the dominant peak, and reanalyze; another would be to determine the multiscale features of these blended peaks in the adaptive learner. While the more sophisticated solution would be the latter, the first approach would be easier to implement. The first-order algorithm performed accurately. A striking feature of the analysis software was the computation time, which was 28 seconds total (12 seconds without a graphical plot) to analyze and output the 43 results on a desktop computer.

Figure 5. Wavelet peak detection of four isotopes in the NaI(Tl) sample. The blue lines show the log spectra, and the red diamonds give the locations and count area of the wavelet peak detection results: (a) ^{239}Pu shows a successful detection of a low signal (left green area) and three blended peaks (right green area); (b) ^{111}In represents the poorest performing result (out of 43 tested) with four missed peak detections (yellow area); (c) uranium ore represents the best performance, with 19 out of 20 peaks correctly located; (d) ^{252}Cf illustrates another poorly performing result with three detectable peaks missed (yellow area). The spectra were all offset by 100 to avoid overlap with the wavelet results.

Categorizing Peak Types

The next issue to tackle was the correct categorization of the various peak types. Several types of peaks were collected, including single photopeaks, Compton edges, backscatter peaks, blended peaks, and annihilation peaks. While there were inherent variances within each category in terms of multiscale behavior, the overall trends were significant enough to consistently separate them, as demonstrated in

Figure 6. The multiscale behaviors of each peak type are very likely to be distinguishable with very few parameters in an adaptive learning or parameterization algorithm designed to reduce computational burden.

Figure 6. Multiscale behavior for a Compton peak versus a photopeak. The photopeak (top) exhibits stationarity (horizontal line) and reflection symmetry about the maxima. The Compton edge (bottom) shows nonstationarity (curved line) and asymmetry. The differences in the behavior, particularly the change in location of a peak from one scale to the next, provide a way to discriminate between different peaks. The same approach can be applied to blended peaks in an adaptive learner.

The method for the determination of the best filtering for NaI(Tl) was done much like that for HPGe. The primary difference was the use of multiple scales to best capture the single photopeak features at low and high energy. In all, a channel scale of 4, 8, 16, 24, and 32 (the rough energy equivalent was 1.5 keV per channel) was used in a piece-wise continuous fashion, with a channel cutoff between each in the peak selection process. Of these choices, the range cutoff was determined by measuring

the multiscale behavior of a subset of photopeaks in that range to determine the appropriate categorization. This simple solution was the first step in addressing the change in resolution exhibited by NaI(Tl) data.

A major issue with all spectral data is the change in statistical characteristics across the energy scale. A fixed-count threshold is a poor choice because the size of a nuisance peak at high-count areas around 100 keV would be much larger than a photopeak at high energies. To account for this statistical variation, our data were simply rescaled by the square root of the local mean, defined to be the same range as the wavelet scales. The choice of this rescaling method had little effect on the outcome as long as it accounted for the change in background. A final modification was to look for the maximum peak in a confined local neighbor of the same scale as the wavelet scale. This was done to mitigate the infrequent occurrences of multiple peaks solutions (only a few channel separations) for the same photopeak. The cause was statistical fluctuations in low-statistics peaks.

Wavelet approach was also applied to other forms of NaI(Tl) from aerial spectral data, identiFINDER, and mobile data; each system varied widely in spectral resolution. The overall benefit of wavelet analysis for each followed the aforementioned pattern for the 3" $\times$ 3" NaI(Tl). To a certain extent, wavelet analysis does not depend on energy resolution as much as it does on knowing the expected peak shape response. With that information, wavelets can easily select the photopeaks through noise, a sloping background, or a curving background, thereby concentrating the critical information.

Conclusion

Wavelets provide a powerful tool to filter and analyze time series, spectral, and even two-dimensional data like imaging. The necessary requirement to its straightforward implementation is an understanding of how that feature behaves in the complexity of operational environments.

Operationally, wavelet spectroscopy works much like windowing methods, but the wavelets scan the whole spectrum with an optimized window (filter) for a specific feature like a photopeak, noise peak, Compton edge, brehmsstrahlung, or blended peaks. The output of this first pass filter (peak location and area) could become the input for a peak search library program. A logical extension of this wavelet filtering and peak selection work would be an adaptive learning isotopic source algorithm. Limiting the adaptive learning inputs to only the relevant information would be the best way to improve processing speed, reduce Type I/II errors, and avoid common adaptive learning shortfalls of having too much superfluous information. Therefore, this project anticipates developing a wavelet adaptive learning, automated spectral analysis package, which will emphasize speed and reliability. Following versions could likely leverage the inherent multiresolution analysis of wavelets to detect features only highly trained spectroscopists can currently perform. This could be realized, provided the spectroscopists are the input key of the adaptive learning tool. Even without the isotopic identification, the ability to compress a spectrum down to the actionable information would provide the spectroscopist with a tool to more quickly determine the captured sources.

One way to improve the performance of the automated algorithms would be to level the playing field between the statistical environments. Local variance was determined to be the best method to rescale the data for HPGe since its low-signal peaks closely resemble noise peaks. The best method for NaI(Tl) currently is to rescale by the local mean, since the peaks are spread wide.

The performance of wavelet analysis in peak detection was surprisingly effective at reducing false alarms, as demonstrated by the results of this study. Both the higher resolution LaBr₃ study and the low-resolution NaI(Tl) study had similar performance for the Mexican hat wavelet in terms of its ability to rapidly and correctly detect peaks with an extremely small false-positive rate. The similarity suggests that wavelet analysis balances out the energy resolution for peak detection. Improved resolution provides benefit mainly in the statistics of near-threshold detections and a lowered probability of blended peaks. The former can be rectified by improved detector efficiencies in lower resolution systems, and the latter can already be rectified as outlined in the sections on HPGe and NaI(Tl) in this report. As such, this method is ideally suited for multiple detector systems, hybrid systems, and high-efficiency spectral systems. As wavelet research continues and more intelligent methods are discovered, these algorithms will evolve. The beauty of algorithm research is that it endures far longer than hardware advances, improves a whole suite of technology at once, and vastly expands the understanding of the physical processes through the eyes of the data.

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DETECTORS AND SENSORS

HIGH BAND-PASS PHOTOMULTIPLIER TUBE AND VACUUM PHOTODIODE DEVELOPMENT (PARTS I AND II)

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This report is presented in two parts. The original project would have developed a high band-pass photomultiplier tube (PMT) specifically for VISAR (velocity interferometer system for any reflector) measurements. However, the inability (or lack of interest) of several phototube companies to provide crucial assemblies and processing services made it impossible to proceed with the original design. Consequently, the PMT design was converted to a configuration that was compatible with National Security Technologies', LLC (NSTec's), Livermore Operations (LO) phototube assembly and processing expertise. Unfortunately, the design changes have most likely compromised the PMT's proposed usage for VISAR experiments.

Another proposal to develop a unique, high band-pass vacuum photodiode (PD) was independently submitted for SDRD funding. Because the PMT and the PD projects were technically quite similar, development funding for both phototubes was allowed on the same project. The PD project (development of the PD42008) is presented as Part II of this report.

Background

The measurement of radiation bursts of short duration has become more crucial to correct physics diagnostics, particularly as the duration has moved into the subnanosecond realm or when an accurate measurement of structure innate to the radiation burst is crucial. Both the PMT2008 and the PD42008 projects were conceived to address these basic measurement challenges. If the predicted temporal response of these devices were valid, then there would be, for example, immediate applications for these devices associated with VISAR and inertial confinement experiments.

Part I: PMT-2008 Development

Project

PMT Design Changes

The most significant change to the original dynode array design replaced the radial bank of cylindrical mesh dynodes with a 15-stage planar mesh dynode array. The two dynode array designs are depicted in Figure 1.

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Figure 1. PMT2008 dynode arrays: (left) the redesigned array is shown with the (right) original array

Figure 2. Overall design of the final PMT2008 design. The stainless-steel housing has been made (artificially) translucent to reveal the phototube's internal components.

The dynode array change also included moving the dynode electrical leads (via a multi-conductor vacuum feed-through) from the face of the phototube to the wall of the phototube's cylindrical metal housing. In addition, the radial electron focus was replaced with a less-efficient cylindrical deflector. Figure 2 shows the internal features of the modified phototube's design.

Dynode Assembly

Each planar disk dynode is made from an ultrafine nickel mesh bonded (with nickel braze) to a 0.007-in.-thick stainless-steel frame. The “working” area of the dynode is within the steel frame's 0.750-in. diameter. The steel frame has a 0.830-in. outer diameter with a 0.313-in.-wide “panhandle” that extends 0.813 in. from the frame's center. The panhandle firmly holds each dynode of a 15-piece assembly in a cantilevered and parallel position to adjacent dynodes. The spacing between adjacent dynodes is ~0.093 in. A thin film of magnesium oxide is deposited on the fine nickel mesh to enhance secondary electron emission capabilities.

Anode

A slight focus of the phototube's electrons (provided by a biased cylindrical deflector) from 0.750 to 0.625 in. assures that all of the charge from the 15 dynodes is collected by the anode. An electron focus also minimizes the anode's required diameter and thereby minimizes the lump capacitance between it and the last dynode. A 50- Ω characteristic impedance from the anode's “collecting” surface to the signal tap-off end of the vacuum feed-through optimizes signal transmission qualities.

Photocathode

The photocathode substrate is suspended (and electrically isolated) from the phototube's front plate. This configuration allows photocathode activation by remote processing techniques. The photocathode's planar surface is prepared with an S20 (or equivalent) spectral sensitivity followed by the subassembly's placement into the phototube's vacuum chamber that contains the properly assembled dynode configuration. A permanent indium vacuum seal is made between the subassembly's flange and PMT housing. In its final position, the photocathode is electrically connected via the dynode subassembly to pin no. 2 of the phototube's multiconductor vacuum feed-through.

Operational Theory

The PMT2008's performance is based on (1) an efficient electron photoemission from an optically semitransparent cathode, (2) the amplification of that small photocathode charge by electron secondary emission from ultrafine dynode meshes, and (3) the collection (and transmission) of the amplified charge by an anode and vacuum feed-through configuration designed for signal high bandwidth.

Although the crucial components of the phototube are designed for signal high bandwidth, there are limitations associated with the secondary emission process of the mesh dynodes. The efficiency of individual electron interactions with the fine “wires” of the dynode mesh determines the phototube’s signal gain in addition to the signal’s rise and fall times.

The signal rise and fall times depend on the probability that electrons emitted from any dynode will pass through one or more of the downstream mesh dynodes with little or no interaction. These variable interactions allow a portion of the amplified charge to arrive at the anode’s collection surface before the main charge deposition. A small fraction of the amplified charge will also arrive at the anode later than the main signal because of very efficient interactions with the dynodes (compared to the average interaction efficiency). From a graphical standpoint, early arrival is manifest by a premature rise of the signal from zero base line. Late arrival will contribute to a slow fall time toward zero base line. Both phenomena compromise the ideal frequency band-pass of the phototube.

Ultrafine metal mesh is used to minimize electron skipping between dynodes. However, there is a trade-off between secondary emission efficiency and skipping reduction. If the mesh is too fine, then its secondary electrons may not escape to the downstream dynodes; electron amplification (gain) is compromised. If the mesh is too coarse, the interaction between the electrons and the mesh will be severely jeopardized; dynode skipping becomes a major problem and electron amplification (gain) decreases. The best configuration would cause the early and the late charge to be equal, thereby providing a Gaussian distribution. Variations in the accelerating voltage between dynodes should help establish this condition.

The optimum bias to the dynodes was theoretically determined with computer software, wherein calculated electron paths were mapped following discrete changes in the applied voltage to individual dynodes. Figure 3 shows electron trajectories with an optimum bias applied to each dynode. The axis of rotation for the cylindrical geometry of the PMT2008 in this figure is the bottom horizontal line (the “Z” axis).

Theoretical predictions for the PMT2008 phototube were made using a simplified model in which the rise time of the signal was solely dependent upon the flight time of the electrons from the last dynode to the anode, a distance of ~0.093 in. Also, the fall time (for this simulation) depended only on the capacitance between those same components (~1.1 pF). The graphic display in Figure 4 shows the resulting signal that exhibits

- a 113-ps impulse rise time (10 to 90%),
- a 122-ps impulse fall time (10 to 90%),
- a 134-ps impulse FWHM, a 185-ps step rise time (10 to 90%),
- and a 1.83-MHz frequency band-pass (at -3 dB).

Figure 3. The predicted electron paths for (theoretically) an optimum bias to the PMT dynodes. For this setup the deflector is biased to -4000 V, the photocathode is biased to -3700 V (left), and the conical anode is at electrical ground (far right).

Figure 4. Simulated temporal signal response of the PMT2008 based on a simplified model

Conclusion

Although the design was completed, we were unable to evaluate the prototype. The parts for assembly and processing were available in early July, but the required assembly and processing were not accomplished. Consequently, no evaluations or absolute conclusions were possible relating to the performance of the modified PMT2008 design.

Part II: PD42008 Development

Vacuum biplanar photodiodes (PDs) have played a major role in the dynamic diagnostics of intense radiation transients. A PD's main advantages over solid-state devices lie (1) in their ability to deliver large linear signals, and (2) in their radiation sensitivities not easily duplicated by other sensors with comparable signal output capabilities. This development work focused on the reasonable possibility that vacuum PDs, designed to meet specific experimental needs, could far exceed their present temporal response capabilities without compromising signal linearity. Preliminary mathematical simulations predict that a vacuum phototube comparable to the proposed PD42008 design will exhibit a signal bandwidth in the 1.9-GHz range.

Background

The measurement of radiation bursts of short duration has become more crucial to correct physics diagnostics, particularly as the duration has moved into the subnanosecond realm or when an accurate measurement of structure innate to the radiation burst is crucial. Both the PMT2008 and the PD42008 projects were conceived to address these basic measurement challenges. If the predicted temporal response of these devices were valid, then there would be, for example, immediate applications for these devices associated with VISAR and inertial confinement experiments.

Project

The PD42008 Concept

The PD42008 vacuum phototube design is based on geometric advantages successfully demonstrated by the VCD-173 vacuum Compton detector (VCD) and an experimental Faraday cup (Davis, 2008). The PD42008 uses nine concentric photocathode rings spaced within the cavity of a conical anode. The photocathode structure is positioned such that the longitudinal location from where photoelectrons are ejected (in time) closely matches the longitudinal position of the associated electrical signal as it propagates on the outside surface of the anode cone.

The geometry allows the charge (induced by transient and incident radiation) from the nine photocathodes to "collect" at the anode surface such that the charge is continuously bunched into a narrow time-packet as the resulting "main" signal propagates toward the apex of the conical anode. The PD42008 geometry also assures that signal distortion is minimized by maintaining a continuous, 50- Ω characteristic impedance between the anode and its outer shield.

In effect, the PD42008 geometry concentrates all of the charge emission from a relatively large photocathode surface without time smear associated with (physically) large photocathodes. Figure 1 is a

concept drawing of the PD42008 detector that includes the PD42008 phototube and its holder (the detector assembly). Figure 2 is a three-dimensional model of the PD42008 phototube without the holder.

Figure 1. The PD42008 photodiode detector with holder. Two signals are induced during normal PD operation; the “main” signal propagates to the right, and a secondary signal moves to the left. The main signal will represent the incident radiation because of charge bunching. The secondary signal will be stretched in time since it propagates in the opposite direction of the incident radiation. The secondary signal is “absorbed” by the 50-Ω coaxial terminator.

Figure 2. Nine-stage PD42008 phototube. The stainless-steel body has been made (artificially) translucent to reveal the internal components.

Tiny grooves machined into the oblique surfaces of the photocathode rings allow photons with velocities parallel to the photocathode's axis to strike a perpendicular surface and thereby optimize their adsorption at those S20 photo-surfaces.

The internal surfaces of the conical anode "collect" the electrons from the photocathodes. These internal surfaces are designed to minimize time smear by collecting electrons from any photocathode ring with about the same electron trajectory. This condition was confirmed by plotting the calculated electron paths using the proposed geometry; the results are shown in Figure 3.

Figure 3. The PD42008 photodiode, showing electron trajectories (black) between the multiple photocathodes and the internal surfaces of the conical anode. The axis of rotation of the PD is the horizontal line at the bottom of the figure ("Z" axis).

The outer surface of the anode is the center conductor for a $50\text{-}\Omega$ transmission line that is established between the anode and the inner surface of a concentric housing, an essential part of the PD42008 holder. The PD42008 detector housing also allows 8000 V to be applied to all nine photocathodes via a contact pin located at the center of the phototube's optical window.

The configuration of the holder for the PD42008 uses basic concepts and in-line termination components common to the VCD-173 prototype detector (Davis, 2008). This detector's coaxial terminator successfully terminates its secondary signal to yield a measured 4.48 GHz (at -3 dB) signal bandwidth. The PD holder was intentionally designed to use several parts common to the VCD-173 detector in an effort to minimize engineering, component, and fabrication costs.

The holder's in-line $50\text{-}\Omega$ terminator is required to absorb the secondary signal that propagates toward the PD's optical window. Without this coaxial, $50\text{-}\Omega$ termination, that signal would undergo a reflection near the optical window and (delayed in time) would follow in the same direction as the "main" signal. That condition would significantly compromise the signal band-pass capability of the PD.

Predicted PD Signal Performance

Signal performance prediction for the PD was based upon a very simple model. The model presumed that the signal rise time was dependent on the transit time of the charge across any of the cathode-to-anode vacuum gaps (76 ps from baseline break-away to peak at 8000-V bias) convolved with the “width” (the “length” of the electron burst emitted from each photocathode) of the individual bunching measured in time (~21 ps). The signal fall time presumed that the capacitance (1.65 pF) between the largest cathode and the anode was the major contributor to signal fall time. Figure 4 depicts a simulated signal (1.89-GHz band-pass) that is expected from the PD42008 based on the described simulation parameters.

Figure 4. Signal simulation based on a simplified mathematical model in which the signal rise time is dependent on the electron transit times between the cathode and anode, and the signal fall time is dependent on the capacitance between the largest cathode and the anode.

Conclusion

All of the preliminary development work (design and simulations) was completed, and the provision of a full complement of PD42008 components to assemble and process one prototype was provided in early July 2008. However, the required assembly and processing of the phototube was not accomplished before the end of September. Without a prototype device, actual performance evaluations were not possible. Despite this, we remain confident that the PD42008 phototube will perform as predicted by the simulations. We have learned that it may be possible that this prototype device could become available through non-SDRD funding some time in FY 2009.

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URANIUM VISUALIZATION CHEMISTRY

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During this second project year, methods for enhancing uranyl (UO_2^{2+}) phosphorescence using chelation by polyhedral oligomeric silsesquioxanes (POSS) were further investigated (Di Benedetto, 2008). Studies were performed in acetone, acetonitrile (ACN), and tetrahydrofuran (THF) with a variety of tri- and tetra-silanol POSS ligands, and, to emulate surface contamination, studies were performed in the presence and absence of iron on metal residue-coated filter papers. Fluorescence spectroscopy and time-resolved imagery data were used to identify new compounds and measure fluorescence lifetime. Spectral measurements suggest that two new POSS-U complexes were formed; however, because they were not isolated, it is difficult to gauge the effect that POSS has on POSS-U fluorescence. At alkaline pH in the absence of Fe(III), POSS appears to quench rather than enhance phosphorescence of the solution.

Background

This FY 2008 project was a continuation of an FY 2007 SDRD project with the same title. More complete background information can be found in that report. One of the characteristic features of UO_2^{2+} compounds (i.e., compounds containing the U(VI) species) is their phosphorescence; remote detection of this uranium-oxide phosphorescence has been useful for locating uranium at contaminated sites (Di Benedetto, 1999). Because the detection of the target emission is often diminished by incomplete oxidation, or matrix effects that either quench the emission or inhibit excitation of the material by the laser excitation source, the overall goal of this project was to overcome these limitations by developing a method, chemical formulation, and kit that could be used for remote delivery and detection of enhanced phosphorescence of uranium complexes on surfaces. The multicomponent formulation would (1) mechanically or chemically extract the uranium complex to the surface for improved visualization, (2) oxidize reduced uranium to U(VI), and (3) chelate U(VI) with a ligand that enhances its phosphorescence by means of a suitably absorbing chelating agent that separates it from iron.

Since weakly bound ligands can cause quenching due to rapid bond breaking of the solvent in the first coordination sphere (Formosinho, 2003), we considered phosphorescence enhancement in which bonding to the metal ions involved non-labile ligands with low numbers of protons. Bimolecular quenching can also be a major loss mechanism for phosphorescence, and, in both diffusion-controlled

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and static mechanisms, iron can contribute significantly to loss of signal. Due to the ubiquitous nature of iron in the environment, part of this work focused on the effects of iron in solution and on dried samples.

Uranyl complexes with virtually every type of oxygen donor, as well as many nitrogen and even sulfur donors, have been reported (Cotton, 1988). Silica gels (Sutton, 2003) and silico-chelates (Lorenz, 2000) demonstrate promising templates for long-lived emissions. Since chelation by these ligands might prevent rapid bond breaking, inhibit quenching by iron, and aid in oxidizing reduced U, we chose to investigate multidentate ligands based on silicon oxides. In our approach, tridentate and tetradentate (with two bidentate coordination sites) functionalized silanols, or silanol salts, from Hybrid Plastics were used. Figures 1a and 1b show the R-substituted trisilanol POSS (SO1450 for R = isobutyl)² and the sodium salt of a phenyl-substituted tetrasilanol POSS (SO1460),³ respectively. Figure 1c shows a possible six-coordinate uranyl complex coordinated by two SO1460s. Lorenz reported that a six-coordinate (POSS)₂-U(VI) species, characterized by x-ray crystallography, was formed with a tridentate silanol POSS (R = cyclohexyl) regardless of the original U oxidation state (2000). Fluorescence data were not collected, however.

Figure 1. (a) TriSilanolR-POSS (R = isobutyl: SO1450); (b) Tetrasodium salt of TetraSilanolPhenyl-POSS (SO1460Na); (c) possible coordination of uranyl by SO1460Na.

In FY 2007, uranyl acetate (UAc) was reacted with a variety of POSS ligands. The reaction of SO1460Na with UAc in EtOH gave a precipitate with blue-shifted emission under 355-nm excitation and longer-lifetime phosphorescence (dominant component: 87 μ s). Preliminary observations suggested that a POSS-U(VI) complex had formed. However, in FY 2007, the significant pH differences between U and POSS solutions were not addressed. Figure 2 demonstrates the significant effect of pH on uranyl speciation in water. We presume that uranyl speciation may be similarly responsive to pH in the organic solvents.

² 1,3,5,7,9,11,14-Heptaisobutyltricyclo[7.3.3.1(5,110)]heptasiloxane-*endo*-3,7,14-trisilanol. Product Name: TriSilanolisobutyl-POSS; Product Number: SO1450.

³ Tricyclo[7.3.3.3(3,7)]octasiloxane-5,11,14,17-tetrasodiumsilanolate-1,3,5,7,9,11,14,17-octaphenyl. Product Name: TetraSilanolPhenyl POSS; Product Number: SO1460Na.

Figure 2. Uranyl speciation (μmolal) versus pH in water, in the presence of phosphate (PO_4^{3-}) (Rockware, 2008)

Project

In FY 2008, we revisited the reaction of SO1460Na with UAc in EtOH and investigated the role of pH on uranium emission in various solvents. To control pH, reactions with SO1460Na and SO1444 in 2-amino-2-methyl-1-propanol (AMP, 500 millimolar, [mM]) and/or triethanolamine-(TEA, 500 mM) buffered EtOH were performed in the absence of and presence of iron. Due to uranyl quenching by EtOH, supernatants and precipitates from reactions of U(VI) in acetone, ACN and THF with promising ligands (1460Na and 1450Li) were also explored. U to ligand ratios varied from 0.5 to 2.0 and solution pHs were noted (Table 1). Emission spectra were collected for all samples and decay rates were calculated for samples with sufficient emission when excited by 355-nm light. Quenching studies were also performed.

We again employed an off-site contractor to prepare test samples and carry out quenching studies due to restrictions on performing reactions with uranium at National Security Technologies, LLC (NSTec). Dr. Marc Schrier (Calchemist) prepared sealed samples for optical analysis and performed pH and quenching studies as part of this work. Calchemist sent evaporated supernatants and precipitates from reactions of POSS and U(VI) to NSTec's Special Technologies Laboratory (STL) in

secure sample holders. At STL, emission spectra were collected for all samples (usually $\lambda_{\text{ex}} = 310, 355$, and 400 nm^4) and decay rates were calculated for samples with sufficient emission when excited by 355-nm light (the laser-induced fluorescence imaging [LIFI] excitation wavelength). At the University of Nevada, Las Vegas (UNLV), U(VI) emission and the extent of iron quenching in the presence and absence of POSS, in buffered EtOH solutions, were investigated.

pH Measurements

We first investigated pHs of relevant solutions and the impact of acid on POSS ligands. Organic unbuffered UAc and uranyl perchlorate [U(per)] solutions are very acidic. For example, the pH of 10 mM U(per) in acetone is 2, whereas solutions of POSS in organic solvents are basic (see first column of Table 1). When acetic acid (HOAc) is added to POSS solutions, the ligand precipitates or the solution becomes viscous, most likely due to silanol condensation. The pHs attained when unbuffered POSS and uranyl solutions were mixed are listed in Table 2. Final pHs were basic, but the ligands were exposed to acid (Table 2); thus, controlling for pH changes, or minimizing them by using buffers, was essential to preserve ligand chelation properties.

Table 1. pHs and observables after addition of HOAc to POSS solutions (1 mL)

	pH (initial)	Volume of HOAc added (μL)	pH (final)	Observed following acid addition
1 - SO1450Li - THF	8.9	10	6.9	viscous
2 - SO1460Na - Acetone	11.3	2	8.7	ppt
3 - SO1450Li - Acetone	10.3	1	9.3	ppt
4 - SO1460Na - ACN	10.9	1	10.1	ppt
POSS concentrations are 10 mM for 1–3, and 50 mM for 4 ppt = precipitate, eq = equivalent				

⁴ $\lambda_{\text{ex}} = 400 \text{ nm}$ usually resulted in stronger fluorescence.

Table 2. pH of POSS + uranyl in unbuffered solutions following addition of 0.5, 1, and 2 equivalents (eq.) of UAc in the same solvent as the POSS ligand

	Equivalents		
	0.5 U	1 U	2 U
1 - SO1450Li - THF	8.3	8.2	8.2
2 - SO1460Na - Acetone	10.8	10.6	9.6
3 - SO1450Li - Acetone	9.7	9.6	8.8
4 - SO1460Na - ACN	10.1	10.3	10.3

Revisiting the Reaction of SO1460Na with UAc in Unbuffered EtOH

In FY 2007 preliminary observations led researchers to speculate that a new POSS-U(VI) complex precipitated out of solution when an excess of SO1460Na was reacted with UAc in EtOH. To determine whether the observed blue-shifted emission was a result of chelation by POSS, or due to the significant change in solution alkalinity, we added UAc to a basic EtOH solution. Emission of the precipitate (sample 5-3 in Table 3, $\lambda_{\text{ex}} = 400 \text{ nm}$) matched the emission ($\lambda_{\text{ex}} = 355 \text{ nm}$) of the precipitate identified in FY 2007 (sample FY 2007 in Table 3), suggesting that the pH of the POSS solution, rather than coordination by POSS, contributes to the observed blue shift (Figure 3).

The dominant lifetime of the FY 2008 sample is 307 μs , 3.5 times longer than that of the precipitate measured in FY 2007. This difference may reflect differences in sample purity or measurement parameters. Reactions performed in FY 2008 in ACN, acetone, and buffered ethanol appear to give a new SO1460-U complex, and a reaction in THF appears to give a new SO1450-U complex, as will be described below.

Reactions Performed in Acetone, ACN, and THF

Calchemist performed reactions with UAc and POSS ligands in acetone and ACN in hopes of avoiding solvent-induced quenching observed in EtOH. THF was also used, despite uranyl quenching,⁵ based on a report that in THF, 2 eq. of POSS coordinates to U in the presence of triethylamine (Lorenz, 2000). U to ligand ratios were increased from 0.5 to 2.0 and final solution pHs were noted earlier (Table 1). Solutions were not buffered; instead base (NaOH) was added to UAc solutions to match the pH (see Table 1) of the solutions prepared when POSS ligand (1460Na or 1450Li) was

⁵ Even after heating to 100°C ($\lambda_{\text{ex}} = 254 \text{ and } 365 \text{ nm}$).

added. Emission spectra of evaporated base and UAc supernatants were collected and compared to emission spectra attained for corresponding reactions with UAc and POSS, in order to confirm that new emission spectra were due to ligand complexation rather than solution pH. Samples prepared with SO1460Na and SO1450Li emitted sufficiently with 355-nm LIFI excitation, and gave emission spectra ($\lambda_{\text{ex}} = 400 \text{ nm}$) suggestive of reactions.

Figure 3. Overlay of the emission spectra collected for the FY 2007 precipitate from the reaction of excess SO1460Na + UAc in EtOH (blue, $\lambda_{\text{ex}} = 355 \text{ nm}$), and the precipitate generated when NaOH is added to a solution of UAc in EtOH (green, $\lambda_{\text{ex}} = 400 \text{ nm}$)

Average lifetimes and emission band maxima for sufficiently emissive samples are listed in Table 3. For comparison, samples 6-7 to 6-10 correspond to UAc in solvent with no POSS added.

The reaction of SO1450Li in acetone (5-8) appears to yield a product with emission similar to UAc + base in acetone, but the reaction of SO1450Li with 2 eq. UAc in THF gives an apparently new product with emission bands at 496, 516, and 538 nm and a 5.6- μs lifetime (Figure 4a). The short decay time is suggestive of U coordination by THF. The reaction of SO1460Na with UAc in acetone (0.5 and 1 eq. UAc) or ACN (0.5 and 2 eq. UAc) gives a product with emission peaks (507, 527, and 550 nm, [$\lambda_{\text{ex}} = 400 \text{ nm}$]) (Figure 4b) corresponding to the product observed when U(per) is reacted with SO1460Na in 500 mM AMP-EtOH-HNO₃. Dominant lifetimes (>97% component) for the products obtained with an excess of ligand (0.5 eq. UAc) were 207 μs (product from acetone) and 386 μs (product from ACN). Differences in lifetime may be due to sample impurity.

Table 3. Summary of lifetime and spectroscopic data collected for some samples prepared by Calchemist;
 $\lambda_{\text{ex}} = 400 \text{ nm}$, with the exception of FY 2007 sample where $\lambda_{\text{ex}} = 355 \text{ nm}$

Sample	Ligand	Solvent	Equivalents U/ supernatant or precipitate	Ave. Lifetime (μs)	Emission bands
5-5	1450Li	THF	2/super/40C	5.6	496, 516, 538
5-8*	1450Li	acetone	2/supernate	44	462, 480, 500, 518
5-6	1460Na	acetone	1/super	127	507, 527, 550
5-9	1460Na	ACN	2/super	218	507, 527, 550 (453 broad band)
6-2	1460Na	acetone	0.5/super	207	507, 527, 550
6-5	1460Na	ACN	0.5/super	387	507, 527, 550
6-7	X	acetone	X	73	484, 503, 523
6-8	X	ACN	X	290	459, 478, 498, 520
6-9	X	THF	X	5	488, 508, 532, 558 (clean)
6-10	X	EtOH	X	10	peaks at 485, 493, 514, on broad band (ca. 521)
5-3	X	EtOH	NaOH/ppt	307	460, 478 (st), 498 (st), 521
FY07	1460Na	EtOH	xs ligand/ppt	ca. 66	460, 478 (st), 498 (st), 521
*This emission spectrum roughly matched that of UAc + NaOH in acetone (no POSS, super) Super = supernatant , ppt = precipitate, st = strong					

Evaporated supernatants from the reaction of SO1460Na with UAc (1 POSS:0.5 U, and 1:1) in acetone and SO1450Li with UAc (1:2) in THF were sent for analysis by fast atom bombardment mass spectroscopy. A peak observed at 1541 is suggestive of a SO1460-U complex.

Figure 4. (a) Emission spectra comparing dried UAc after exposure to THF (red trace) and the product from the reaction of 2UAc + SO1450Li in THF (brown trace); (b) emission spectra comparing dried UAc after exposure to acetone (blue) and the product of the reaction of 0.5 UAc + SO1460Na in acetone (red).

Quenching Experiments in Acetone

Quenching experiments for the SO1460Na reaction in acetone were also performed by Calchemist. To determine the effect of SO1460Na on U(VI) emission intensity (I), the I of UAc was measured before and after ligand addition. When 10 mM SO1460Na (1 mL) was added to a 10 mM UAc (1 mL) solution in acetone, emission ($\lambda_{\text{ex}} = 365$ nm) was reduced by nearly 85% (band at 518 nm: $I_{1460\text{Na}}/I_{\text{U-only}} = 5/31$). (An absorbance spectrum of the UAc + SO1460Na solution confirmed that there was indeed absorbance at 365 nm.) Iron (0.5 mM) did not have a significant effect on quenching in the presence of SO1460Na ($I_{\text{Fe}}/I_{1460\text{Na}+\text{U}} = 42/50$). Greater quenching by iron was observed in the absence of POSS. As EtOH experiments suggest (see below), POSS may be inhibiting bimolecular quenching by iron, as a result of iron coordination by POSS. It is also possible that there is a pH effect in the unbuffered acetone, because the iron salt is acidic.

Emulating UO_2^{2+} Surface Contamination

To emulate surface contamination, 10 μL of 5 mM uranyl perchlorate [U(per)] were pipetted onto a non-fluorescent hydrophilic Whatman Anodisc 25 membrane. After drying, excess POSS (20 mM SO1444 or SO1460Na in EtOH) was deposited over the U residue and allowed to interact for 30 min prior to vacuum filtration. Emission spectra were collected for filter residues and filtrates following filtration ($\lambda_{\text{ex}} = 414$ nm).

In the presence of SO1444 in AMP- or TEA-buffered EtOH, there was no obvious shift in emission bands. However, red-shifted emission with maxima at 507 and 527 nm was observed for the residue

and filtrate after addition of SO1460Na in 500 mM AMP-EtOH-HNO₃ (HNO₃ = nitric acid). Reactions performed in acetone and ACN (described earlier) also appeared to yield this product based on emission spectra. The addition of SO1460 Na (pH 11.9) resulted in an overall decrease (~20%) in uranyl emission intensity (relative to AMP-HNO₃ buffered EtOH only (pH 9.5)).

Emulating UO₂²⁺ Surface Contamination in the Presence of Fe(III)

The same reactions were also performed in the presence of an 11.5-fold excess of iron (III) perchlorate [Fe(per)] dried on the Whatman membrane with U(per). At this concentration, uranyl solid fluorescence was quenched by 50% and uranyl solution fluorescence (in EtOH) was quenched by about 10%. Filtrate pHs did not differ significantly from those listed in Table 4 for U-only reactions. Uranyl emission bands did not shift in the presence of SO1444. A red-shift with maxima at 507 and 527 nm was again observed for the SO1460Na in 500 mM AMP-EtOH-HNO₃ residue and filtrate, and increased emission (up to 2-fold) was observed as a result of the addition of SO1460Na. Since increased emission was not observed in the absence of iron, the increased emission observed with iron may be due to iron chelation by POSS. Chelation of Fe(III) may restrict Fe(III) contact with U(VI), thereby inhibiting quenching (Hay, 2007).

These “surface contamination” experiments indicate U(VI) chelation by SO1460Na, in the absence and presence of iron, but ethanol, despite its low toxicity, is not a suitable carrier solvent for POSS since it causes U(VI) quenching. Acetone is a better solvent for the SO1460Na + U(VI) reaction, while THF appears to be a better solvent for the SO1450Li + U(VI) reaction.

Conclusion

We studied the effect of POSS ligands on the emission of U(VI) under a variety of conditions. It appears that although POSS (SO1460Na and SO1450Li) may coordinate to U, it does not increase uranyl emission to any appreciable extent, and in most cases causes quenching. In AMP-HNO₃-buffered EtOH (a quenching solvent), a slight decrease in emission was observed with SO1460Na in the absence of iron. In the presence of Fe(III), there was a slight increase in emission in EtOH solutions when SO1460Na and SO1444 were added. This may be a result of iron coordination by SO1444 and SO1460Na. In non-quenching solvents, emission nearly disappeared. It is possible that the U(VI) complex formed does not contain the brightly fluorescent uranyl group. IR spectroscopy would likely have revealed whether or not representative O=U=O symmetric stretches were present, but our ability to make such measurements at STL was limited due to restrictions on working with anything but sealed uranium samples. Since a POSS-U compound was not isolated, we were not able to study the emission properties of a pure POSS-U compound. Still, results thus far do not suggest that POSS is a suitable candidate for spray-on use, as hoped.

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HANDHELD NEUTRON SPECTROMETER

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The goal of this project was to develop a proof of concept for a portable neutron identification detector array with directional detection capability utilizing spectral discrimination algorithms for evaluation. In support of this proof of concept, Monte Carlo N-Particle Extended (MCNPx) modeling was performed on an ideal and perfectly matched hexagonal array of borated polyvinyl toluene (PVT) light pipes coupled to photomultiplier tubes (PMTs). Standard spectral histogram techniques and directional detection capability were proposed and simulated with this ideal setup. Follow-on spectral measurements were performed with a single, large, borated PVT detector to demonstrate the viability of detecting radiological sources. After the hexagonal detector array was fabricated, very careful and detailed multiple spectral measurements were carried out. Preliminary analysis indicates that this hexagonal array has limited capabilities for neutron detection and cannot provide directionality without running an extremely laborious algorithm. Its utility for material identification will require further extensive investigation and testing, including other detector materials and configurations, for evaluation of concept feasibility. Based on limited experimental results, the single, large detector shows some applicability in the field of operational health physics as a dual gamma-neutron rate meter, but warrants further effort.

Background

This project intended to develop a lightweight, deployable, handheld neutron spectrometer that could be used by radiological emergency response programs to identify the type of neutron signature from special nuclear materials. We proposed the use of spectroscopy and multiplicity counting along with directional source position to determine neutron type. This work may potentially support radiological searches, if the technology leads to a field-worthy neutron detector that would provide a rapid, reliable neutron spectroscopy capability.

Project

Extensive MCNPx modeling was performed (Hayes, “A theoretical,” 2008; Hayes, “Preliminary,” 2008). These MCNPx simulations demonstrated that a compact hexagonal array of detectors may be feasible for use in neutron source identification, while simultaneously finding the direction of the source relative to the detector array. For these simulations, a compact hexagonal array of borated PVT hexagonal light pipes was used as the detection system. In the MCNPx model, background spectra

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were generated as part of the preliminary proof of concept effort prior to obtaining the detectors. Both the background spectra and source spectra generated with MCNPx proved to be poor estimations of actual spectra acquired due to a lack of knowledge on the quantum light conversion efficiency for the detector material. Background spectra produced were subtracted from each of the seven detectors so that the residual spectral information from any radiological sources could be summed together to produce a so-called “sum spectrum” from the array for analysis. The preliminary calculations looked very promising.

For the experimental studies, two detectors of different sizes were investigated:

- **Saint-Gobain BC-454 Natural Boron-loaded plastic scintillator:** Figure 1 depicts the large detector coupled to a large PMT. The BC-454 has a 5% by weight loading of natural boron, resulting in a ^{10}B weight content of about 1%. The height and diameter of the detector are each 76.2 mm. Its density is 1.025 g/cm^3 and it has a hydrogen to carbon ratio of 1.169. The light output of this BC-454 detector is 48% anthracene, as compared to 64% for an unloaded plastic such as BC-408 (Kamykowski, 1992).
- **SCIONIX Model H38B114/H40-E2-P-X light pipe (plastic scintillator):** The hexagonal array individual detector contains 5% boron, and is 38 mm from face to face. The light pipe is 114 mm long and is coupled to a 40-mm (face to face) hexagonally shaped PMT (Photonis XP 3062B). The hexagonal array, consisting of the borated PVT detectors, the light pipes, and the compact PMTs, is presented in Figure 4.

Figure 1. Saint-Gobain BC-454 Natural Boron-loaded Premium Plastic Scintillator (large single detector). This detector was used in the initial spectral measurement, but was not part of the hexagonal array due to its size.

It was proposed that, for completeness, measurement and testing were to be carried out on each detector type to complete the investigation into the total capabilities of the hexagonal array using a range of gamma emitters (^{133}Ba , ^{137}Cs , and ^{60}Co), as well as a couple of neutron sources ($^{241}\text{Am-Be}$ and ^{252}Cf). In subsequent measurements with the hexagonal array, only ^{137}Cs and ^{252}Cf radioactive sources were used to characterize the detector set.

Characterization of the Large PVT Detector and the Hexagonal Array Detector Set

After the MCNPx modeling results were obtained, two sets of characterization measurements were made of the large single-crystal detector and each individual detector within the hexagonal array.

Characterization of the Large PVT Detector

There was a notable difference in performance (efficiency) between the larger detector and the individual elements in the hexagonal array. The single detector behaved predictably, showing a much higher sensitivity (count rate) to both gamma and neutron sources than the smaller hexagonal array detector set, due to the larger detector volume. Subsequent spectral measurement results indicated little difference between the large and smaller hexagonal array detectors in terms of energy resolution. The energy resolution for all the detectors showed a monotonic increase in their Compton continuum edges with increasing energy. Figure 2a shows a ^{137}Cs spectrum collected using the large PVT detector. As seen in the figure, the Compton edge of the 661-keV energy peak is so broad that only energy range information would be readily obtainable if the detector were appropriately calibrated. The effects from multiple isotopes for the same detector are not clear. Figure 2b shows a spectrum collected with both ^{137}Cs and ^{60}Co sources. As seen in the figure, the Compton edges are present and resolved, indicating that continual development is necessary if these types of detectors would be used to differentiate and identify multiple radioactive sources. A ^{252}Cf neutron source was also used to characterize the large PVT detector. The measured spectrum, along with other radioactive source spectra, taken at 1 m for 5 min is shown in Figure 3.

Characterization of the Hexagonal Array Detector Set

The prototype hexagonal array of borated PVT detectors was fabricated and tested. This prototype is depicted in Figure 4. Figure 5 shows the relative position and placement of the radioactive sources in respect to the array. Each detector was characterized independently as a single detector unit to investigate if matching of the detectors could be obtained. Based on the results, it was determined that all the seven-detector units had their own intrinsic efficiencies and coupling bias to the individual light pipes. In addition, characterization included obtaining the detector response for the type of radiation (gamma and/or neutron), combination of gamma/neutron sources, source distance, and angular dependency, as well as bias from the ORTEC DigiDART to the PMT. In addition, a Tektronix T2430A oscilloscope was used to trace the pulses from each of the detectors.

Figure 2. ^{137}Cs spectra obtained from the large single detector (left); measured spectrum with both ^{137}Cs and ^{60}Co present (right). Two Compton edges from the ^{60}Co can be discerned at 1.33 and 1.17 MeV.

Figure 3. Measured spectra of different sources taken at 1 m for 5 min with the large PVT detector. Note the unique response obtained from the ^{252}Cf neutron source in red.

Figure 4. Borated PVT hexagonal array

Figure 5. Radioactive source placement location at mid-plane and 0.5 m from detector face

Figure 6 shows typical spectral data collected with one of the seven PVT detectors within the hexagonal array. Based on the measurement results obtained from the smaller detector set, it was determined that, due to the poor resolution and small size of the borated PVT detectors, the proposed neutron/gamma discrimination technique would be ineffective in the array. This determination was consistent with literature search results indicating that borated PVT not only provides significantly poorer quality energy resolution ($\sim 30\%$) than the high-purity germanium (HPGe) ($>7\%$), but also even poorer quality energy resolution than for sodium iodide (NaI) detectors ($\sim 14\%$) (Vo, 2002).

Figure 6. ^{137}Cs spectral measurement obtained from hexagonal array detector #1 with radioactive source located at 0.5 m from the detector face at mid-plane

The capability to discriminate n, γ contributions, as well as directional neutron and gamma detection, were discussed and introduced as part of the testing during the measurement phase, but were not stated as an objective of this project. During individual detector characterization with ^{252}Cf (Figures 7 and 8), we noted that the spectral information obtained with the rear detector (#4) yielded better resolution than the spectra from the front detector (#1). We determined that this was due to insufficient moderating material in the front detector. This led us to conclude that the current setup of the hexagonal array detector was incapable of achieving directionality or identification of neutron radiation from a source.

As a result of these measurements, it was determined that the development of pulse-shape discrimination and pulse-height analysis were not warranted.

Figure 7. ^{252}Cf spectral measurement obtained from hexagonal array detector #1 with radioactive source located at 0.5 m from the detector face at mid-plane

Figure 8. ^{252}Cf spectral measurement obtained from hexagonal array detector #4 with radioactive source located at 0.5 m from the detector face at mid-plane

General Observations

Presented below are several general observations noted throughout this project.

- The ORTEC digiDART is not designed for high bias and milliampere current. Significant drift was noticed during operation. The digiDART (Vo, 2002) is also not suited for use with high-gain PMTs (Guerrier, 2008).
- The current hexagonal array setup does not provide suitable spectra for the objectives of this project.
- The seven borated PVT detectors were not matched, and each had its own intrinsic efficiency and required separate bias settings and gain adjustments.
- Current configuration with Pixie-4 is not proven and is not capable of discriminating n and γ components.
- Ongoing work with the Pixie-4 multiplexers may prove beneficial for other design projects. However, it is apparent that pulse-shape discrimination can only occur using a list mode to obtain the variables that can be used to discriminate gamma from neutron pulses.
- A hexagonal array can provide a framework for directionality. A patent currently exists for a hexagonal array utilizing liquid organic scintillating material for directionality determination of incoming fast neutrons. (Aykan, 2003).
- Smaller hexagonal detectors provide poor energy resolution. Only Compton edge signatures were visible.
- ^{252}Cf spectra taken with smaller hexagonal array detectors are indistinguishable from background spectra.
- The current hexagonal configuration would require more algorithm development to deduce directionality.
- The PVT array is extremely temperature-sensitive.
- Further work is warranted using the single large detector, but the application is unrelated to the intended purpose of this work and so, although surreptitious, has potentially lucrative possibilities if developed.

Conclusion

Detailed spectroscopy measurements demonstrated that the spectral resolution of the borated PVT system tested and evaluated for this work does not support the proposed shape-discrimination capability. The concept seemingly has merit if all the correct components are carefully evaluated, tested,

and chosen based on their compatibilities (matching), cost, and ease of operation to meet potential mission definition. If additional funding were available, other solid materials should be considered. There are several compatibilities that should be matched between array elements, such as intrinsic efficiency for each individual detector unit, PMTs, biases, and power requirements for each detector. In terms of discrimination between neutron and gamma rays, the borated PVT should be carefully evaluated to determine the optimal size (thickness versus length, circular versus mini-hexagonal) for such a physical arrangement (i.e., hexagonal or other configurations).

MCNPx modeling is a powerful tool to determine the viabilities of a measurement concept, but carefully designed fundamental physical measurements are equally important to supplement the deficiencies and limitations of MCNPx.

The single large detector could be pursued as a potential dual gamma-neutron dose rate meter for applications in the nuclear industry as a whole, as it exhibited high sensitivity and weighs less than commercial units currently available.

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CEBr₃ AS A ROOM-TEMPERATURE, HIGH-RESOLUTION GAMMA-RAY DETECTOR

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Cerium bromide (CeBr₃) has become a material of interest in the race for high-resolution gamma-ray spectroscopy at room temperature. This project quantified the potential of CeBr₃ as a room-temperature, high-resolution gamma-ray detector. The performance of CeBr₃ crystals was compared to other scintillation crystals of similar dimensions and detection environments. Comparison of self-activity of CeBr₃ to lanthanum bromide (LaBr₃:Ce) was performed. Energy resolution and relative intrinsic efficiency were measured and are presented.

Background

This work investigated a new room-temperature, high-resolution material that has potential to provide performance superior to LaBr₃:Ce, which is often used because its energy resolution at room temperatures is better than sodium iodide (NaI:Tl). LaBr₃:Ce suffers from the presence of a radioactive constituent, ¹³⁸La. CeBr₃ may be the answer for room-temperature, higher resolution spectra with greater sensitivity because of substantially lower internal activity.

Gamma-ray spectrometers are an important tool in the characterization of radiation sources. Ideally, gamma-ray spectrometers should have good energy resolution, high detection efficiency, and low power requirements, and also should be compact in size, lightweight, and portable. No available spectrometer satisfies all these requirements. High-purity germanium (HPGe) systems require liquid nitrogen or cooling engines. Room-temperature, high-resolution systems often have other limitations, such as maximum crystal size or low detector efficiency. In general, scintillation detectors suffer from poor resolution when compared to semiconductor detectors. This project quantified the potential of a new scintillation detector material, CeBr₃, as a room-temperature, high-resolution gamma-ray detector. CeBr₃ is an attractive room-temperature, high-resolution scintillator material because it is very similar to LaBr₃:Ce in terms of scintillation properties, but has a much lower self-activity. The property of scintillators that make both of these preferred over semiconductor detectors is that they can operate at temperatures that semiconductor detectors cannot, eliminating the need for cooling to tens of kelvin.

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Detector-system developers can use this emerging and novel scintillator material whenever there is a requirement to perform the identification of nuclear radioisotopes. CeBr_3 allows for exploitation of high-resolution spectral characteristics with low self-activity. This should promote identification of special nuclear material (SNM).

Gamma energy spectra from CeBr_3 have lower intrinsic backgrounds than those from $\text{LaBr}_3\text{:Ce}$. This is due to the very nature of the crystal material. The isotopic contaminant ^{138}La emits conversion electrons and β -particles with energies of up to 1.7 MeV. The self-activity due to ^{138}La in LaBr_3 has an intrinsic count-rate of ~ 1.5 events/(cm^3/sec) (Shah, April 2005). In addition, the presence of ^{138}La in the crystal leads to the two gamma energy lines in the energy spectrum at 789 and 1436 keV (Mukhopadhyay, 2005). These make the $\text{LaBr}_3\text{:Ce}$ crystal unsuitable for detection scenarios involving extremely low signal strength, or in situations where an extremely large detector mass is required, such as in standoff detection missions. The large mass of material (associated with a large detection surface area and volume) required for these missions would lead to a very large radiation background for $\text{LaBr}_3\text{:Ce}$, hence reducing the sensitivity. CeBr_3 , on the other hand, performs similarly to $\text{LaBr}_3\text{:Ce}$, but without such a large self-activity. This results in an increased sensitivity for this detector material, making this investigation very interesting, particularly for the aforementioned challenging low signal strength or standoff missions. Furthermore, CeBr_3 shows excellent energy resolution (Ra, 2008). Shah (April 2005; December 2005) measured the energy resolution to be about 3% FWHM for both the CeBr_3 and $\text{LaBr}_3\text{:Ce}$ crystals for 662-keV gamma rays (^{137}Cs source) with the scintillators coupled to a photomultiplier tube (PMT).

Project

Part of this work included growing large crystals of the proposed scintillator and evaluating their properties. Three CeBr_3 crystals were grown and coupled to PMTs; the first crystal was $0.5'' \times 0.5''$, and the other two crystals were each $1'' \times 1''$. Radiation Monitoring Devices, Incorporated (RMD), fabricated and used a $0.5'' \times 0.5''$ CeBr_3 crystal (Shah, April 2005; Shah, December 2005). They had agreed to provide their crystal to Saint-Gobain in order to assist Saint-Gobain in growth and benchmarking of a new crystal specifically for this study (Shah, 2007). Figure 1 is a picture of the $0.5'' \times 0.5''$ crystal grown at Saint-Gobain. All of these crystals were used for comparisons with sodium iodide doped with thallium (NaI(Tl)), and lanthanum bromide doped with cerium ($\text{LaBr}_3\text{:Ce}$) of two different-sized crystal dimensions. Another material, lanthanum chloride doped with cerium ($\text{LaCl}_3\text{:Ce}$), which included impurities of rare earth metals, was also compared for performance and self-activity (Mukhopadhyay, 2005).

Data Acquisition

Each of two sizes of CeBr_3 crystals were obtained and benchmarked against NaI(Tl) and $\text{LaBr}_3\text{:Ce}$. We also benchmarked a $\text{LaCl}_3\text{:Ce}$ crystal. The team documented energy resolution and counting efficiency characteristics using a series of isotopes and gamma-ray energies (^{60}Co , ^{137}Cs , ^{133}Ba , ^{152}Eu , ^{241}Am , highly enriched uranium, and plutonium). A resolution of 3.7% was documented for the 662-keV gamma line.

Figure 1. The 0.5" × 0.5" CeBr₃ crystal epoxied to a PMT

Figure 2 displays the background spectra obtained with the newer 1" × 1" CeBr₃ detector taken inside the Lead Shielding Pyramid at the University of Nevada, Las Vegas (UNLV), which was constructed for this project, providing no less than 2-in. of lead on all sides of the detector. This spectrum represents the gross sum of counts acquired over an interval of time of 115 hours.

Figures 3 and 4 show spectra acquired for a 2" × 2" LaBr₃:Ce crystal, also prepared by Saint-Gobain in conjunction with earlier studies on LaBr₃:Ce (Kernan, 2006), and the 1" × 1" CeBr₃ crystal, respectively. The count times were 10 minutes for each spectrum. One immediately sees the features associated with the intrinsic activity for LaBr₃:Ce from the background count. From Figure 3 one sees in all of the spectra features of the background for the LaBr₃:Ce crystal that include the 789-keV gamma-ray peak, along with an associated β decay spectrum (0–255 keV), and the 1436-keV gamma-ray peak, along with an associated 32-keV x-ray. The isotope source strengths for this work were deliberately chosen in the μ Ci range. The source was placed 40 cm from the LaBr₃:Ce detector to highlight the difficulty of seeing the signal from a weak source in the presence of a high internal background. A couple of salient features bear notice:

- If one compares the ¹³⁷Cs spectra in Figures 3 and 4, one readily sees how much clearer the ¹³⁷Cs stands out when using CeBr₃. Our preliminary data showed a gamma energy resolution of 5.8% (FWHM) for the larger 1" × 1" CeBr₃ crystal (still superior to NaI:Tl). This was compared to the 3.7% resolution obtained with the new 0.5" × 0.5" CeBr₃ detector.
- For the ²³⁹Pu isotope, the peaks were on a larger background when acquired with the LaBr₃:Ce crystal. The signal-to-noise ratio was better for the 1" × 1" CeBr₃ detector. (A larger ²³⁹Pu source was used to enhance the peak definition. The LaBr₃:Ce spectrum for this specific case for ²³⁹Pu was taken with the 1" × 1" LaBr₃:Ce crystal.)

Figure 2. Background gamma-ray spectrum using the 1" $\times$ 1" CeBr_3 detector taken inside the UNLV Lead Shielding Pyramid. This count was taken for 115 hours. Counts are shown as a function of energy (keV).

Modeling

The Monte Carlo N-Particle (MCNP) model of self-activity for the 1" $\times$ 1" $\text{LaBr}_3\text{:Ce}$ detector was refined. Modeling investigation was performed to better understand and model the effects of crystal size on performance. Reported here are the MCNP calculations for the $\text{LaBr}_3\text{:Ce}$ detector. The β spectrum associated with the 789-keV gamma peak was successfully included in the MCNP model. Not included in the MCNP model were the 32-keV x-rays associated with the 1436-keV gamma peak. A Gaussian-Energy Broadening routine was implemented. The simulation results shown in Figure 5 do include coincidence of the 255-keV maximum β with the 789-keV gamma, but not the x-ray coincidence with the 1436-keV gamma. The model for the $\text{LaBr}_3\text{:Ce}$ detector may be extended to the case of CeBr_3 . However, the self-absorption presented by CeBr_3 was low. The plot in Figure 5 shows a comparison between the 1" $\times$ 1" and 2" $\times$ 2" $\text{LaBr}_3\text{:Ce}$ MCNP simulations. The significance here was to provide or project a metric of how self-activity would scale with detector volume.

Figure 3. (a) LaBr₃:Ce background, and LaBr₃:Ce spectra of (b) ⁶⁰Co, (c) ¹³³Ba, (d) ¹³⁷Cs, (e) ¹⁵²Eu, (f) 5% ²³⁵U, (g) ²³⁹Pu, and (h) ²⁴¹Am

Figure 4. (a) CeBr₃ background, and CeBr₃ spectra of (b) ⁶⁰Co, (c) ¹³³Ba, (d) ¹³⁷Cs, (e) ¹⁵²Eu, (f) 5% ²³⁵U, (g) ²³⁹Pu, and (h) ²⁴¹Am

Figure 5. Comparison between the 1" $\times$ 1" (pink) and 2" $\times$ 2" (blue) $\text{LaBr}_3\text{:Ce}$ MCNP simulations

Analysis

We measured the non-linearity of photon yield versus energy for CeBr_3 detectors and determined the linearity to be better than 2% in the 100–1500 keV energy range. The NaI non-linearity is an order of magnitude worse than that for CeBr_3 . This work also showed that the non-linearity for $\text{LaBr}_3\text{:Ce}$ was somewhere in between these two extremes. This finding for $\text{LaBr}_3\text{:Ce}$ is consistent with what has been reported (van Eijk, 2005).

Preliminary estimates of intrinsic efficiency, size effect, self-activity, and resolution were defined. In Figure 6, preliminary results for the self-activity are shown. Figure 6 is a compelling spectral comparison that argues for CeBr_3 . Our work documented a lower background with CeBr_3 than for $\text{LaBr}_3\text{:Ce}$. The intrinsic background observed in the CeBr_3 spectra is mostly due to contaminants associated with the α signatures that are evident in Figure 2. In the critical low-energy areas of the spectrum, where the lower-energy SNM peaks occur, the CeBr_3 had a reduction in intrinsic background.

Figure 6. Self-activity for CeBr₃ (dark blue), LaBr₃:Ce (pink), LaCl₃ (light blue), and NaI:TI (yellow)

Figure 7. Notice that the resolution of the small 0.5" x 0.5" CeBr₃ detector was comparable to the resolution of the 1" x 1" LaBr₃:Ce detector

In Figure 7, results for resolution are given. For the first 1" × 1" crystal delivered, it was determined that the resolution was significantly worse than that of the 0.5" × 0.5" crystal. However, it was learned that Saint-Gobain had prepared a second large 1" × 1" CeBr₃ crystal, which they delivered on request. Saint-Gobain reported the detector resolution for this second large crystal to be 4.4% at 662 keV.

Conclusion

The potential of CeBr₃ as a room-temperature, high-resolution gamma-ray detector has been quantified. CeBr₃ has superior self-activity properties and comparable detector resolution to other room-temperature, high-resolution detectors. This work determined the self-activity properties for CeBr₃ are superior, while the efficiency and resolution properties are comparable to LaBr₃:Ce. Indeed, it has been determined that CeBr₃ may be better than LaBr₃:Ce for detection of certain SNM gamma rays.

Acknowledgments

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DUAL NEUTRON-GAMMA DETECTORS AS NEUTRON ENERGY SPECTROMETERS

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The objective of this project was to explore the feasibility of using dual neutron-gamma detectors as a viable alternative to multiple detector packages. We also examined the possibility of extracting useful neutron energy information from fast neutron detection media, such as liquid organic scintillators (LOS), lithium gadolinium borate (LGB)-loaded plastics, a cesium lithium yttrium chloride (CLYC) scintillator, and a polyvinyl toluene plastic scintillator (BC-408) with an embedded ^3He tube. Presented are significant results of advances made in the determination of neutron energy and time-of-flight (TOF) measurements.

Background

Neutron energy measurements are a small but very significant part of radiological emergency response applications. Mission-critical information can be obtained by analyzing the neutron energy given off from radioactive materials. When trying to locate special nuclear materials, neutron energy information from an unknown source can be of paramount importance.

At the Remote Sensing Laboratory (RSL) of National Security Technologies, LLC (NSTec), a series of materials, namely LOS, LGB, or $\text{Li}_6\text{Gd}(\text{BO}_3)_3$ in a plastic matrix, a recently developed crystal of $\text{Cs}_2\text{LiYCl}_6:\text{Ce}$ (called CLYC), and a BC-408 scintillator with a ^3He tube were studied as to their effectiveness as a portable neutron energy spectrometer. Comparisons illustrating the strengths of the various materials are provided.

Of these materials, LGB offers the ability to tailor its response to the neutron spectrum by varying the isotopic composition of the key constituents (lithium, gadolinium [yttrium], and boron). All three of the constituent elements possess large neutron capture cross section isotopes for highly exothermic reactions. These compounds of composition $\text{Li}_6\text{Gd}(\text{Y})(\text{BO}_3)_3$ can be activated by cerium ions (Ce^{3+}).

CLYC, on the other hand, has a remarkable gamma response in addition to superb neutron discrimination, comparable to that of europium-doped lithium iodide ($^6\text{LiI}:\text{Eu}$). Comparing these two materials, CLYC has higher light output (4500 photoelectrons [phe]/MeV) than that of $^6\text{LiI}:\text{Eu}$ and it shows better energy resolution for both gamma and neutron pulse heights. Using CLYC, gamma-energy pulses can be discriminated from the neutron signals by simple pulse-height separation.

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For the cases of both LGB and LOS, careful pulse-shape discrimination (PSD) is needed to separate the gamma energy signals from neutron pulses. Both analog and digital methods were applied to obtain a clear gamma and neutron energy spectrum in a mixed radiation field. A waveform digitizer manufactured by Agilent Technologies, Inc., was successfully used to digitize the signal and separate the gamma and neutron signals. These results, along with some interesting data from a BC-408 scintillator and ^3He dual gamma-neutron detector, are presented.

Project

BC-408 with an Embedded ^3He Tube

For this project, RSL – Andrews Operations (RSL-A) characterized a dual neutron and gamma detection system consisting of a large (5" × 12") BC-408 scintillator with a central access (>1" diameter × 10" long) to accommodate a pressurized ^3He tube for simultaneous gamma and neutron measurement. The objective of this SDRD work was to quantitatively characterize the gamma-ray response of the BC-408 scintillator and evaluate the neutron-counting efficiency of the ^3He tube (using BC-408 as a moderator). Figure 1 shows the dual neutron-gamma sensor along with the associated Nuclear Instrumentation Module (NIM) electronics.

Figure 1. A large BC-408 scintillator with a pressurized ^3He tube inserted at the center for use as a dual gamma and neutron detector

A check source of the radioisotope ^{22}Na was used to quantify the gamma response from the BC-408 in a source pull-by experiment. A significant alarm level of G7 was recorded, corresponding to a net count of 64–128 times higher than the standard deviation of the ambient gamma background counts.

Gamma alarm levels are defined on a decade scale as the number of times the net gamma counts exceed the standard deviation of the ambient background. If T , B , and σ are the total, background, and standard deviation of the background, respectively, then the gamma alarm, GN , will be defined as:

$$GN = 1 + A \log_2 (\text{Int}(T-B)/\sigma). \quad (1)$$

This same formula applies for neutrons.

Figure 2. A small gamma source (^{22}Na) was pulled by the BC-408 scintillator with -975 V and MACS-II electronics, causing a gamma alarm of level 7

With the ^3He tube powered at a high voltage of $+1750\text{ VDC}$ and surrounded by a plastic cylinder, a small ^{252}Cf neutron source was used for a neutron source pull-by experiment. This time, a neutron alarm level of N6 was obtained, corresponding to a net count of 32–64 times higher than the standard deviation of the ambient neutron background (Figure 3).

Neutron count rates from the ^3He tube are shown in Figure 4 (using Radiation and Environmental Data Acquisition and Recorder [REDAR] electronics) (EG&G, 1981) with and without the plastic cylinder providing moderation; a factor of 6 gain in count rates was observed when the plastic was covering the ^3He tube.

Monte Carlo N-Particle (MCNP) Simulation of BC-408

A Monte Carlo simulation of the track-length estimates of neutron flux (F4 tally in MCNP 5) was performed for the pressurized ^3He tube with and without the BC-408 scintillator/moderator. The actual physical setup for MCNP 5 is shown in Figure 5. A large ^{252}Cf source was placed 100 cm

away from the ^3He tube. The simple particle flux obtained within the pressurized gas tube was $9.95036 \times 10^{-6} \text{ cm}^{-2}$ (with BC-408), and that inside the BC-408 was calculated to be $1.04142 \times 10^{-5} \text{ cm}^{-2}$, accounting for 95.5% efficiency of the ^3He tube to detect thermal neutrons.

Figure 3. A small ^{252}Cf was pulled by the plastic scintillator with an embedded ^3He tube (at +1750 VDC) and MACS-II electronics, causing a neutron alarm of level 6

Figure 4. Effect of plastic cylinder wrapped around the ^3He tube, yielding a factor of 6 gain in neutron counts due to thermalization of the energetic neutrons. A bare ^{252}Cf source was used.

Figure 5. (left) ray tracing within the cells (cell 1 is ^3He tube, cell 4 is BC-408); (right) ^3He tube inside the BC-408 scintillator. The track-length estimates of particle flux (in units of $1/\text{cm}^2$) in the ^3He tube (with and without the BC-408 cylinder) are 9.95036×10^{-6} and 8.45114×10^{-6} , respectively, but when the absorption cross sections are folded in, the effective fluxes are 5.68605×10^{-7} and 2.00158×10^{-9} , respectively.

CLYC Study

$\text{Cs}_2\text{LiYCl}_6:\text{Ce}^{+3}$ (CLYC) is one of the most promising new scintillators for dual neutron-gamma detectors. For small crystals measuring up to 1 cm^3 , light output of 20,000 photons per MeV of gamma energy and 70,000 photons per neutron have been measured (Glodo, 2008). In our work, a FWHM gamma-energy resolution of $\sim 5\%$ yielded at 661.6 keV (from a check source of ^{137}Cs) (Figure 6). The main optical emission due to the activators Ce^{3+} was at 390 nm. Crystals also exhibited core-to-valence luminescence, observed only by gamma excitation at a wavelength of 325 nm, allowing us to distinguish between neutron and gamma pulses via PSD.

CLYC Response to ^{252}Cf Source

Figure 7 shows the pulse height spectrum taken under irradiation from a moderated ^{252}Cf source. The neutron peak was well-separated from low-energy gammas and is equivalent to 3.2 MeV (equivalent gamma energy). Considering that the neutron interaction on ^6Li produces two charged particles of alpha and tritium ions at an energy of 4.79 MeV, the conversion efficiency is about 67%. The

measured neutron peak has a regular Gaussian shape with a 2.9% FWHM energy resolution. This excellent energy resolution comes from the tremendous number of photons generated from the neutron (~70,000 per neutron).

Figure 6. ^{137}Cs gamma-energy spectrum as observed by the CLYC sample. The peak at 661.6 keV and the Compton edge are clearly seen. The FWHM resolution is about 4.5% (much better than NaI:Tl resolution).

Figure 7. The thermal neutron peak appears at 3200 keV; the peak at 477 keV is source-related. The spectrum needs further study to ascertain the source of this line, as we've determined it is not self activity. Curiously, the energy is coincident with the energy from a $^{10}\text{B}(n, \alpha)\gamma$ reaction. The polyethylene shielding materials must have been ^{10}B -loaded.

LOS

Figure 8 depicts the setup for the BC-501A LOS used in this study; it was procured for earlier SDRD work (Leander, 2007). The LOS is ideal for discriminating between neutrons and gammas. It is well documented (Lee, 1998; Sellin, 2008) that PSD may be used to distinguish neutron and gamma events detected by an LOS. The basis for our neutron/gamma selectivity was PSD on the signals from the LOS. The neutrons that deposit energy into the LOS take longer to do so than a gamma ray because of their mass and interaction with protons in the detection medium, with the result that neutrons take much longer to deposit their energy into the detection medium than gammas; therefore, a differential time-to-pulse-height spectrum should show a fast and relatively sharp gamma response followed by a slow and spread out neutron response. Using a leading-edge trigger as the timer start and a crossover trigger as the timer stop provided the best neutron discrimination from gammas. The sources used to obtain the data included ^{137}Cs , ^{133}Ba , ^{22}Na , ^{241}Am , and ^{252}Cf . The sources were placed roughly 10 cm away and had an activity of $\sim 1 \mu\text{Ci}$.

Figure 8. The BC-501A LOS used as a dual gamma and neutron detector

First, a comparison of the figure of merit (FOM) for neutron/gamma PSD was performed using the rise-time, fall-time, pulse-width, and zero-crossing techniques. A FOM of 3 was established using the edge/crossover technique. The starting point was the creation of the PSD spectrum, created by digitizing time for an aspect of the pulse generated by the LOS. A ^{252}Cf source was used, which provided a wealth of gammas and neutrons. One can define a FOM as:

$$FOM = \frac{\text{centroid}_n - \text{centroid}_\gamma}{FWHM_n + FWHM_\gamma}. \quad (2)$$

Using this formalism, the PSD spectrum may be analyzed and the experimental setup tuned or modified to enhance the FOM, and thereby the discrimination between neutron and gamma signatures

in the LOS. In Figure 9, the progress made on improving the circuitry for the PSD is depicted, which was key to the selection of neutron signatures in the LOS. The gamma signatures were typically more narrow in time (left peak), while the neutron signatures were broader in time (right peak). Using this discrimination to gate a differential pulse-height spectrum yielded a response spectrum. The energy of the neutrons was found to within a few hundred keV. Significantly, this detector can simultaneously obtain energy spectra in the gamma and neutron channels. More significantly, changes in the energy spectra measured for the AmBe and ^{252}Cf sources were observed, indicating there is a signature that correlates with the energy field of neutrons.

Figure 9. The edge/crossover PSD technique yielded the best FOM. PSD results for the fall-time, pulse-width, and rise-time techniques are also shown.

The efficiency measurement using a TOF technique was determined using a plastic detector for the associated gamma coincidence counting. The TOF spectra were generated for neutrons to a specific point from a specific fission event in the ^{252}Cf source (Figure 10). The TOF was measured using the gamma detection as the clock “start” (using a BC-408 scintillator) and the neutron detection as the clock “stop” (using the LOS). The next step was to calculate the energy efficiency of the LOS using the 1-m TOF measurement, along with the known partition function for the energy spectrum of neutrons emitted from a ^{252}Cf source. Significantly, gamma-energy spectra typical of an LOS were

obtained simultaneously. The energy spectra obtained for ^{137}Cs (blue) and for ^{60}Co (green) sources acquired with the LOS in the ungated mode are shown in Figure 11. Notice the clean difference in energies for the Compton edges associated with the 662-keV peak (blue) and the 1173 and 1332-keV peaks (green).

Figure 10. TOF spectrum for ^{252}Cf . In the ungated spectrum (blue), both gammas and neutrons are observed. In the gated spectrum (pink), only the neutron signature is evident.

Figure 11. LOS energy spectra obtained for ^{137}Cs (blue) and ^{60}Co (green)

Aerial Neutron Data Acquisition Flight

A flight test was performed to compare the performance of LOS to ^3He detectors for air missions. Six-foot ^3He neutron-detector modules, used by the NNSA Office of Emergency Response Program, were used. Detector brackets for the helicopter for this flight were designed for this investigation. The neutron data acquisition flight was conducted on September 18, 2008. Figure 12 provides a comparison of the data for the two detector systems.

Figure 12. LOS data (left); ^3He detector data (right). A helicopter was flown for this data acquisition over Government Wash near Lake Mead.

Conclusion

A BC-408 scintillator with an embedded, pressurized ^3He tube was effectively used as a single-package, dual neutron-gamma detector for performing search and localization operations. The prototype package, when fully assembled, weighs less than the current NNSA backpack systems, yet has approximately the same sensitivities. As well, a large crystal of CLYC showed excellent promise of being a single-crystal source for dual neutron-gamma detection. Its energy resolution was better than that of $^6\text{LiI:Eu}$. A small radioisotope identification device could be developed using a $2'' \times 2''$ crystal of CLYC. Finally, the LOS demonstrated potential to serve as a dual neutron-gamma detection tool, able to acquire aerial data and distinguish between various neutron sources.

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NEUTRON ACTIVE INTERROGATION USING A SMALL NEUTRON GENERATOR

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This work focused on investigating the use of the associated particle technique to detect fissionable material. A tagged neutron generator produces 14-MeV neutrons that generate fission within any fissionable material that may be present in the neutron flux. The fission process produces up to about 14 gamma rays as well as several neutrons. A time window is opened for 90 ns after the tagged neutron is produced and triple coincidences (α , γ , γ) are counted as a function of neutron flight time. This work has shown that it is possible to distinguish between fissionable and non-fissionable materials based on these coincidence count profiles. This technique has proven to be sufficiently sensitive and relatively hazard-free, such that it can easily compete with other known neutron interrogation techniques.

Background

The tagged neutron technique uses the deuterium-tritium reaction that produces a 14-MeV neutron and an alpha particle that propagates at about 180 degrees opposite the neutron. A two-dimensional alpha particle detector (Figure 1) placed in close proximity to the neutron generator target registers the arrival and location of the alpha particle on the alpha detector. The detection of the alpha particle therefore gives the moment of production and the direction of travel of the neutron. Such a neutron is said to be tagged by the alpha particle. The speed of a 14-MeV neutron is fixed at 5.13 cm/ns; for a gamma-ray photon, it is fixed at 30 cm/ns. Since the moment of production of the neutron and the time of arrival of the photon are known, it is possible to determine the interaction point of the tagged neutron in the interrogated object. Such data can then be used to form an image of the object. Forming an image in this way is called associated particle imaging (API). This technique was developed at the Special Technologies Laboratory (STL) in Santa Barbara, California, in the early 1990s (Beyerle, 1991). Development work focused mostly on the imaging and identification of explosives; however, imaging work was performed at STL in 1998 using depleted uranium (Tinsley, 1998). The scope of this project was carried out using the API system located at STL, because the system could be easily modified to meet the specific needs of this project.

The concept here involved using a tagged neutron generator to irradiate an object to produce fission if fissionable material were present. The fission process was expected to produce about 14 gamma rays (7 prompt and 7 delayed) and several neutrons. The scope of work presented in this report focused on measuring triple coincidences between the alpha particle in the generator and the simultaneous detection of gamma rays in two NaI detectors as a function of neutron time of flight. The tagged

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Figure 1. Principle of tagged neutron generation (not to scale). Both deuterium and tritium are accelerated into a target that is also loaded with deuterium and tritium. The detection of the alpha particle gives the time of production and direction of travel of the neutron, thereby “tagging” the neutron.

neutron was expected to generate a gamma-ray “spike,” due to the production of prompt gamma rays from fission at the location of the material. If the target material were fissile, then a tail would be expected, due to further fissions caused by neutron multiplication.

The goal of this project was to determine the potential to provide a tagged neutron interrogation system that would be safe, easy to use, and deployable. The objectives were to perform proof-of-principle experiments and to develop a technically solid and confident design.

Project

The API system at STL has a tagged neutron generator that produces about 3×10^7 n/s into a 4π solid angle. The tagged neutron beam is continuously emitted into a cone that has a vertex angle of 13 degrees. The system has two detector “arms” separated from each other by 45 degrees, as shown in Figure 2. Each arm consists of a 4×4 -in. NaI detector shielded from the generator target by stacked lead bricks, so that one arm has a lead thickness of 30.5 cm and the other 17.8 cm.

We initially used the system to perform a background run with no interrogation target in the tagged neutron beam. It was found that there was an interaction between the two detectors caused by neutron-induced gamma-ray production in one detector being “seen” by the other detector. This resulted in

an inflated accidental triple coincidence rate that was expected to reduce the signal-to-noise ratio (SNR) of the technique. The problem was solved by the placement of 3.8 cm of lead between the two detectors, so that one detector would be blind to the other (not shown in Figure 2).

Figure 2. Layout of the API system used for this work (not to scale)

The first run using an interrogation target in the tagged neutron beam was performed using a 10-kg depleted-uranium (DU) plate located 60 cm from the neutron generator. The plate was sufficiently large, such that the tagged beam only irradiated a portion of the plate that had a mass of 3.7 kg. The irradiation time was 2.7 days. The lower-level discriminator was set at 60 keV. The system measured the time between the production of a tagged neutron and the detection of a gamma ray in either detector. The result is shown in Figure 3.

As can be seen, the tagged neutrons reached the material and generated a prompt gamma-ray spike due primarily to fissions. Two coincidence arms are also observed radiating away from this spike at 90 degrees relative to each other. These arms peak at about 19 ns after the main gamma-ray spike, and are due to the arrival of prompt fission neutrons at the detectors. The background level at the base of the spike is 2.2 counts per channel, and therefore gives an SNR of about 140:1. This was computed

Figure 3. Results for the DU run. This is a three-dimensional graph of the number of coincidences versus time between neutron production and gamma-ray detection in each detector/arm of the system.

by dividing the maximum of the peak by the background level. Application of the Currie equation to calculate the minimum detectable mass indicated that the smallest mass distributed evenly over the tagged beam area that could be seen at 60 cm was 116 g, assuming the beam size was small enough to be completely intercepted by the mass. This corresponds to being able to see 48 g of ^{235}U or 55 g of ^{239}Pu at 60 cm. Since the cross sections of both of these nuclides is at least a factor of 2 greater than that of ^{238}U , it is reasonable to expect at least twice as many counts from the same mass of fissile material. This DU result is estimated as equivalent to being able to see 1.6 kg of ^{239}Pu or 1.8 kg of ^{235}U under similar conditions of distance and beam size. Fissile material will also display some degree of neutron multiplication, and is therefore expected to give enhanced coincidence arms beyond what was observed for DU.

Runs were also performed for lead, tungsten, and iron interrogation targets placed at 60 cm. The run times were 4.1 days, 3.1 days, and 5.4 days, respectively. Each plate was 1.3 cm thick and had dimensions sufficiently large to cover the area of the tagged neutron beam. The system setup was the same as that used for the DU run. The results are shown in Figure 4, where the count times are normalized to that of DU. These results show a gamma spike for each material and weak coincidence arms radiating

away from it. Note that the amplitude and profile of the DU gamma spike is far greater than that of the other materials studied. The DU result is also different due to the presence of the prompt neutron peak on each coincidence arm.

Figure 4. Comparison of results for DU, lead, tungsten, and iron for one of the coincidence arms (irradiated volumes were equal in each case)

Calculations performed by a Russian group (Kuznetsov, 2006) that built a similar system show that both a prompt gamma pulse and a prompt neutron pulse that can be sensed by detectors sensitive to each type of radiation must be present. NaI displays some sensitivity to neutrons, which is why we saw a prompt neutron peak in our data. Their calculated coincidence versus neutron flight-time profiles are remarkably similar to the coincidence arms observed in our data.

Conclusion

The data collected in FY 2008 indicate that this technique gives a unique signature of fission. We also observed both prompt gamma and neutron pulses in our data. Fissionable material was distinguished based upon the gamma-ray spike alone; however, the observation of a neutron spike displaced by the neutron flight time was a definitive indicator of fission. The signature of fissile material is expected to be even more unique than observed here for DU.

Path Forward

Dose rate calculations indicate there are unlikely to be issues during operational use, due to exceeding the recommendations of ICRP-60 (1991). The microchannel plate (MCP) used in this work was about 3% efficient; however, a new MCP is anticipated to be about 85% efficient. The MCP used for

this work was about 3 years old and had been used extensively during that time. Its efficiency had decreased due to degradation from use. Modifications of this system to incorporate a new MCP and larger NaI detectors, or one or two more detectors, may result in an increase in count rate by several orders of magnitude. This leads to the hypothesis that data presented in this report may be acquired in minutes of operation rather than days. If all coincidences (triple and above) are accepted, then the coincidence rate would be expected to rise approximately according to $2^n - (n + 1)$, where n is the number of detectors.

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RADIATIVE DECAY ENGINEERING FOR IMPROVED SCINTILLATORS

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Our goal was to explore the effect of covalently bonding, singlet-emitting organic dyes to metallic nanoparticles (NPs) as a means to create faster and brighter scintillator materials. Toluene-soluble silver (Ag) NPs were fabricated, and the synthesis of a conjugated polymer with thiol endcaps (CP-SH) for coordination to AgNPs was nearly completed. Due to an unforeseen synthetic hurdle, however, CP-SH could not be delivered by the subcontractor. While the synthesis of CP-SH was being attempted, a ~160 nm Stokes-shifted conjugated polymer, PFPB, was synthesized and characterized. Synthetic schemes for CP-SH and PFPB are provided, and the results of scintillation tests and fluorescence studies for PFPB and a ternary cocktail, comprised of 2-(4-tert-Butylphenyl)-5-(4-biphenyl)-1,3,4-oxadiazole (tBu-PBD) and PFPB in toluene, are described. We report that the two-solute mixture of tBu-PBD (1%) and PFPB (0.2%) (1-PB02) emitted at ~520 nm with 44% scintillation strength relative to a comparably sized, commercially available plastic cylinder (BC-454).² Scintillation yield (SY) from 1-PB02 was greater than two times stronger than that from 3-hydroxyflavone (0.25%), and the cocktail had a scintillation decay time of 2.5 ns.

Background

Metal NPs to Improve Scintillator Output

Faster and brighter scintillators are required for improved nuclear radiation detection and radiography. A method to accelerate scintillator speed, without compromising light yield, and improve scintillator brightness would be of great value in the areas of radiation sensor development and pulsed-power diagnostics.

It has been demonstrated that the interaction of phosphorescent materials (excited optically or electronically) with surface plasmon (SP) resonances (such as from AgNPs) can both amplify emission intensity, due to the metal increasing the local incident field on the fluorophore (the “lightening rod” enhancement effect), and accelerate radiative processes, leading to faster decay. These effects are results of (1) the metal allowing the fluorophore to absorb more light, and (2) the metal introducing a new radiative decay path. This complex phenomenon has been termed metal-enhanced fluorescence (MEF). MEF can be achieved using appropriately sized metal NPs, metal gratings, or rough films in close proximity to singlet and triplet emitters. The effect observed depends upon many factors,

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² All SY values were corrected for the wavelength-dependent sensitivity of the photomultiplier tube (PMT) photocathode using a typical S-20 sensitivity curve (Mansour, 2008).

including chromophore absorption properties, the distance between metal and chromophore, the refractive indices of the metal and its immediate environment, and colloid size and shape. Larger metal colloids are expected to absorb less and scatter more, resulting in enhancement (Lakowicz, 2005). If parameters are not optimal, metal surfaces can cause quenching. Lakowicz explains the metal-induced phenomenon based on the concept of the metal surface as a radiating plasmon. According to this model, the effect of metals on fluorophores is explained by electrodynamics, Mie theory, and Maxwell equations.

This field of study is new and evolving. Zhang (2007) reports that gold particles enhance emission of Alexa Fluor dyes (synthesized through the sulfonation of coumarin, rhodamine, xanthene, or cyanine) by 6- to 10-fold. Decay rates are also increased. In addition to a 20-fold increase in emission intensity, an 8-fold decrease in lifetime was reported for Rh800-doped, silica-coated silver nanoparticle assemblies (Aslan, 2007).

We proposed studying the effect of metal nanoparticles on fluorophore emission generated by ionizing radiation. Since plasmon-induced phenomena require that the metal surface and chromophore be in close proximity, we proposed covalently bonding singlet-emitting conjugated polymer dyes to metallic NPs. These hybrid NPs could then be dissolved in an aromatic scintillator solvent, such as toluene, and SYs and decay times could be measured and scintillation strengths compared to determine the effects of the metal NPs on chromophore emission.

Stokes Shift

When a molecule is excited, excitation is generally into a higher vibrational state, but emission is from the lowest vibrational level of the excited electronic state. Therefore, the average wavelength of light emitted is greater than that of the absorbed radiation. This difference is called the Stokes shift. Overlap between the relatively broad absorption and emission bands causes self-absorption of emitted light, resulting in reduced intensity of transmitted light. A large Stokes shift, leading to minimal overlap, helps decrease reabsorption. Large Stokes shifts are especially important in applications that require large detector dimensions, such as in scintillating fibers (D'Ambrosio, 1991), where light must propagate efficiently over distances of several meters or more.

Project

Synthesis of AgNPs, PFPB, and the attempted synthesis of an emissive conjugated polymer with thiol end caps (CP-SH) were performed in the laboratory of Dr. Guillermo Bazan at the University of California, Santa Barbara (UCSB), by UCSB staff scientists.

PFPB, a conjugated polymer (CP) with a large Stokes shift, was synthesized by incorporating a green-emitting dye, 2,1,3-benzothiadiazole (5%), into a blue-emitting polymer. Energy transfer from the blue to the green-emitting portion can occur radiatively and/or non-radiatively (Forster resonance

energy transfer). The suitability of the polymer as a primary and secondary scintillation solute was studied. A Stokes-shifted polymer could eventually be thiol-capped to permit coordination to a metal NP.

AgNP Synthesis and Description

The synthesis of oleic-acid-protected, toluene-soluble AgNPs was achieved following the procedure described by Lee (2007). AgNPs were characterized by transmission electron microscopy (TEM) (Figure 1) and absorbance spectroscopy. Maximum absorbance was at 423 nm. Prior to use, AgNPs were dispersed in toluene and filtered. It was found that the surfactant molecules (oleic acid) were only weakly associated with the NPs, and dissociation of surfactant and aggregation of NPs occurred when the toluene solution was exposed to light. We expect that stronger CP-SH coordination would diminish or prevent AgNP light sensitivity in liquid solutions. In solid organic polymer solutions such as polyvinyl toluene (PVT), aggregation in the presence of light would not be a concern.

Figure 1. TEM of AgNPs shows good dispersion in toluene. A Gaussian fitting of the data (inset) reveals a mean diameter of 7.2 nm with a standard deviation of 1.4 nm.

CP-SH Synthesis

The synthesis of CP-SH was attempted according to Figure 2. The final step to deprotect the thiol group was not successfully completed by the subcontractor. Without this polymer, we could not carry out the proposed studies.

Figure 2. Scheme for the synthesis of CP-SH ($\text{R} = \text{octyl}$). Deprotection of the thiol group in product **6** could not be achieved. The lower right cartoon represents CP-SHs coordinated to a AgNP. On the order of 2000 chromophores may coordinate to a AgNP.

PFPB Synthesis and Description

PFPB, a polymer with a 160-nm Stokes shift ($\lambda_{\text{ab}}/\lambda_{\text{em}} \sim 360/520 \text{ nm}$), was studied next. The large Stokes shift is achieved by replacing 5% of the fluorene monomers in poly-((9,9-bis-(2'-ethylhexyl)-fluorene-*co-alt*-1,4-phenylene with green-emitting 2,1,3-benzothiadiazole (BT). It is possible that thiol capping of such a polymer might be especially advantageous when working with AgNPs to prevent reabsorption in a solution with CP and AgNP. Efficient energy transfer between absorbing (PFO component) and emitting (BT) states has been reported for solid-state PFBT (without the additional phenylene units) by Vehse (2004). PFPB was synthesized at UCSB according to the synthetic scheme in Figure 3. The insertion of phenylene groups simplifies the synthesis but likely decreases Forster energy transfer.

Figure 3. Scheme for the synthesis of PFPB ($n = 95\%$, $m = 5\%$)

Evaluation of PFO and AgNPs in PVT

While the synthesis of CP-SH was being attempted, PVT-based thin films containing AgNPs and a fluorophore were fabricated. Pulsed x-rays were used to irradiate binary toluene control solutions and the PVT-based thin films. The excitation source was a Platts x-ray tube with end-point energy up to 450 keV and pulse width of ~ 40 ns. Tests were performed with AgNPs (1.25% in PVT and 0.1, 0.3, and 0.5% in toluene) and unmodified poly[(9,9-dioctylfluorenyl-2,7-diyl) PFO (0.5%).³ PFO does not have thiol groups for tethering to AgNPs, and the subcontractor-provided CP emitted where the AgNP absorbed ($\lambda_{\text{em}}(\text{PFO}) = 414$ nm). Not surprisingly, scintillation tests in toluene demonstrated that when AgNP concentration increased from 0 to 0.5%, signal decreased (in this case by a factor of nearly 10). The exercise resulted in a technique for preparing PVT films (230–700 μm thick) containing PFO solute and NPs, and further emphasized the need for tethering CPs to NPs.

Evaluation of PFPB in Toluene

Figure 4 (top) shows an emission spectrum for a dilute toluene solution (0.06 a.u. in 10-mm cuvette) of PFPB. At an excitation wavelength of either 290 or 360 nm, the emission spectrum differs significantly depending upon solution concentration. The dilute sample emission spectrum contains two bands. The bluer band has peaks at 405 and 427 nm and emulates PFO-only emission; the redder band has a peak at ~ 520 nm and is due to emission from the BT unit. In more concentrated solutions, only

³ When referring to concentrations, % values are by weight.

the redder band is observed due to self-absorption of the bluer band. Photoluminescence (PL) decay times for the bluer emission are short (470 ps), whereas decay times associated with BT emission are longer (2.4 ns) (Figure 4 [bottom]).

Figure 4. (top) PFPB emission from dilute solution. In more concentrated solutions (0.25%), only the redder emission is observed; (bottom) PL decay plots for the ~410-nm and 520-nm emission bands.

Results comparing SYs for PFPB in toluene demonstrate that signal strength increased from 18% (0.125% PFPB) to 35% (1 and 2% PFPB) relative to that from a 10-mm-diameter cylinder of

PVT-based, Bicorn-blended polymer (BC-454 [$\lambda_{em} = 425$ nm]).⁴ Results are summarized in Table 1, Groups 1 and 2. PFPB emission was strong relative to a 0.25% toluene solution of a well-known scintillator with a 190-nm Stokes shift, 3-hydroxyflavone (3-HF)⁵, (D'Ambrosio 1991; Barni 1997), and anthracene (0.25%).⁶

Table 1. Comparison of SYs resulting from x-ray irradiation

Group	Sample	SY relative to BC-454 ⁺	% signal relative to no filter:	
			Short-pass filter [*]	Long-pass filter ^{**}
1	BC-454 cyl	100		
	BC-490	38		
	0.25% 3-HF [#]	18		
	0.25% Anthracene [#]	11		
	0.5% PFBT	16		
2	2% PFPB	35		
	1% PFPB	35		
	0.5% PFPB	28		
	0.25% PFPB	22		
	0.125% PFPB	18	4	80
3	1% tBu-PBD	106	62	3
	1% tBu-PBD+ 0.05% PFPB	61	20	62
	1% tBu-PBD+ 0.1% PFPB	51	13	72
	1% tBu-PBD+ 0.2% PFPB (1-PB02)	44	5	87
4	0.5% tBu-PBD	99		
	0.5% tBu-PBD+ 0.05% PFPB	57		
	0.5% tBu-PBD+ 0.1% PFPB	51		
	0.5% tBu-PBD+0.2% PFPB	51		
⁺ corrected for S20 photocathode sensitivity [*] Oriol 57372: 450-nm short pass ^{**} Corning 3-72: 450-nm long pass [#] At concentrations above 0.25%, emission from these standards decreased				

⁴ BC-454 has a reported scintillation light output of 48% anthracene (anthracene light output = 40–50% of NaI(Tl)), and is composed of PVT (>85% by weight), natural boron (5% by weight), and aromatic fluorophores (<5% by weight). Decay constant (main component) = 2.2 ns.

⁵ 3-HF has been used for particle detection ($\lambda_{ab} = 343$, $\lambda_{em} = 534$ nm, and QY = 0.42 in xylene).

⁶ $\lambda_{em} = 400$ nm.

PFPB SY was also compared to the commercially available PFO-based dye: poly[(9,9-dioctylfluorenyl-2,7-diyl)(90%)-co-(1,4-benzo-{2,1',3}-thiadiazole)(10%)], (PFBT, American Dye Source #ADS233YE) [$\lambda_{em} = 528$ nm, 143 nm Stokes shift]). Scintillation output from PFPB was stronger than that from PFBT at equivalent concentrations (0.5% PFPB, SY = 28%; 0.5% PFBT, SY = 16%).

Addition of tBu-PBD to PFPB in Toluene

The overlap between toluene emission and PFPB absorption was not ideal. To improve light absorption by PFPB, tBu-PBD (2-(4-tert-Butylphenyl)-5-(4-biphenyl)-1,3,4-oxadiazole) ($\lambda_{ab} \sim 305$ nm; $\lambda_{em} = 366$ and 383 nm in toluene, QY ≈ 0.84) was used as a wavelength shifter. The results are shown in Table 1, Groups 3 and 4. Note that 1% tBu-PBD is as strong a scintillator as BC-454. Although the addition of PFPB decreased scintillation from the blue-emitting tBu-PBD-only solution by at least 40%, SY from solutions of PFPB (0.05 to 0.2%) with 1% and 0.5% tBu-PBD nearly doubled relative to scintillation from solutions of PFPB alone. Fluorescence spectroscopy (collected at 90 degrees from the source) demonstrated that, in this configuration, fluorescence emission comes only from PFBT (0.05 to 0.2% in toluene) when tBu-BPD (1%) is a primary solute (Figure 5).

Fluorescence spectra show maximum intensity emission from the 0.05% PFPB with 1% tBu-PBD mixture. At higher PFPB concentrations, emission drops. Scintillation results from the ternary solutions also indicate lower emission with higher concentrations of PFPB. To determine whether the bulk of the scintillation from the cocktail was from the redder PFPB band (as indicated by fluorescence), we placed filters in front of the collection fiber and performed pulsed x-ray tests on the 1% tBu-PBD samples with PFPB concentration ranging from 0 to 0.2%. Data were also collected for a 0.125% PFPB-only solution. A T<450 filter was a good block for PFPB signal, and a T>450 filter was a good block for tBu-PBD signal. Although the solution containing 0.05% PFPB gave the strongest scintillation signal, studies with filters indicated that some of the signal came from tBu-PBD; therefore, not all emission was Stokes-shifted. As PFPB concentration increased, the amount of signal from the bluer emissions became less significant, and for 1-PB02 the scintillation signal was due only to the BT component of PFPB (emission at ~ 520 nm).

Scintillation lifetimes for 1-PB02 and PFBT (0.5%) were measured by time-correlated single photon counting using the Special Technologies Laboratory (STL) fluor-decay system (Flournoy, 1994) with a ^{90}Sr beta emitter. The decay time measured for 1-PB02 (~ 2.5 ns) agreed with the emission coming from the red-shifted BT component of PFPB ($\tau_{PL} = 2.4$ ns). As shown in Figure 4 (top), dilute PFPB had two distinct emission bands (4:1 ratio for peak at 406 nm: peak at ~ 520 nm) with very distinct emission times. In contrast, there was better intramolecular energy transfer in dilute PFBT such that the redder band (528 nm) was 4x stronger than the bluer band (414 nm) ($\lambda_{ex} = 290$ nm). However, SY for PFBT was lower than that for PFPB, and an estimated 50% longer scintillation τ was recorded for the redder band of PFBT in toluene ($\tau_{PL} = 2.75$ ns). To improve the absorbance of toluene emission, tBu-BPD could also be used in a ternary cocktail with PFBT.

Figure 5. Fluorescence spectra, $\lambda_{\text{ex}} = 290$ nm, showing emission from 1% tBu-BPD alone and with 0.05 to 0.2% PFPB present. As PFPB concentration decreases, the bluer PFPB bands (at 409 and 432 nm) become apparent, and PL intensity at 520 nm increases.

Conclusion

The primary goal of this work, to study the effect of coordinating emissive dyes to metal NPs, was not achieved due to problems with the final step in the synthesis of a thiol-capped CP. Organic soluble AgNPs were synthesized, and techniques for the fabrication of AgNP and emissive dyes in PVT mixtures were successful. PFPB, a CP with a large Stokes shift, was studied with and without tBu-PBD as a primary solute in toluene. In the absence of tBu-PBD the SY from 1 and 2% PFPB solutions was 35% relative to BC-454, exceeding that from 3-HF and anthracene at their optimal concentrations (0.25%). SY from the commercially available PFBT (0.5%) was on par with that from 3-HF. Addition of tBu-PBD to PFPB solutions improved PFPB excitation. A fast, green-emitting scintillator with an ~160-nm Stokes shift, 1-PB02 scintillates at ~520 nm with 44% signal strength relative to BC-454. Scintillation decay for 1-PB02 ($\tau \sim 2.5$ ns) agreed with the PL lifetime for the PFPB red-emitting component. For thermal neutron detection, toluene-soluble carborane could be added to the cocktail.

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LASER PLASMA VAPOR DETECTION

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A means of detecting and identifying fluorinated chemicals in the ambient atmosphere was explored. A laser-generated plasma was used as the excitation source for producing atomic emission spectra of chemicals in air. An emission spectrometric system was designed and constructed for field portability and battery operation. Concentration curves were constructed and limits of detection for target compounds were estimated. A means of providing calibrated emission intensities in situ was explored. Additional excitation sources were also investigated. The efficacy of the plasma vapor detection approach for air sampling was demonstrated.

Background

Fluorinated chemicals such as sulfur hexafluoride (SF_6) and 1,1,1,2-tetrafluoroethane (R134a) are routinely released at the Nevada Test Site (NTS) Nonproliferation Test and Evaluation Complex (NPTEC) in support of many chemical release and process simulation campaigns. Both SF_6 and R134a are employed as targets for determining the limit of detection of long-wave infrared imagery and spectroscopy systems deployed by customers at the NTS. SF_6 is widely used as a tracer gas for long-wave infrared imagery for real-time chemical plume trajectory mapping. R134a is used as both a tracer for plume tracking and as a direct-target chemical for chemical process simulation. Detection and identification of open-air or process-stream chemical vapors is an integral component of chemical release activities, wherein stand-alone vapor detectors are required for ground truth diagnostics to provide both real-time chemical identification and concentration monitoring at the release source and in the field. While NPTEC and the Special Technologies Laboratory (STL) currently deploy a suite of chemical vapor detectors during release campaigns, gaps exist in the list of chemicals capable of being monitored. The aforementioned fluorinated gases are invisible to the photoionization detector array and other sensors that are deployed to detect plume position and concentration in time in the release field. Additionally, recent metal vapor chemical releases have been performed without field diagnostics due to the lack of an available detection technique. The goal of this project was to explore a cost-effective means of detecting such chemicals in real time.

The use of atomic emission spectroscopy would allow detection of a broad suite of compounds, with specific identification of individual compounds. The method proposed is based on the formation of a transient high-temperature plasma in air generated by a focused laser pulse, followed by collection and detection of emission lines originating from excited-state atoms and molecular fragments present in the plasma. This technique is generally referred to as laser-induced breakdown spectroscopy (LIBS)

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and has been the subject of numerous investigations over the past two decades (Cremers, 2006; Palleschi, 2006). Most LIBS efforts have focused on solid-sample analysis and, to a lesser extent, liquids. A backpack-sized portable LIBS system has been reported for on-site analysis of soils and other solids (Yamamoto, 1996). Innovations in recent years of very compact, pulsed, solid-state lasers (including battery-operated lasers), and palm-sized spectrometers with small, uncooled, sensitive array detectors allow for the design of a small portable unit for field deployment and real-time vapor detection. The design of such a system and the use of laser-induced plasma atomic emission was explored as a means of detecting chemical vapors in open air and in production streams.

Project

The end goal of this project was the demonstration of gas-phase LIBS as a means of detecting and identifying chemical targets in air. The prototype LIBS system was to be of a size and weight amenable to deployment on a surveyor's tripod, and to be of a cost such that multiple units could be purchased if desired. The system would be required to operate in both manual and automated modes, with samples being collected directly from the ambient atmosphere for real-time interrogation of the presence and identity of chemical species. The tasks leading to the realization of this goal were (1) design of the prototype optical-emission spectrograph system with laser-excitation source, (2) spectroscopy of target gases in air, (3) investigation of calibration sources, and (4) investigation of alternative excitation sources.

Prototype Spectroscopy System

The largest source of weight, power consumption, and size for a LIBS system is the laser-excitation source. The principal investigator was aware of a generation of handheld, self-contained, pulsed Nd:YAG laser units from prior work in the 1980s on laser-induced, second-harmonic generation in crystals (Eddy, 1988). As well, Kigre, Inc., now produces high-peak-power, handheld, flashlamp-pumped lasers providing 25 mJ of energy at 1064 nm in 10-ns pulses for under \$10,000. The Kigre MK-367 laser is shown in the Figure 1 inset. It was found that focusing the laser output from the Kigre laser with a 1-cm focal length plano-convex lens provided sufficient laser fluence to produce a plasma "spark" in ambient air, as shown in Figure 1. The Kigre is also capable of producing a double pulse with 45 mJ total energy. The Kigre control module was modified to allow external triggering, which would be required for autonomous computer-controlled operation.

The second enabling technology for the development of a lightweight, portable LIBS system is a generation of commercially available, small, high-resolution spectrographs by Ocean Optics. Our initial intent was to specify and procure a single spectrograph providing ± 0.1 -nm resolution over the wavelength region corresponding to the most intense fluorine atomic emission lines, 600–800 nm. However, Ocean Optics recently began production of the LIBS2500plus unit, which allows up to four individual spectrographs to be operated simultaneously, with software that "stitches" the spectra together to produce a single high-resolution spectrum over a broad wavelength range. Three

spectrographs, covering the spectral ranges of 197.2–423.3 nm, 423.4–626.3 nm, and 626.4–808.6 nm, were procured to provide spectral coverage from the UV to the near infrared. Each spectrograph is equipped with a 2048-pixel CCD array for light signal collection at the focal plane. Control software provided by Ocean Optics allowed the collection of single-shot, multiple-shot, and multiple-shot-averaged data to be collected and displayed, with an optional elemental peak identification function built in.

Figure 1. Layout of the prototype laser plasma vapor deposition system. The upper right inset depicts the scale of one of the enabling technologies, the pulsed Kigre Nd:YAG laser. The lower left inset shows the plasma glow in air produced by focusing the Kigre laser output.

A block diagram of the prototype system is depicted in Figure 1. The data collection sequence is initiated from the control software, pretriggering the spectrograph via USB. An external trigger is sent to the laser to initiate a laser pulse. A delay board was designed to provide a 32.8-ms delay

followed by a 400- μ s-wide lamp-sync pulse, which is used to trigger a 2-ms CCD collection of the dispersed atomic emission. The timing is set such that the lifetime of the plasma and associated atomic emission signal (tens to hundreds of microseconds) falls well within the CCD collection window to negate any effect of jitter in any of the triggers. The 1064-nm laser pulse from the Kigre Nd:YAG laser is focused using a 1-cm focal length lens to a spot located on the optical axis of a 600- μ m quartz optical fiber. The fiber is mounted on an x,y,z translational stage for signal optimization. The sampling chamber, consisting of a 1-cm pathlength quartz flowthrough fluorescence cell, and the 12-VDC, 500-ml/min miniature air pump were not utilized during data collection due to delays in receipt of the flow cell. Instead, sample gas was directed to the laser interaction area via a Tygon tube at a flow rate of 2 liters/min. Gas mixtures ranging from 0 to 100% (2.5% minimum) were produced using a dual input mixing chamber with 0 to 2 liters/min manual flow control meters by Cole Parmer.

Laser Plasma Spectroscopy of Target Gases

The atomic emission spectrum of neat gas samples, defined as a flow of 2 liters/min gas through the laser interaction zone, is shown in Figure 2. The nonlinear background observed in each of the three spectral regions is due to a white light plasma continuum emission resulting from bremsstrahlung and electron recombination radiation as the plasma cools. The peaks observed in the spectra are due

Figure 2. UV and visible atomic emission spectra of gases utilized in this investigation, demonstrating the unique spectral fingerprints of each chemical

to the subsequent light emission from excited-state neutral atoms and singly or doubly charged excited-state atomic ions formed in the plasma (Palleschi, 2006). A general observation applicable to this study is that emission in the 200–400-nm region is from metals, 400–600 nm is from nonmetals and molecular fragments, and 600–800 nm is from gaseous elements.

Figure 3. Visible atomic emission spectra of SF_6 and R134a, illustrating the disparate features of the two fluorinated chemical compounds

The spectrum of air was dominated by emission lines from neutral and singly ionized nitrogen (N) and oxygen (O) atoms derived from the breakdown of N_2 and O_2 , respectively. The key spectral lines used for analysis in the current study were 500, 742, 744, and 746 nm for N, and 777 nm for O (San-sonetti, 2007). Flowing 2 liters/min of argon (Ar) diminished but did not eliminate ambient air from the laser interaction zone, the consequence of which was that gas concentrations reported in this study are approximations, and subsequent calibrations based on reported concentration values will illustrate observed trends rather than absolute values. The key Ar spectral lines utilized herein were 763, 751, and 738 nm. The spectra of the two fluorinated gases were dominated by fluorine (F) peaks in the 600–800 nm region. The key F peaks used for analysis were 677, 683, 685, 690, 704, and 713 nm (Williamson, 1998). Differences in the spectra arose from the disparate atomic composition of the two gases, allowing the identification of the two species. Figures 2 and 3 illustrate the unique spectral features of these compounds. Fragmentation of SF_6 yields sulfur (S) emissions in the 400–600-nm region, with peaks at 543, 545, and 560 nm being used in the identification of SF_6 (Dudragne, 1998). Fragmentation of R134a ($\text{F}_3\text{CCH}_2\text{F}$) yields a dominant hydrogen (H) atom emission at 656 nm, and

carbon atom emissions in the 200–300-nm region. Also evident are molecular fragment emission lines in the 400–600-nm region due to the diatomic C_2 radical (Xiaoxiang, 1987), and 386 nm due to the CN radical (Tatematsu, 1977), resulting from the reaction of C with N_2 in the air.

Measurements of spectral line intensities at varying gas concentrations in air were performed to establish concentration calibration curves, estimate the limit of detection of each gas in air, and delineate the individual behavior of each gas as a function of concentration. Emission spectra of Ar as a function of concentration and the corresponding calibration plot of peak intensity versus concentration for three Ar emission lines are shown in Figure 4. Immediately apparent is the variation in the baseline intensity of the spectra. The differences in emission intensity peak heights and baseline were used for the calibration curves to correct for this variation. The resulting plot exhibits a linear dependence of Ar with respect to concentration, with an estimated limit of detection (LOD) of 1% based on a signal-to-noise ratio (SNR) of 3. This LOD is illustrated in Figure 7, wherein the 763 nm peak of atmospheric Ar (1.29% atmospheric abundance) is just detectable within the system SNR. It is noted here that the peak emission intensities of the 742 and 746 nm lines of atmospheric N varied by only $\pm 4\%$, indicating that the actual levels of Ar in the laser interaction zone were being diluted by ambient air due to the open nature of the sample introduction. Thus, values and trends reported here are for illustrative purposes and serve as an upper estimate of the actual LOD obtainable by the system.

Figure 4. (left) Red emission spectra of Ar at varying concentrations in nitrogen gas; (right) a calibration plot of peak intensity versus Ar concentration is linear.

Shown in Figure 5 are the S emission lines of SF_6 as a function of concentration and the corresponding calibration plot of peak intensity versus concentration for three S and four F baseline-corrected emission lines. A nonlinear dependence of peak height on concentration is observed. The leveling effect of higher concentrations suggests that self absorption of the emission line is occurring. This is more evident in the corresponding data for R134a (Figure 6), wherein leveling is observed

for all nonzero concentrations evaluated. Thus, the linear portion of the concentration curve is not observed under the current experimental conditions. The LOD of each gas is approximated from the slope of the emission intensity line between the lowest concentration of gas and air, yielding 200 parts per million (ppm) R134a based on the H emission line, and 300 ppm SF_6 based on the 703-nm F line. These are presented as likely upper limits given the constraints of the experimental conditions reported.

Figure 5. (left) Visible emission spectra of the sulfur lines of SF_6 at varying concentrations in nitrogen gas; (right) the variations in peak intensities of the sulfur and fluorine lines (not shown) yield a nonlinear calibration curve indicative of saturation at higher concentrations.

Figure 6. (left) Red emission spectra of the fluorine and hydrogen lines of R134a at varying concentrations in nitrogen gas; (right) the variations in peak intensities of these lines yield a nonlinear calibration curve indicative of saturation at all investigated concentrations.

Intensity Calibrations

The spectra presented in Figures 4 through 6 illustrate the variability in baseline intensity, and thus the variability in absolute emission line peak heights. A series of at least ten spectra was collected for each concentration of gas recorded during this study. The variance in peak height from laser shot to laser shot was observed to be as much as 80%, making a calculation of concentration in air highly unreliable for a single-shot spectrum. Sources of this variability include shot-to-shot laser power fluctuations, variability in the location of the plasma relative to the collection optical fiber, and variations in plasma parameters influenced by the chemical composition of the sample being probed (Palleschi, 2006). Means of correcting for variations in sample intensity were investigated.

For solid or liquid LIBS samples, a known quantity of a calibration standard is mixed with the sample. While this is not achievable for open atmospheric air sampling, we sought to emulate this approach using components of air as the calibration source. Although N_2 and O_2 were obvious choices, the possible employment of the LIBS system for detection and measurement of targets containing N or O made each of these elements a variable rather than a standard. Argon is ubiquitous in the atmosphere at 1.2% abundance, and is not found in stable molecular species, thus making it an invariant marker. The emission spectrum of ambient air did yield a peak for natural abundance Ar at 763 nm (Figure 7). However, as noted above, the peak intensity was at the SNR limit of the system, making it unreliable as a calibration peak.

Figure 7. Emission spectrum showing the most intense Ar peak (763 nm) in ambient air. While present, the low SNR of the peak precludes its use as an intensity calibration standard.

A second means of providing intensity calibration was the introduction of a stationary platinum (Pt) wire at the focal point of the laser. In this way, both air excitation and Pt excitation via localized metal vaporization occurred within the same laser point. The emission spectrum of Pt was comprised of multiple peaks between 200 and 300 nm, well removed from the “fingerprint” peaks of most organic and nonmetallic species (400–800 nm). A portion of the Pt spectrum is shown in Figure 8. This technique proved successful for correcting Ar peak intensities. However, Figure 8 demonstrates the failing of this technique at higher concentrations for the fluorinated compounds. For example, as the concentration of SF_6 increased, the emission intensity of Pt decreased by a factor of 5 and did not exhibit the same trend in peak intensity variability as the F and S peaks of SF_6 . It appears that passivation of the Pt occurred at the concentrations investigated.

Figure 8. UV emission spectra of laser plasma-ablated Pt wire in the presence of SF_6 . The inset demonstrates the passivation effect of added SF_6 on the Pt emission intensity.

Alternate Plasma Sources

Plasma sources other than the Kigre Nd:YAG laser were investigated in an effort to further simplify the system in terms of required power supplies and drivers, and to provide a plasma at a fraction of the cost of the laser-based system. Plasmas generated by dielectric breakdown of air in the presence of a high electric field between two electrodes were investigated. Platinum electrodes were used to provide both a fairly inert electrode material and to directly introduce ablated Pt vapor into the plasma as an emission intensity calibration source. The first source tested was an EMCO Q20-5 DC-to-high voltage DC module, approximately 1 cm³ in volume, and requiring only an input of 1–10 V to produce up to a 10,000-V output at very low current. It should be noted that, for the purposes of this experiment, the Q20-5 units were driven well beyond their specified outputs to produce a plasma spark. The second source was a handheld lead-zirconium-titanate piezoelectric fire-starter, producing tens of thousands of output volts when mechanically activated. The interest in the latter source was due to the possibility of utilizing the shaft motion of the sample air pump as an actuator for the plasma without any additional power requirements for the system. The feasibility of both plasma sources was demonstrated in the production of Ar atomic emission, as shown in Figure 9. The inset illustrates the system configuration, with the spark gap between two Pt electrodes aligned along the optical axis of the collection fiber optic. The apparently weak signal and continuum-free spectra depicted are due to a timing mismatch between the CCD signal collection system in free-run

Figure 9. Plasma emission spectra of Ar and air using alternative plasma ignition sources

mode, and the timing of the sparks, which were at random intervals. Introduction of synchronization between the sparks and signal collection would be required before a quantitative evaluation of these spark sources could be performed. It should be noted that introduction of either SF₆ or R134a results in the complete quenching of the high voltage-induced spark. This is due to the high threshold for dielectric breakdown, and thus the strong electrical insulating properties, of these gases and perfluorinated chemicals in general.

Conclusion

The efficacy of LIBS laser plasma vapor detection for the detection of fluorinated gases in ambient air has been demonstrated. Both ppm-level detection and compound identification are feasible with this system. A means of calibrating the system was investigated, promising results for the Pt-standard technique at lower concentrations of passivating materials such as the fluorinated compounds. The system has been proven to a sufficient point to satisfy the field diagnostic needs for NNSA NA-22 ground truth monitoring at chemical release events at the NTS. A field prototype can be produced from the laboratory prototype demonstrated herein.

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CONCEALED DIRECTIONAL DETECTOR WITH RADIONUCLIDE IDENTIFICATION

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The Concealed Directional Detector (CDD) was originally developed in FY 2007 (Maurer, 2008) as a first-generation advanced gamma-radiation detection prototype incorporating a low-profile footprint and directional capability. The second generation of the CDD prototype was developed in FY 2008 and incorporates several advanced engineering features, including miniaturization of the electronics and integration of a lanthanum bromide (LaBr₃) high-resolution radionuclide identification system. The new prototype unit will detect, direct, track, investigate, and identify an anomalous moving radiation source. This integrated system provides emergency responders and law enforcement officials with a new, innovative tool for radiological surveillance, interception, and adjudication at high-profile special events.

Background

The current approach employed by nuclear emergency responders conducting a radiological search is to utilize gamma radiation detectors concealed in backpacks. Since the terrorist attacks on September 11, 2001, security officials at large-scale events do not allow visitors to bring backpacks past security checkpoints due to ongoing concerns with terrorism. Consequently, the use of backpack detectors by nuclear emergency response personnel has been significantly curtailed except when working directly with law enforcement. In addition, backpack detectors in use today employ single-detector elements and, as such, are not capable of providing directional information. The direction, tracking, and localization of a moving radiation source are increasingly important capabilities for assisting law enforcement in the adjudication of anomalous radiation alarms in large crowds. In an effort to remedy this situation, a first-generation radiological search instrument prototype was developed in FY 2007 (Maurer, 2008), and was further advanced in the second-generation prototype in FY 2008.

Project

In FY 2008, the project goal was to enhance the original engineering prototype with novel features, including radionuclide identification. Other enhancements included significantly reducing the footprint of the electronics, adding a Bluetooth interface, utilizing a cell phone computer, and incorporating radionuclide identification.

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Front and rear views of the FY 2007 prototype are shown in Figure 1. The unit employs four gamma-radiation detectors affixed to a belt and oriented at 45, 135, 225, and 315 degrees and worn by the emergency responder. The belt provides a convenient conduit for routing the detector bias and signal wires to the signal processing electronics and power supply in an enclosure on the rear of the belt.

Figure 1. Concealed directional detector showing front (left) and back (right) views

The four detectors and the inherent self-shielding provided by the emergency responder's body mass allows the individual detectors to intercept gamma radiation with varying intensities from a source located at an angle relative to the frame of reference. The following equations are used to calculate the direction of the radiation source:

$$X = (D_1 - D_3), \quad (1)$$

$$Y = (D_4 - D_2), \text{ and} \quad (2)$$

$$\Theta = \text{Arctan}(Y, X). \quad (3)$$

The detector count rates (in counts per second) are denoted by D_1 through D_4 , and the angle of the radiation source relative to the emergency responder is given by Θ . Once a trigger level, defined by a preselected number of standard deviations above background, is attained in one of the detectors, the angle is then calculated. After being activated, the direction of the radiation source is updated once every second.

The identification unit is installed in the rear enclosure and is used by the first responder to assist law enforcement in the adjudication of radiation alarms. The integrated detection/identification system will allow nuclear emergency responders working with law enforcement to detect, direct, track, and adjudicate radiation alarms caused by a moving radiation source.

Results

The advanced CDD unit employs four 2.5" × 4" × 0.5" cesium iodide (CsI) scintillation detectors made by Scionix. Each detector is coupled to a photodiode and hermetically sealed in a $\frac{1}{16}$ -in.-thick aluminum enclosure. The CsI/photodiode detector combination was selected for its low power requirements, compact form factor, and low bias voltage. The photodiode operates with a bias voltage from 35 to 70 volts DC. The detector size selected provides the same sensitivity as the current commercially available backpack systems. (This project also investigated the use of plastic scintillators. However, the CsI detectors are preferred, due to their higher sensitivity to energetic gammas.)

The signal and power leads from the CsI detectors are directed along the belt to the electronics control unit housed at the rear of the belt. A compact electronics package, developed by the Remote Sensing Laboratory (RSL) was utilized, which includes a battery, low-voltage power supply, a microprocessor operating Linux, signal processing, and a Bluetooth transmitter. Figure 2 shows the compact electronics unit.

Figure 2. RSL compact electronics unit in the CDD

A Bluetooth module integrated in the electronic unit pushes the data from the detectors to a cell phone having an onboard computer. The computer records the data and performs the direction calculation. The result of the calculation is displayed via an arrow indicator on the cell phone screen.

The CDD utilizes an AT&T Tilt Model HTC 8900 Smart Phone with Bluetooth, and employs software previously developed by RSL for data acquisition, analysis, and display configured for the CDD. The computer employs the Windows Mobile 6 operating system. The smart phone and an example of the directional display are shown in Figure 3.

Figure 3. AT&T Smart Phone Model HTC 8900 (top and bottom left) and screen capture of display showing the directional indicator (bottom right)

Radionuclide identification is performed with a LaBr_3 high-resolution scintillation detector from Saint-Gobain engineered into an integrated compact unit by Bridgeport Instruments. The LaBr_3 detector measures $1" \times 1"$ and is coupled to a photomultiplier. The dimension of the identification unit is approximately $1" \times 1" \times 4.5"$. The LaBr_3 detector was selected for its superior resolution of 3–4% for cesium-137 (661 keV) as compared to sodium iodide (6–7%), which is commonly used in commercial portable radioisotope identification units. The LaBr_3 identification unit with associated electronics is shown in Figure 4.

Figure 4. Compact, high-resolution LaBr_3 identification unit and electronics

The LaBr_3 identification unit was developed for the project but did not arrive in time to be integrated into the CDD at the time of this writing. The analysis software incorporates spectroscopy algorithms that will be utilized with the identification unit.

Conclusion

The first-generation prototype CDD developed in FY 2007 has been advanced by engineering compact, low-power electronics and a high-resolution identification unit. The unit represents the second generation of the CDD low-profile detection system for nuclear emergency responders and law enforcement personnel. The CDD will detect, direct, track, and adjudicate radiation alarms caused by a moving radiation source.

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FREQUENCY-MODULATED DETECTION OF PHOSPHORESCENCE ON SURFACES

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A frequency domain fluorescence detection method established in the laboratory was adapted for the measure of solid material fluorescence lifetimes under field conditions. A microcontroller-based circuit board was designed and fabricated, providing a modulated excitation light source, an amplified photo receiver, and data capture and control logic functions. The board was packaged to run on battery power, and was then used to capture frequency domain fluorescence decay waveforms from samples with a known lifetime. The observed waveforms are in good agreement with prior work, and future system optimization paths have been identified.

Background

The use of frequency-modulated fluorescence in the identification of solid materials is a technique developed under an FY 2007 SDRD effort (Di Benedetto, 2008). In that work, a specific class of materials was identified through a measure of the lifetime of the fluorescence (or phosphorescence) light induced by illuminating the materials with modulated light. Fluorescence is defined here as optical emission resulting from optical excitation. Further treatment of the scientific basis of the fluorescence process and the developed techniques for solid material fluorescence measurement can be found in the FY 2007 SDRD final report. The end result of that work proved the frequency domain fluorescence technique to be viable for use in the identification of solid materials in the presence of ambient light and secondary sources of fluorescence. The challenge leading to the current project was to create a new fluorescence measurement system that would utilize many of the same methods, but be portable and adapted for use under field conditions. The field portability requirement dictated that the system operate on self-contained power, be housed in a compact package, and support the ability to operate in high ambient light conditions. The goal of this year's project was to design and fabricate such a system, and test it using known fluorescent samples to verify functionality.

Project

Design

The system design is shown in the block diagram in Figure 1, where the microcontroller provides a timing reference for driving excitation light from the LED. With the battery-powered LED directed

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at a fluorescent sample, the incoming fluorescence light is amplified and then digitized in the analog-to-digital converter (ADC). The microcontroller then compares the phase of the driving and received waveforms and either stores or reads out the result.

Figure 1. Block diagram showing the fluorescence measurement system

The first step in the design of the system was to address a method for modulating the excitation light source, a 365-nm UV LED. The desire for simplified circuitry dictated that a square wave be used to generate a simple power on/off function. However, the challenge proved to be in maintaining bandwidth into the hundreds of kilohertz range while driving the LED with 4 to 5 V at 0.5 A. Several iterations of drive circuitry had to be developed and tested to overcome the basic limitation of rapidly switching large amounts of power on and off. An operational amplifier (op-amp)–driven JFET (junction gate field-effect transistor) circuit capable of generating an acceptable square wave in

the LED light output up to around 200 kHz was eventually developed. To tailor the light output to a variety of field measurement conditions, a current-adjustment capability was added to drive the LED at five discrete current outputs ranging from 100 to 500 mA.

The second part of the system development involved the capture and amplification of weak optical signals coming from a fluorescent material. The initial effort focused on the selection of an appropriate photodiode; both amplified and nonamplified variants were considered. In general, the amplified photodiodes, also known as hybrid photodiodes, did not perform well in this application due to bandwidth limitations at higher frequencies. In the end, an OSI PIN5-DI nonamplified photodiode was selected for its optimal combination of fast response and sufficient sensitivity, two generally competing attributes in any photodiode detector. Due to the lack of an integrated amplifier in the PIN5-DI, a first-stage amplifier had to be selected that could feed the signal to either a second-stage amplifier or directly to the ADC. After considerable additional testing and research, it was determined that a single-stage transimpedance amplifier circuit would be sufficient, utilizing an OPA657 op-amp. An adjustment to select between five different gain settings was incorporated into the circuit to account for varying intensities of incoming fluorescence light. The circuit was capable of amplifying photodiode optical signals in the range of 10 nA to 1 mA up to a level of 1.2 V going into the ADC.

The final part of the system design consisted of integrating the power and control functions to support the MSP430 chip. The 16-bit chip specifications include 48 kB of flash memory, 2 kB of RAM, and three onboard Sigma-Delta ADCs. To help mitigate cross talk issues between the driver, receiver, and logic portions of the circuit board, the ground planes and power supplies were isolated. The final design utilized a six-layer board with a single 14-V battery powering the LED driver circuit and MSP430 microcontroller, and two 9-V batteries powering the receiver circuit.

Figure 2. Completed prototype assembly showing (left) circuit board and (right) base-mounted 14-V battery and detector head

With a completed board prototype fabricated, it was finally necessary to address the issue of controller firmware. The firmware was written to make use of four momentary push buttons mounted on the card to drive interrupts to ports on the MSP430, thus allowing the manual selection of three important functional parameters: LED drive frequency, LED output power, and the amplifier gain on the photodiode. The firmware was set up to cycle through a predetermined set of values for each parameter in a closed loop. In addition, the firmware had to control the input onto the ADC and write waveforms coming from the ADC to the flash memory onboard the MSP430 chip.

Data Collection and Analysis

The completed prototype was used to collect data on two fluorescent samples with known lifetimes, coumarin 522B and UO_2F_2 . The coumarin sample has a very short lifetime in the nanosecond range, which places its phase shift well beyond the frequency range of this instrument as configured. The UO_2F_2 , on the other hand, has a relatively slow lifetime on the order of several hundred microseconds. Since fluorescence lifetime is the inverse of the frequency about which the phase shift is centered, the slow component of the UO_2F_2 can be expected to shift in phase from zero to ~ 90 degrees centered about a frequency of a few hundred hertz. Figure 3 shows a plot of the fluorescence light emission from the two samples as driven by the UV LED with square wave intensity modulation. The light intensity was sampled at 128 points over eight waveform periods, repeated at nine different frequencies. Note that only the first three waveform periods for each frequency are shown in the figure. When compared with the fast-fluorescence coumarin 522B, the phase shift in the UO_2F_2 is evident in the slowing rise and decay times of the waveform with increasing frequency (Figure 3). The roll-off observed in the coumarin 522B waveforms at the higher frequencies is a result of the bandwidth limitations of the system, and does not represent a change in response to the dye (Di Benedetto, 2008).

Also evident is the associated phenomenon of demodulation in the slow phosphorescence. As the drive frequency begins to approach and then exceed the inverse of the sample lifetime, so the amplitude of the phosphorescence light modulation decreases and trends towards DC light. This results from the fact that in this frequency range a percentage of the fluorophores excited at the peak of the excitation continue to emit past the excitation minimum (Lakowicz, 2006).

Figure 3. Phase and amplitude signals as captured by the onboard microcontroller for (a) coumarin 522B fast-fluorescence sample and (b) UO_2F_2 sample

Suggestions for Future Work

Although the data collected with the handheld instrument showed in a qualitative sense the difference in lifetime between the two samples, a waveform analysis algorithm will be needed if actual lifetime values are to be extracted from the waveforms. This analysis algorithm would be capable of discerning multiple lifetime components in a single sample, and would require translation into firmware sufficiently compact to function on the MSP430 processor. In addition, a digital lock-in amplifier may need to be incorporated as a second-stage amp, if warranted by high optical noise or detector saturation. Finally, the PC board would need to be redesigned to facilitate a smaller size instrument with lower power consumption.

Conclusion

A compact, portable test apparatus was designed and assembled to demonstrate a frequency-modulation technique for the measurement of phosphorescence lifetimes. The device was tested on samples used in the prior-year SDRD work, and gave qualitative indications of sample lifetime consistent with both benchtop instrumentation as well as other time-domain measurement techniques. Lastly, the team also identified improvements that could be made to the device packaging and recommended further developments of the device firmware and functionality.

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NANOSECOND SINGLE-PHOTON DETECTION OF DIFFRACTED X-RAYS

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We have designed, built, and tested a novel, fast, single-photon detector using microchannel plate (MCP) technology. Testing of the nanosecond single-photon detector (NSPD) showed it was capable of observing 0.71 Å x-ray quanta within a 1.7-ns gate time. We have also developed a single-pulse x-ray diffraction (XRD) diagnostic, which was validated with high explosives-driven experiments on tin, zirconium, and aluminum at the Special Technologies Laboratory (STL) Boom Box.

Background

The purpose of this SDRD project was to develop a phase transition diagnostic for shocked polycrystalline materials. Because XRD provides a direct correlation between the diffracted image and the solid-state structure of the material under investigation, this diagnostic should be extremely useful to the Stockpile Stewardship community. The scope of work for this project was two-fold:

1. Develop a new, fast, single-photon MCP detector for use in conjunction with single-pulse XRD, and
2. Perform experiments with shocked polycrystalline metals using single-pulse XRD as the primary diagnostic using the high-explosives drive at the STL Boom Box.

The scientific concept for the use of an MCP as an x-ray detector in the 17–22 keV energy range is based on measurements by Rochau (2006) at Brookhaven National Laboratory's beam line X8A at the National Synchrotron Light Source. Sensitivities were measured for x-ray energies ranging from 100 eV to 6 keV, and the results were consistent with a leaded-glass photocathode and not a gold photocathode. This opened the possibility for the use of gated MCP detectors in the 17–22 keV energy range, a region where the Pb L-series photoabsorption results in high quantum efficiency for a thin MCP. Figure 1 shows the high probability for an Auger electron or photoelectron reaching the MCP channel, initiating the cascade.

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Figure 1. Continuous slowing down approximation shows a high probability for Pb Auger electrons reaching a channel, thus starting the electron cascade

Project

NSPD Design and Laboratory Testing

Design work and drawings for the NSPD were completed during the FY 2007 SDRD project (Morgan, "Nanosecond," 2008). The FY 2007 NSPD assembly drawing is shown in Figure 2. In early FY 2008, prototype NSPD parts were procured and assembly began. A leak was discovered in the fiber-optic faceplate, but the original order included a replacement faceplate, which held vacuum at 2×10^{-6} torr. The MCP coatings that were applied by Burle were observed to have numerous defects, and applied voltages were below the desired values for the experiments to prevent arcing.

The initial tests of the NSPD were performed in the laboratory with the setup shown in Figure 3. A molybdenum anode produced bremsstrahlung and characteristic 0.71- $\text{\AA}$ line emission, and a slit was placed in front of the source. A silicon (220) single crystal was placed 6 in. from the slit at a Bragg angle of 10.7 degrees and served as a spectrometer, so the $\text{K-}\alpha_1$ and $\text{K-}\alpha_2$ lines could be resolved on the detector. DC bias voltages ranging from -1000 VDC to -1200 VDC were applied to the MCP input, and +800 VDC was applied to the phosphor. The CCD camera was gated to record an image during the 40-ns x-ray pulse generated by the x-ray source. The 0.709- $\text{\AA}$ and -0.713- $\text{\AA}$ $\text{K-}\alpha_1$ and $\text{K-}\alpha_2$ lines were resolved by the detector, as shown in Figure 4. Also, the individual photons that form these lines are also apparent.

To achieve fast gating of the NSPD, a short, -1000-V, 4-ns rectangular pulse was applied to the input of the MCP. The pulse was transmitted through the MCP transmission line, attenuated by 60 dB, and recorded by a 1-MHz bandwidth digitizer. The digitizer simultaneously recorded the 40-ns x-ray pulse as shown in Figure 5. Because the gain of the MCP goes approximately as V^{11} ,

Figure 2. FY 2007 assembly drawing of the NSPD (Morgan, 2008)

Figure 3. Laboratory setup for NSPD testing. The MCP bias is fed into the detector at the top, and the phosphor voltage is applied on the left. During pulsed operation, the output on the bottom of the NSPD was used to record the waveform.

and because the pulse waveform is rectangular, the effective response gate of the detector is about 2 ns, which is considerably shorter than the pulse width. The effective time response is also reduced by the transit time of electrons in the MCP, so we estimate the overall time response of the detector in the experiment shown in Figure 5 to be about 1.7 ns. Only a few 0.71-Å x-rays are observed by the detector under these conditions.

Figure 4. Molybdenum K- α spectrum observed with the NSPD in DC mode

Figure 5. Nanosecond single photon detection

Experiments at the STL Boom Box

Several improvements were made to the single-pulse XRD system in FY 2008. The improved design is shown in Figure 6. The FY 2007 prototype single-pulse XRD system used a P-43 phosphor coupled to a CCD camera by a coherent fiber-optic bundle as the x-ray detector. For FY 2008, a large-format x-ray imaging system was developed that used a Fujifilm BAS-MS image plate. The image plate was kept light-tight in a cylindrically bent re-entrant fixture. The large format allowed the entire XRD pattern to be recorded in the image, including the direct beam, which was attenuated by a tungsten beam stop. Multiple layers of high-density polyethylene were used as blast protection for the x-ray window. The tungsten shielding was increased on the x-ray source to eliminate all x-rays except through the pinhole. A new design was developed for the 1.5-mm-diameter tungsten pinhole collimator with improved shielding that limited the spot size on the sample and the beam divergence to ~ 1 degree. Also, a new wax fixture was developed that aligned the collimator with the target package at a fixed 12.2-degree angle.

Figure 6. The new design of the single-pulse XRD features a large-format imaging system, increased target shielding, a redesigned 1.5-mm pinhole collimator, and an improved alignment fixture

The system was transported from Los Alamos Operations to the Boom Box at STL and set up as shown in Figure 7. The high-voltage pulser used to generate the x-rays is a 37-stage Marx with 8.1 nF per stage, charged to -30 kV. The Marx develops an output pulse of approximately 400 kV, which travels through a DS-2158 coaxial transmission line to a needle-and-washer x-ray diode, generating line and bremsstrahlung x-ray emission in a 40-ns pulse. The anode spot size is ~ 1.5 mm, which is the same size as the pinhole. The x-ray diode was placed inside a blast containment tube to allow the placement of the x-ray source close to the target package. Experimental timing of the shock breakout on the back surface of the vitreous carbon window was measured by photonic Doppler velocimetry (PDV), which also allowed an estimate of the shock-loaded interface pressure of the target sample. An electromagnetic pickup near the x-ray diode was used to determine the timing of the x-ray pulse with respect to the PDV shock breakout signal.

Figure 7. Setup of the single-pulse XRD system at the STL Boom Box shows (a) a close-up of the setup inside the vessel, (b) the vessel, and (c) the Marx bank

The first experiments were performed with fine-grained aluminum samples with a 0.4-mm-thick vitreous carbon back window, shocked with Detasheet high explosives. The experimental static and dynamic images are shown in Figure 8. New software, developed to integrate the Debye rings, includes the correction for the small movement of the sample interface. From the results of the integration shown in Figure 9, we observe that the aluminum diffraction lines have shifted to larger scattering angles, with larger shifts occurring for larger scattering angles. The angular shift for the diffraction lines of aluminum are compared with the shift expected for 8.6% isotropic volume compression of the

face-centered-cubic (fcc) aluminum lattice in Figure 10. From the bulk modulus of aluminum, 8.6% volume compression with no temperature increase corresponds to an applied pressure of 60 kbar. This compares to the WONDY code calculation of 71 kbar generated using PDV data.

Figure 8. Static and dynamic images of shocked aluminum. The crystal grains remain in the fcc structure.

Figure 9. Integration of the Debye rings from static and dynamic fcc aluminum experiments

Figure 10. Compression of the aluminum fcc lattice

In FY 2007, our smaller field-of-view imaging system was used to observe shock-loaded tin. For these experiments, we observed complete disappearance of the strong (311) and (400) diffraction lines, which were replaced in the dynamic image with a broad interference peak that we have associated with a liquid or amorphous dynamic phase. This year, experiments were conducted to re-examine the phase transition we observed for shocked tin. Also, we attempted to observe the phase transitions that are assumed to occur in shocked zirconium. For these experiments, a thin, 0.1-mm-thick tin or zirconium foil was placed between the aluminum buffer and the vitreous carbon back window.

A shocked tin experiment was conducted with 0.71-Å x-rays and Detasheet high explosives. Figure 11 shows experimental results. The large grain size in the static image is consistent with 0.1-mm nominal grain size for tin. As with our FY 2007 experiments, a broad interference peak or “halo” appears in the dynamic image at a scattering angle consistent with observations of liquid tin diffraction by Vahvselkä (1978). Additionally, Debye rings are observed in the dynamic image associated with solid structures, which do not appear to be the static tin β -phase. These Debye rings indicate a much finer grain tin structure, and the rings were not observed in our FY 2007 experiment because of the limited field-of-view. Further analysis would be required to determine this structure.

We conducted two dynamic shock experiments on zirconium this year with 0.71-Å x-rays and PBX-9501 high explosives. Both zirconium experiments gave essentially the same result. The results of a shock-loaded zirconium experiment are shown in Figure 12. Similar to tin, a broad interference peak or halo is observed in the diffraction pattern. This is indicative of a shocked zirconium liquid or amorphous solid state, which is interesting because the extremely high melting point of zirconium precludes the possibility of what is traditionally considered melt. Also observed in the dynamic image is a new, undetermined solid phase.

Figure 11. Static and dynamic tin diffraction patterns. The dynamic image shows evidence for a mixed liquid or amorphous and solid-state phase, as well as reduction of the grain size.

Figure 12. Static and dynamic XRD images of zirconium. As with tin, the dynamic image shows evidence of a mixed phase.

Conclusion

We have developed the NSPD, and tests demonstrated its capability for single-photon detection with gating <2 ns. We have also performed high explosives-driven experiments at the STL Boom Box to investigate the dynamic material properties of shocked aluminum, tin, and zirconium. An article describing the recent single-pulse XRD system has been published in *Review of Scientific Instruments* (Morgan, “Real-time,” 2008).

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RADOPTIC SENSORS (PHASE 2)

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Substantial progress was made in demonstrating the GaN-based RadOptic sensor in the laboratory. For the optical probe, we completed procurement, installation, and testing of a tunable, single-mode, single-frequency, continuous wave (CW) 405-nm semiconductor laser system. The InGaN RadOptic material was designed, acquired, and tested. A functional interferometer was built and GaN and InGaN films were tested with the interferometer. With proper balancing, the interferometer routinely operated at a contrast ratio of >96%. We have conducted experiments using a femtosecond laser, which, when operated at 200 nm, acted as a pump beam for the GaN and InGaN materials. The experiment results were inconclusive; however, we expect the GaN sensor would be successful if the probe wavelength can reach 360 to 366 nm to better match the GaN band gap. A number of improvements to the experiment are recommended in this report.

Background

Ultrafast, high-dynamic-range, low-cost x-ray and optical detectors are in demand by the national laboratories for high-energy density physics and for inertia-confined laser fusion research. RadOptic sensors are a new class of fast semiconductor detectors, relying on x-ray-induced refractive index change as the detection mechanism. When energetic ionizing radiation is absorbed by the semiconductor material, electron-hole (e-h) pair productions alter the spectral refractive index, which changes the phase of an optical probe beam. Laser interferometry is then used to read out the phase variation. The difficulties in detecting broadband x-rays thus become a problem of optical interferometric detection.

Our goal was to create GaN-based RadOptic sensors to meet such needs (Sun, 2008) as an improvement over GaAs sensors (Vernon, 2006). Because the GaN material has a very fast relaxation time, below 1 ps, and may go as low as 600 fs (Lowry, 2005; Nakamura, 2000; Stanton, 2001; Matulionis, 2002), it is well-suited for high bandwidth measurement and synchronization control at the National Ignition Facility.

The GaN material as a sensing medium is a wide-band-gap semiconductor with an absorption edge in the 360 to 366 nm range. In modified material such as InGaN, the band gap can be tuned to a lower energy (longer wavelength). Therefore, we employed two approaches of developing and

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demonstrating the GaN sensor: (1) tune the probe laser wavelength, or (2) tune the material band gap with a different component ratio for InGaN alloy. The goal was to demonstrate a high-performance, ultrafast, and cost-effective GaN RadOptic sensor.

Project

The GaN RadOptic sensor is a complex experiment to demonstrate. It requires two lasers at two different wavelengths: a pump laser with a shorter wavelength that is absorbed by the GaN/InGaN materials, and a probe laser with a wavelength that matches the band edge transition region. The pump laser induces the refractive index change in the GaN materials. The phase shift experienced by the probe laser beam traversing the GaN material is detected by an interferometer. For the GaN material the band gap is ~ 3.44 eV, equivalent to a laser wavelength of ~ 360 nm.

For the pump laser we selected a Ti:Sapphire laser with an amplifier, followed by a fourth-harmonic generator. The laser can deliver a ~ 10 -mJ pulse with a pulse width of ~ 200 fs. Because the interferometer readout was built on a movable cart, we were able to conduct the experiment in the Short-Pulse Laser lab at National Security Technologies', LLC, (NSTec's) Livermore Operations (LO).

For our work the probe laser needed to operate in single mode due to the interferometric detection requirement. The probe laser's wavelength could be based on pump laser intensity, and this requirement is still a subject of research. There are two approaches to calculate the refractive index change for GaN material. The free electron Drude model, as often used in terahertz detection, indicates a wider selection of probe wavelengths. However, a more complex model considering exciton generation dynamics would require the probe laser wavelength to be near the band gap, within a small wavelength range of 1–3 nm.

The probe laser we acquired was a custom-made, CW, GaN semiconductor laser with a wavelength of 405 nm (tunable to within 2 nm) and an emission power of 35 mW. We experienced a significant delay in procuring the laser and having it installed, as the manufacturer, Sacher Laser Technique, is located in a foreign country; this extended the approval process. However, the laser performed quite well. Figure 1 shows the laser head, the laser emission spectrum, and the laser beam profile. The single-mode, single-frequency modulation was further confirmed using a scanning Fabry-Perot interferometer and a Michelson interferometer with an arm length difference of 50 cm.

The pump/probe experiment design, construction, and fine tuning required considerable effort. Figure 2 shows the probe laser experimental setup. The probe section, including the CW 405-nm laser, two alignment lasers, the interferometer, the detectors, and other instruments, are all densely assembled on a cart with a heavy optics breadboard. The mobile cart could be moved to any laser source lab for experiments.

Figure 1. The 405-nm, CW, GaN laser (left), its emission spectrum (upper right), and beam profile (lower right)

Figure 2. The laser probe apparatus, set up on a cart

Our experiment used a Michelson interferometer configuration. Figure 3 shows a simplified optics layout for this. We also set up several other configurations that used Mach-Zehnder and Sagnac interferometers. However, the Michelson interferometer offered ease of alignment with clear signature. It also promised a pulse-length measurement with echo confirmation. Alignment was carefully done to achieve 96% contrast ratio. The residual leakage was due to optics depolarization at 405 nm. Nevertheless, we can observe the fringe change using an oscilloscope directly down to 10^{-4} radians or better. The time-integrated observation was not attempted, because our primary interest was to observe the fringe change in a single-shot event.

Figure 3. Simplified optics layout, showing the Michelson interferometer

Figure 4 shows a typical interference fringe scan. The smoothness indicates the stability of optics mounting is practical and enabling. To simultaneously observe the near-DC fringe, and the dynamic fringe variation at faster time scale, three detectors with different response speeds were used at the same time. For this purpose, the interferometer output beam was split into three.

We also searched for an optimized material whose band gap matched the 405-nm laser wavelength. Based on extensive literature studies and interactions with the vendor, we chose InGaN thin film as a second candidate material for the RadOptic sensor. The indium composition for $\text{In}_x\text{Ga}_{1-x}\text{As}$ thin film was selected as $x = 10\%$. The spectral transmission of the $\text{In}_{0.1}\text{Ga}_{0.9}\text{As}$ thin film was tested, and the measurement results are shown in Figure 5. The transmission is near zero for wavelengths shorter than 362 nm, and it approaches 66% at 405 nm, with more headroom in the longer wavelength region. Thus, the band-gap edge did shift to longer wavelength compared with GaN-only thin film.

However, the band-gap edge slope widened an equivalent amount, ~ 40 nm. The measurements were taken for two polarizations. Slightly lower transmission is observed for 45-degree polarization input. However, the overall transmission behaviors are similar, as Figure 5 shows.

Figure 4. An example of the Michelson interferometer fringe pattern, taken when the interferometer is illuminated with the 405-nm CW probe laser

We conducted the pump/probe experiment at the LO Short-pulse Laser Laboratory using our interferometer and an ultrafast laser as the pump. The laser emits ~ 10 -mJ laser pulses at 200 nm, with a pulse width of ~ 200 fs.

Two kinds of GaN-based materials were used in the experiment: (1) A 7- μm -thick GaN film grown on sapphire with double-sided polished surfaces, and (2) a 5- μm -thick InGaN film grown on GaN film on a sapphire substrate. GaN and InGaN samples were inserted in both of the interferometer arms; this balanced the arm intensity and preserved the fringe contrast ratio. An additional green alignment laser was used to ensure the pump spot was overlapping with the interferometer arm laser path.

Figure 5. Transmission measurements of the $\text{In}_{0.1}\text{Ga}_{0.9}\text{As}$ thin film material, showing (upper) a scan from 200 to 800 nm and (lower) a detail from 360 to 500 nm

The experiment results were inconclusive in that we saw no definitive change in interferometer readout at the main detector. It is possible that:

1. The 405-nm wavelength of the probe laser is still too far away from the true band edge.
2. Our main detector, a 12-GHz photodiode, is still too slow for the 120-fs laser pulsewidth. The detector might also be too noisy for the dynamic intensity change.

However, according to our calculations and some of the published results, we believe we are close to an observation of the refractive index change induced by a single-shot pump laser. We suggest the following improvements be tried first in future work:

1. Use of a tunable laser to operate the experiment with scanning wavelength.
2. Use of a fixed laser with wavelength close to the band gap. A commercially available laser operating at 372 nm is a likely candidate.

Conclusion

We have made a systematic first step toward demonstrating the GaN-based RadOptic sensor, although experiments performed on GaN-based materials were inconclusive. Several improvements to the experiment setup are recommended. With these improvements, it is likely that one can observe the refractive index change induced by a single-shot pump laser to give a clear demonstration of the GaN-based RadOptic sensor.

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FIELD-TESTING A GAMMA-RAY TELESCOPE FOR SEARCH AND CONSEQUENCE MANAGEMENT MISSIONS

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Gamma-ray imaging is potentially useful for Homeland Security applications, since it would allow a searcher to survey a scene. The ability to pinpoint hidden sources in a single measurement and from a single vantage point affords the emergency responder an opportunity to not only maintain safe distances but to expedite the search mission. The National Aeronautics and Space Administration (NASA) has designed, built, and put into orbit a wide field of view (FOV) gamma-ray telescope to survey the sky for extragalactic gamma-ray bursts. This telescope, the Burst Alert Telescope (BAT), consists of a coded aperture and a focal-plane array of 32,768 CdZnTe (CZT) detectors. The NASA Goddard Space Flight Center (GSFC) maintains a smaller engineering model of this instrument, the Goddard Rad Source Imager (GRSI), on the ground. Collaboration between National Security Technologies, LLC (NSTec), and GSFC was initiated to evaluate the effectiveness of this 2048-element model for Homeland Security applications. Side-by-side measurements done with the gamma-ray imaging spectrometer (GRIS), contributed by Lawrence Livermore National Laboratory (LLNL), provided a baseline against which the BAT was evaluated.

Background

NASA chose a coded-aperture approach for the BAT because of its superior sensitivity when imaging point sources. For this same reason, it is a good approach for search and consequence management (CM) missions. Other coded-aperture cameras developed for Homeland Security purposes generally suffer from low energy and spatial resolution. This collaboration between NSTec and GSFC provided the opportunity to push the limits of coded-aperture technology.

Imaging with Coded Apertures

A coded aperture or coded mask is a pattern that creates a shadow on an image plane. For a point source, if the position of the image and the mask are known, then the direction of origin of a source can be derived. This method can be applied even if multiple sources are present. The shadows may not be recognizable to the naked eye, but the positions of the sources can be deconvolved by computer reconstruction. In principle, any shape can be used for the mask, but the most efficient mask pattern is one that is 50% open; also a more closed pattern obstructs too much of the detectors and leads to low efficiency, while a more open pattern reduces the contrast (Phlips, 2006).

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The BAT Instrument and the Engineering Model

The orbiting BAT consists of 32,768 pieces of $4 \times 4 \times 2$ -mm CZT to form a 1.2×0.6 -m sensitive area in the detector plane. Groups of 128 detector elements are assembled into 8×16 arrays and each array is connected to 128-channel readout application-specific integrated circuits (ASICs). Detector modules contain two such arrays and are further grouped by eights into blocks. The engineering model consists of one of the 2048-elements blocks and uses electronics identical to those on the BAT. A single 8×16 array, termed a “sandwich,” is shown in Figure 1.

Figure 1. Subarray of CZT detectors, called a “sandwich,” consisting of 128 individual detectors with a total active detector area of 328 cm². The sandwiches are the basic control unit within the larger detector array.

For use in the field, the GRSI was modified to ruggedize the instrument against stresses commonly encountered during search and CM missions (Figure 2). Furthermore, the algorithms that reconstruct the images were modified for Earth-based (i.e., non-infinite) source distances.

The BAT has a coded-aperture mask consisting of approximately 54,000 $5 \times 5 \times 1$ -mm³ lead tiles mounted on a 5-cm-thick composite honeycomb panel. The panel is mounted 1 m about the detector plane. The BAT coded aperture uses a completely random, 50% open/50% closed pattern rather than the commonly used uniformly redundant array pattern. The GRSI employs a similar type of coded aperture scaled to the smaller size of the detector (Figure 3). The mask is located 37 cm from the front of the detector array.

Figure 2. The GRSI. The control unit is contained in a cart measuring $48 \times 25.5 \times 22$ in. The CZT detector array, mask, and cooling unit are housed in a cart measuring $70 \times 36 \times 24$ in.

Figure 3. The coded-aperture mask of the GRSI, composed of open and closed cells measuring 0.5×0.5 cm. The closed cells are 1-mm-thick lead tiles.

CZT as a Detector Material

CZT is the acronym for the ternary compound $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$, where x is the blending fraction of ZnTe in CdTe. The range of x is generally between 0.04 and 0.2, corresponding to band gap energies of 1.53 eV and 1.64 eV (Knoll, 2000). CZT has many advantages when compared to other gamma-

ray-detecting materials. For example, the large band-gap energy and the sufficiently low amount of energy needed to create an electron-hole pair are properties unique to CZT. The wide band gap allows CZT detectors to be operated at room temperature, a desirable feature for field use. A result of the low energy for electron-hole pair creation is better resolution when compared to other room-temperature gamma-ray detectors (5%, or 3.3 keV at 60 keV). Other advantages of CZT are its superior intrinsic photopeak efficiency and good photopeak-to-Compton ratio, both of which derive from CZT's high atomic number.

CZT has several disadvantages that should be noted. First, both the mobility and lifetime of the electrons are very different than those of the holes. The measured mobilities are $1350 \text{ cm}^2/\text{V}\times\text{s}$ for electrons and $120 \text{ cm}^2/\text{V}\times\text{s}$ for holes (Knoll, 2000). The measured hole lifetime is between 100 ns and several microseconds for electrons and from 50 to 300 ns for holes (Knoll, 2000). However, the low mobility and short lifetime of the holes results in their being trapped very quickly, thereby preventing them from contributing to the formation of a full energy signal. Consequently, the photopeak suffers from low-energy tailing. Another disadvantage of CZT is the difficulty in producing high-quality, large, homogeneous single crystals. CZT is currently in high demand and remains one of the more expensive detector materials. Nevertheless, it is expected that new manufacturing techniques will develop, allowing the production of thick CZT. Also, the price of CZT is expected to decrease over time.

Project

Side-by-side Comparison

Tests to evaluate the performance of the GRSI were conducted using a ship container configuration at two GSFC locations: the Building 2 Hi-Bay facility and behind Building 76. The GRSI and the GRIS instruments were placed side by side, each viewing the source under test at the same distance to within 2 or 3 cm. Trials were conducted in the "on-axis" orientation for distance determination accuracy and spatial resolution. Off-axis trials were conducted for angular resolution determination, where, again, each instrument was viewing the source at the same off-axis angle.

Instrument Comparison

Both the GRIS and the GRSI use a coded-aperture design, are semi-portable, and can be turned on and ready to operate within a few minutes. The LLNL data analysis is ongoing during the data collection, whereas the Goddard instrument requires several minutes of post-collection analysis time to yield an image and source location. Figure 4 shows a typical *imaging* result from the BAT. The system also provides a host of numerical results. Its imaging capabilities simply tell you whether or not a source is present, along with an estimate of its intensity level.

Figure 4. Typical BAT image. The green circle highlights the bright spot in the image, indicating the presence of a radiological source.

Results

Detection Significance

Background Rate

The background rate is 5 to 20 counts/sec/sandwich. The amount varies within this range from run to run. Given the sources involved during the testing, the instrument was operating in the source-dominated regime (not the background-dominated regime).

Noisy Detectors

Periodically, a single CZT detector becomes noisy, and then the “noisy detector” algorithm is run (takes 2 minutes) to find and disable the noisy detector. It is not mandatory to disable noisy detectors—the coded-aperture technique is very forgiving in this sense, yielding only a very slight loss in sensitivity with a noisy detector. Also, there is only a very slight loss in sensitivity for having disabled detectors (the sensitivity loss is directly related to the square root of the detector area loss).

Detection Significance

Given the number of pixels in the image FOV, we require at least a 5- σ detection to guarantee that any “bump” in the image domain is not a chance coincidence noise fluctuation. Given the activities of the sources used (from 50 μ Ci to 60 mCi), we were routinely achieving 10- σ (or greater) detections in less than 30–60 sec of integration time.

Shielding

Shielding of the source creates a dramatic and problematic effect on detection ability. For example, imaging ^{137}Cs showed that a 47- σ detection, with no shielding, was dropped to an upper limit on the detection of 4 σ using only 0.9 cm of lead shielding. A stronger ^{137}Cs source (at roughly twice the distance) dropped the detection significance from ~ 30 σ , with no shielding, to 6.6 σ (just above the 5- σ threshold) with 1.1 cm of lead shielding.

Container Environment

A series of trials were conducted with a ship container (door open, door closed, and finally shielded by miscellaneous material in the container). The 62-mCi ^{239}Pu source went from a 27- σ detection (door open) to a 13- σ detection (door closed, 0.6-cm steel for the door). This was further reduced to a non-detection upper limit (4.2 σ) when the beam passed through an additional 17 ft of miscellaneous material (steel sheets, plywood, cardboard, wires, etc.). Essentially the same results were obtained for a series of runs using a 46-mCi ^{137}Cs source.

Distance Determination Sensitivity

To estimate the radial distance between the detector and the source, the coded-aperture technique relies on the divergence of the beam being detected. The GRSI estimates the distance from the detector, then looks at the σ value at that location. The maximum σ value yields the final estimate of the source location. As the source is moved farther away (i.e., the beam gets closer and closer to a parallel beam), it is more difficult to measure small differences in an already small amount of divergence. Table 1 shows the as-measured uncertainty of the distance determination.

Table 1: Uncertainty in the distance-to-source determination

Distance to the Source (cm)	Distance Uncertainty (cm)
540	50
1020	90
1650	300
1950	500

Note that this detector was not designed for this type of near-field activity. It was designed for sources at infinity (i.e., parallel beams), where distance determination is not considered necessary. We also have to note that with the GRIS there is no mechanism for distance determination. The instrument requires an estimate of distance before a measurement can begin. This requires the responder to essentially approach the source, which, at times, may be impractical.

Angular Resolution

Of interest in this measurement is the maximum FOV for the GRSI, compared to that of the GRIS. The imaging pixel size of the detector is 14 arcmin, which translates to an X,Y location accuracy of ± 2 in. at 50 ft for a 10- σ detection (± 4 at 100 ft; ± 8 at 200 ft). The Goddard instrument images and locates sources within a 90-degree FOV. Figure 5 shows a 2x zoom image of an unshielded 700- μCi ^{133}Ba source, using the GRSI. The source was placed 485.14 cm (15 ft, 11 in.) from the detector. Data were acquired for 15 minutes. The count rate was 50–200 counts/sec/sandwich. The detector was rotated 45 degrees off axis. The σ value for this configuration was 5.5, just above the unit's threshold of detection.

Figure 5. The off-axis limit of detection for the GRSI. The green circle around the white dot indicates the location and intensity of the ^{133}Ba source.

The GRIS can only image and locate sources within about a 30-degree FOV. Figure 6 shows an image of the same ^{133}Ba source, with the GRIS instrument situated at 15 degrees off axis. In this case, the source was 500.38 cm (16 ft, 5 in.) from the detector. The data acquisition time was 10 min with a count rate of ~ 530 counts/sec. The zoom level was set at one. Performance, as it pertains to the angular resolution of the GRIS, worsened with an increase in zoom.

Figure 6. The off-axis limit of detection for the GRIS. The greatest intensity (bright yellow pixels) corresponds to 20,220 counts being accumulated over an integration time of 10 min.

When configured at an orientation of 15 degrees off-axis, the GRSI acquired data for 13 min. The count rate in this case was 130–170 counts/sec/sandwich. The σ value was 32.3. It's worth noting that a source becomes “partially coded” as the angle of detection increases. As a result, a decrease in the σ value is observed (from 32.3 at 15 degrees to 5.5 at 45 degrees) and the image quality is noticeably diminished.

Spatial Resolution

In this measurement, the GRSI and the GRIS acquired images of four ^{133}Ba sources, each with an activity of 237 μCi . The sources were positioned 5.08 m from the detector. From left to right, the sources were spaced 129.5 cm (51 in.), 36.2 cm (14.25 in.), and 71.1 cm (28 in.), respectively. The data acquisition times were 10 min for the Goddard instrument and 20 min for the GRIS. The GRSI distinctly resolved the two (middle) sources closest together, separated by only 71.1 cm (Figure 7).

Conversely, Figure 8 shows that the GRIS system could not resolve the two middle sources; they appear as one. From the image, it is possible to localize the source by comparing source strength and pixel size. At a scaling factor of 1.01242, the calculated horizontal pixel size, as it appears on the bit-map image, is 11.516 cm. The calculated vertical pixel size is 10.665 cm. The highest intensity pixels are bright gold, as indicated in the figure. At these points, 11,015 counts were detected. Based on intensity and pixel size, one could argue that the sources in question are located at the points of highest intensity. However, even with pixel-related information, it is difficult to resolve the two sources with any amount of certainty without prior knowledge of their respective positions. The image still suggests that only one source is present in its center.

Figure 7. GRSI of (a) four ^{133}Ba sources and (b) two middle (of the four total) sources. Note the orientation of the sources. The true “up” direction (i.e., towards the ceiling) is to the left in the image and true “left” is down in the image (compare with GRIS image in Figure 8).

Figure 8. GRIS image of the four ^{133}Ba sources used for spatial resolution trials

Conclusion

The current GRSI has a good FOV and excellent spatial resolution, especially when compared to the GRIS. Depth determination was problematic; however, that is to be expected with such a unit. When detecting sources at “infinity,” distance determination is a non-issue. In general, the current GRSI was not designed nor optimized for this work.

Recommendations

There are several changes that could be made with the fabrication of a “second generation” instrument:

Thicker CZT

Thicker CZT (on the order of 5 mm) would allow the detection of higher energy lines. Currently, the BAT CZT at 2 mm is about 50% transparent at ~150 keV. CZT at 5 mm has good response up

nto the 500-keV range, and reasonable response up to 1 MeV. This higher energy response would provide much better full-energy peak performance for certain key radioisotopes (e.g., ^{137}Cs , ^{60}Co , U, and Pu).

Thicker Mask

Pushing to higher energies (thicker CZT) would require a thicker mask. A consideration would be 5-mm tungsten over the current 2-mm lead. This would maintain the mask modulation and greatly improve sensitivity at higher energies.

Pixelated Detector (Better Energy Resolution)

The “small-pixel effect” of pixelated detectors could improve the energy resolution. The peak widths of the gamma-ray lines in the spectrum would be about 2.5 times narrower. This would help in isotope identification. Incorporating improved charge collection performance would also improve the energy resolution.

Larger Width Dimension of the Detector Array

Increasing the width of the detector array could improve the distance determination of a radiological source by improving the ability to measure the divergence of the beam. The current packaging of the CZT array within the instrument envelope is not efficient. This could be improved by increasing the width of the CZT array by two, while maintaining the overall width dimension of the instrument (3 ft). Doubling the array width would improve the uncertainty values in Table 1 by a factor of two. Similarly, tripling would reduce the distance uncertainties by a factor of 3.

Compacting the Instrument

Currently, the instrument is contained in two mobile carts. The new design could combine the computer and power supplies into the main instrument cart ($2 \times 3 \times 5$ ft).

Improve Software

The current software package does not carry out the detector-by-detector gain and offset corrections to pulse heights of the photon energies. This corresponds to an approximate 10% error in typical photon line energies. These corrections are performed for the BAT on-orbit flight data, but are not incorporated into the program for the GRSI. A second-generation instrument should incorporate this correction, allowing energy cuts to be made in the imaging process, thereby reducing the background and improving the source-detection significance.

Currently, the imaging and location data processing occurs after the data is collected. This is because the current package was designed for functional testing of sub-units coming off the production line,

not for full-up instrument imaging function. A second-generation unit should incorporate image processing into the main instrument control program to achieve real-time imaging/locating functionality. This could also be combined with video-camera imaging of the scene being monitored.

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ADVANCED MICROWAVE ANTENNA ARRAY AND MULTIBAND RECEIVER

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Numerous organizations require the ability to detect microwave signals in a variety of bandwidths over a wide spectrum. In order to facilitate this necessity, an antenna or array of antennas capable of receiving the frequencies of interest must be designed and constructed. Because a span of nearly one decade must be kept from the lowest to the highest center frequency, as well as meeting additional challenges presented by microwave signal propagation, careful consideration must be taken in designing the geometrical and physical characteristics of the radiating structure. These design requirements can be realized via the definition of boundary conditions and material properties in an electromagnetic simulator. Evaluations of various electronic design automation (EDA) software packages were conducted, and a decision was made as to the most capable. Following this survey, an electromagnetic design simulator was purchased. Finally, proficiency in software operation was obtained through online training, and the generation of a preliminary antenna design commenced.

Background

For several years, the Mini Microwave Infrared and Acoustic (MIRA) instrument developed by the Security Systems Technology section at the Remote Sensing Laboratory (RSL) has been the premier tool for microwave detection. The original instrument incorporates a 9.3- to 10.8-GHz X-band microwave receiver. The X band has been the primary frequency band used by commercial manufacturers of microwave products of interest for many years. However, the commercial market is shifting to include microwave sensors that operate at 2.45, 5.8, and 24.15 GHz. All bandwidths are 50-MHz wide (except for the X band, which is 1.5-GHz wide).

Project

The ultimate purpose of this project was to design, develop, and build an antenna array that would be capable of covering all the frequency bands listed above, as well as a multiband receiver capable of processing those signals. In order to be effective for the intended purpose, the receiver must provide precise amplitude measurement (± 1 dB over each frequency band).

Design

The conceptual design anticipated that each antenna in the array would be similar in design, but that their individual sizes would be determined by their associated wavelength and substrate permeability.

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There would be three to four antennae making up each array. Each antenna of the array would be manufactured on individual substrates or possibly together on a single substrate. In order to negate the effects of the transmitted signal's polarization, all of the antennae would be circularly polarized.

Survey of Simulation Software

A vendor search was performed to locate potential candidates that could provide an electromagnetic simulation software package for evaluation. The investigation included Agilent's Electromagnetic Design System (EMDS), Zeland's IE3D, AWR's Microwave Office (MO), and Ansoft's Maxwell. Of these, MO and Maxwell were not capable of simulating far-field radiation patterns. Additionally, IE3D assumed an infinite substrate in its method of moments calculation, rendering it impossible for the program to calculate edge effects. This was problematic due to our radiating elements possibly lying near the edge of the substrate. Furthermore, IE3D did not possess an intuitive interface or easy-to-learn tutorials. EMDS, on the other hand, performed a finite element analysis, which captures all antenna and substrate edge effects. Through the calculation of Maxwell's equations, the electric and magnetic fields are derived, as well as the calculation of dispersion effects on the structure. Also, EMDS was simple to use and offered online tutorials that could provide easy-to-understand explanations.

During the evaluation period, we noted that EMDS provides the user with excellent feedback with regard to the radiating structure. The drawing environments of all evaluated software packages did not offer the drawing functions available in a full CAD environment, yet the chosen software needed to have the ability to represent two- and three-dimensional (2-D and 3-D) objects in an AutoCAD-like environment, while allowing some import or export functionality to other solid modeling programs such as Pro Engineer or Solidworks. This would enable the user to utilize the extended capability of these high-fidelity solid model design environments. The ability to draw and define shapes in Solidworks or Pro Engineer allows the user to define shapes with ease by means of equation definitions, leading to faster optimization of the design.

We chose EMDS because it has the ability to import and export 3-D solid model files in the ACIS SAT format to facilitate drawing interoperability between Solidworks or Pro Engineer CAD software.

A limitation of the EMDS software is that it does not directly generate Gerber files. This is the file format used by manufacturers to fabricate printed circuit boards. This minor issue could be addressed by implementing a work-around that would export the SAT files to a program capable of creating Gerber files.

Assigning Attributes

After the preliminary mechanical structure of an antenna is developed, using both the high fidelity solid model program and the mechanical portion of the EMDS program, each of its substructures are assigned attributes in the EMDS program (Figures 1 and 2). These include the real or complex

parameters of permeability, permittivity, electric-loss tangent, conductivity, resistivity, and magnetic-loss tangent. This completely defines each isotropic or anisotropic substructure. The surfaces of these structures can be assigned to be conductive, resistive, magnetic, or electric. This leads to the definition and modification of boundary conditions such as ports (with accompanying field direction and polarity), voltage sources, symmetry planes, and ground planes surrounding the objects. An interim design of the original Mini MIRA antenna is represented in Figure 1. Additional work needs to be performed concerning this structure, including the reflective cavity back and realization of the physical parameters.

Figure 1. Depiction of the original Mini MIRA antenna without the cavity back, radome, absorbing foam, or accurate material properties

Figure 2. Portrayal of coaxial back-fed 10.5-mm radius spiral with a duroid substrate

Simulation

Once the structures and the environment surrounding them are completely defined, a full simulation across a user-defined frequency range, as well as at discrete frequencies, is carried out. A conceptual design for the highest band antenna was completed (Figure 2).

Figure 3. Return loss at port one of the coaxial cable for the first propagating mode over the relevant section of the K-band antenna

Figure 4. Smith chart revealing a tight loop around a value of 1.0, which suggests a high resonance at that frequency

Further, the simulation convergence criterion can be specified based on changes in the scattering parameter calculations. EMDS provides the calculation and display of modal and nodal scattering parameters for each mode and port across the specified frequency range. This is represented by the magnitude and phase with associated real and imaginary components or as a graphical view. As can be seen in Figure 3, a plot of the K-band antenna scattering matrix for $S[1,1]$, a very deep resonance of -45 dB at 24.5 GHz. is demonstrated This graph indicates that most of the power is transmitted and is not reflected at the port. Furthermore, by analyzing the Smith chart in Figure 4, we can see the resonance is occurring around 50Ω due to the tight loop. The generalized S-matrices can be renormalized to particular impedances, such as 50Ω , and converted to admittance or impedance matrices. De-embedding of the ports and recalculation of the scattering matrix can also be performed, as well as a determination of the number of transverse electric or magnetic modes propagating at a port.

For simulation software to be truly useful in the area of antenna design, the electromagnetic fields propagating from the structure should be modeled. EMDS can plot normalized and unnormalized far-field radiated power intensity and electric field magnitude as a function of distance from the antenna in associated logarithmic or linear form. The electric, E-field magnitude, magnetic, H-field magnitude, cross- and co-polarized, and left- and right-hand circularly polarized far-field radiation patterns can be plotted either in polar or rectangular and two- or three-dimensional forms. The modeled far-field left- (green) and right-hand (red) polarized radiation patterns can be seen in Figure 5. Also, the ability to plot both surface current intensity and the Poynting vector is useful for simulation accuracy comparisons with other EDA software programs such as MO. The antenna data can be viewed at each port and mode for all simulated frequencies. This information includes radiated power, effective angle, directivity, gain, maximum intensity and associated maximum angle, and the maximum electric field magnitude in both polar and Cartesian coordinates in related directions.

Figure 5. Left- (green) and right-handed (red) polar plot of the far-field radiation pattern simulated at 24.5 GHz

Conclusion

It has been determined that the chosen EMDS antenna simulation package is very capable of modeling not only the far-field radiation patterns, but other antenna parameters as well. We suggest that future work verify the K-band antenna through laboratory tests after fabrication is completed at a board production facility. Before this occurs, however, further optimization is needed in the design to obtain a more directional circularly polarized far-field radiation pattern. After the conclusion of the initial antenna designs for the other bands and verification and optimization of the design for the X band, the antenna array could be finalized and a multiband receiver design could follow. It will be interesting to observe the manner in which the antennas interoperate and affect each other's frequency response as they are added to the array.

DETECTION OF CHLORINE GAS

Richard J. Venedam^{1,a}

The primary purpose of this project was to design, build, test, and deploy embedded piezoresistive microcantilever (EPM) sensors toward the detection of chlorine gas (Cl_2) in the atmosphere. The sensing materials used consisted of polymer matrices combined with sodium iodide (NaI) crystals. Sensors constructed from a silicone-based matrix exhibited the greatest response to Cl_2 , with detection limits in an outdoor exposure setting of approximately 20 ppm.

Background

Chlorine gas, Cl_2 , is a yellowish-green gas that may be recognized by its pungent odor, similar to that of bleach. Under pressure and with cooling the gas may be liquefied for transport and storage. When released at atmospheric pressure, it quickly becomes gaseous again, forming clouds of heavy chlorine gas that remain low to the ground. Chlorine gas is a pulmonary irritant that exhibits intermediate water solubility. Recently, in 2007, Al Qaeda in Iraq conducted a three-pronged suicide car bomb attack using chlorine gas in the Anbar province. Suicide truck bombers armed with chlorine gas struck at targets in the cities of Ramadi, Amiriyah, and Fallujah, resulting in 3 deaths and over 300 injuries.

Project

EPM sensors may be constructed for a variety of sensing applications. In each application, a custom sensing material is designed that will respond volumetrically to the desired analyte. For this project, we constructed EPM sensors for the detection of Cl_2 .

Design

All aspects of the design phase met initial objectives. Candidate sensing materials (such as NaI) were identified, synthesized, and successfully tested. The most successful sensing material consisted of a silicone polymer matrix with a 30% by weight loading of NaI crystals. This material proved to be stable over long periods of time and resistant to high temperatures, and also to have good sensitivity response to low levels of Cl_2 gas. Sensor hardware designs were finalized, including the incorporation of new sensor electronic interface boards with simplified design and lowered cost. The electronics package consists of three stacked boards occupying a volume with dimensions of approximately $2.5'' \times 1.75'' \times 1.5''$. (Figure 1).

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Figure 1. Circuit boards used in Cl_2 mote sensors

Build

Two types of EPM sensors were successfully constructed: USB interface EPM sensors for in-laboratory testing, and wireless “mote” sensors for outdoor remote deployment. The USB interface sensors, operated from any device with an unused USB connector, were tethered by a single USB cable that provided power to the sensors and two-way data communication. The mote interface sensors (Figure 2) consisted of the same sensor package interfaced to small, battery-powered motes. Individual motes had a range of approximately 400 meters and could communicate with each other in a network mesh, effectively extending the sensor range to several kilometers or more, depending on the number of motes deployed.

Figure 2. Fully assembled EPM sensor interfaced to wireless mote

Testing

Cl_2 sensors were laboratory-tested for both low levels of detection (100 ppm) and high levels of detection (1000 ppm). Sensors easily met these criteria, as illustrated in Figures 3 and 4.

Deployment

Sensors were tested at the Nevada Test Site (NTS) during an outdoor Cl_2 release. The release at very low levels (a few ppm) was successfully detected by the EPM sensors (Figure 5).

Figure 3. EPM sensor reading for Cl_2 exposure of 1000 ppm. The exposure occurred at a time of 135 seconds, and the sensor responded within 3 seconds of initial exposure. As the exposure continued the sensor continued to respond, indicating an integrating response.

Figure 4. EPM sensor reading from exposure to 100 ppm Cl_2 . Cl_2 was introduced at 70 seconds and the sensor response was almost immediate. This plot is of raw, unfiltered, unprocessed data.

Figure 5. Detection of Cl_2 at NTS at very low levels (tens of ppm). This test was conducted using a sensor interfaced to a battery-powered mote. The sensor was located 80 m downwind from the release stack and was subject to small variabilities in the wind speed and direction. The data shown have been filtered for noise. These data indicate an approximate level of exposure of 20 ppm.

Also, we have designed, built, and tested a portable, battery-powered quadrupole mass spectrometer to provide a measure of ground truthing in support of the chemical sensor development operations at NTS and elsewhere. This spectrometer was also tested for the first time at NTS during this year's effort in the development of a chlorine sensor. The tests indicated that measurement levels are achievable with partial pressures in the 10^{-9} to 10^{-10} torr range. Overall battery life in the field exceeded two hours at continuous operation. All measurements and data were read in real time on a small (8.4-in.) laptop computer integrated into the lid of the spectrometer case (Figure 6). The spectrometer can also continuously monitor specific, user-chosen masses up to 300 AMU, for tracking the presence of known agents.

Conclusion

We have constructed a fully portable, battery-powered quadrupole mass spectrometer system using primarily low-cost, off-the-shelf components. The case dimensions housing the system are $48 \times 34 \times 24$ cm. The system is capable of sampling continuously from the atmosphere using a differentially pumped inlet system, or it may be used with samples sealed in glass vials containing liquids, solids, or gases. The system has been tested on vapors from water samples contaminated with MTBE, TCE, or carbon tetrachloride.

Figure 6. (top) Internal components of portable quadrupole mass spectrometer and (bottom) spectrometer (including computer) in case

Results from this and a companion project (Porter, “Sensing materials,” 2009) have been presented at two international conferences (2008 International Meeting of the American Physical Society, New Orleans, Louisiana, March 2008, and 2008 International Meeting of the Materials Research Society, San Francisco, California, April 2008), and two manuscripts have been prepared and are currently undergoing review prior to submission to appropriate publications (Porter, “Sensing materials,” 2009; Porter, “A portable,” 2009). Also, another presentation at an international conference is planned (2009 International Meeting of the Materials Research Society, San Francisco, California, March 2009).

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MINIATURIZING MASS SPECTROMETERS

Stephan J. Weeks,^{1,a} Manuel J. Manard,^a and Rusty Trainham^a

The technical goal was the development of a handheld mass spectrometer (MS) for the detection of chemical, biological, and explosive compounds under field operating conditions. Paul- and Penning-type ion trap MS were designed, assembled, and tested. Simulations were developed to guide and verify instrument design and experiments. Initially, a Penning-type ion trap, constructed in an FY 2004/2005 SDRD project (Trainham 2005; 2006), was modified. Serial improvements to the designs were modeled and implemented. Multiple experimental trapping conditions and detection schemes were used. Although no evidence for the presence of trapped ions was acquired with either trap, further design modifications hold the promise for detection and miniaturization. Direct-air sampling systems were also developed and field-tested at the Nevada Test Site (NTS) on commercially modified quadrupole MS.

Background

Users continue to require more sensitive, selective, mobile, and real-time ground-truth chemical, biological, and explosive (CBE) compound diagnostic measurements for release events at the Non-proliferation Test and Evaluation Complex (NPTEC) at the NTS. For accurate, high-precision field analytical chemistry measurements, mass spectrometry is the gold standard to which all other trace atomic and molecular analysis technologies are compared. For this reason, a miniature MS would be a major step forward in analytical capability, and could lead to development of high-precision handheld instruments and small-profile (mote) wireless mesh sensor networks for CBE detection. Paul- and Penning-type ion traps both hold promise as low-power, low-cost, miniature sensors for integration into systems operations.

An ion trap MS is a device that utilizes magnetic and/or electric fields to confine an ion to a region of space within a vacuum system (March, 1995). Once stored, the trap can then facilitate ion detection based on mass-to-charge ratio. The Penning trap uses a magnetic field, together with a hyperbolic electric field, to trap ions. A constant magnetic field prevents excursion of the ions out of the trap in the radial direction while a low DC potential is used to trap ions in the axial direction. To localize the trapping region, the electric field must be at least quadrupolar in character. The resulting arrangement of magnetic and electric fields generates a three-dimensional pseudotrap at the origin, which is a potential energy saddle point. Ion motion in this geometry is characterized by three canonical frequencies: the cyclotron, axial, and magnetron. These frequencies arise from the interaction of the ions with the applied magnetic and electric fields and are functions of the mass-to-charge ratio of the

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ions, the magnitude of the field strengths, and the dimensional properties of the trap electrodes. Ion detection is performed with a high-sensitivity RF spectrum analyzer, which measures characteristic frequencies of image currents induced in the trap electrodes by the motion of the ions. In principle, any of the three canonical frequencies can be driven to generate image currents for ion detection. The Penning trap provides high levels of mass accuracy and selectivity allowing very high precision mass measurements.

A Paul trap MS functions similarly to a Penning trap. However, a Paul trap uses an oscillating RF field, as opposed to a magnetic field, to radially confine the motion of the ions in the trap. Typically, a three-electrode configuration is used in the Paul trap design, consisting of a ring and two endcap electrodes. The RF field is applied to the ring, while the endcaps are either grounded or a small DC voltage is applied. Ion motion in the Paul trap is complex, with ions oscillating between being forced in radially while simultaneously being pulled out axially, and vice versa. Mass selection is generally accomplished by destabilizing the trajectory of an ion by ramping up the RF potential. The orbital stability of an ion is dependent upon its mass-to-charge ratio, with lighter masses being ejected from the trap at lower RF potentials. Ions are ejected axially through small orifices in the endcap and can be detected using typical ion counting methods. Miniature (<0.5 mm) Paul traps can be operated at relatively high pressures (~1 torr) and give very high resolution.

Project

Two benchtop ion trap MS were designed, assembled, and tested. A Penning-type ion trap, first constructed in an FY 2004/2005 SDRD project (Trainham, 2005; 2006), was modified. The Paul-type ion trap was constructed for direct, side-by-side performance evaluation with the Penning trap. The results of this comparison were expected to help define the final design parameters for a smaller, fieldable ion trap MS. Both benchtop ion traps were directly mounted to standard 2.75-in. ConFlat vacuum hardware. The necessary high vacuum was generated by a 20-l/sec ion pump. The characteristic length parameter of both ion traps was $z_0 = 0.1$ cm.

Penning Trap

The benchtop Penning system utilized small permanent magnets mounted in vacuum to produce a trapping field of approximately 4.6 kG. Similar to a typical Paul trap geometry, a three-electrode configuration was used in the Penning design, consisting of two conical endcaps and a ring electrode (Figure 1). The design of the system was modified slightly from its original form to correct problems encountered in FY 2005. Specifically, the leads to the filaments, which are mounted directly to the outside of the endcaps, were spot-welded to maintain a closed-circuit connection to the electron beam. The methods through which the in-vacuum electrical connections are made to the trap were also improved to simplify the design. These modifications allowed for long periods of operation without any system failures.

Figure 1. Benchtop Penning trap. Copper endcaps are separated by a magnetic ring electrode. In-vacuum pill magnets are shown mounted above and below the trap electrodes.

Multiple detection schemes were used in attempts to detect trapped ions. Initially, RF voltage was applied to the trap endcaps to drive the ion motion; the induced signals were then measured on the ring electrode. Additionally, the RF drive voltage was also applied to the ring in an attempt to measure image currents on one of the endcaps. Since the magnetron motion is inherently unstable, the axial and cyclotron frequencies were independently driven to induce the image currents. Detection windows were set to search for both the primary and secondary harmonics of the drive frequency. Ramping of both the endcap DC bias and the RF drive frequency were employed to search for resonances. Since the effective z_0 is known only to a precision of 10%, the drive frequency and detection windows

were systematically scanned. In addition to these experimental parameters, the kinetic energy of the electron beam produced by the ion source was varied up to 60 eV, and the pressure of the sample gas in the vacuum chamber was varied from 10^{-8} to 10^{-6} torr. The gases sampled were H_2 , He, and N_2 .

Figure 2. Theoretical trajectory calculation of 1 eV H^+ under Penning trap experimental conditions. Endcap voltage = 5 V. Ion tracks are shown in dark blue.

Ultimately, no trapped ions were successfully detected with the Penning trap. This result could be due to two possible reasons. One, the ions may be radially diffusing out of the trap; even though simulations suggest that the strength of the 4.6-kG magnetic field is sufficient to trap ions (Figure 2), in actuality this may not be the case. Two, the magnetic field may only be strong enough to trap the ions for very short periods of time. The time required to make an image current measurement is approximately 10–50 ms. If the ions only spend 0.1–1 ms in the trap, no image currents will be detected. Possible solutions to these problems are to strengthen the magnetic field by fabricating all the electrodes that make up the trap from the same magnetic material as that of the in-vacuum magnets.

This could increase the field strength by up to an order of magnitude, greatly enhancing the trapping capability of the system. An RF field could also be superimposed over the magnetic field, yielding a Penning/Paul hybrid trap. Here, the RF field could compensate for the shortcomings of the magnetic field, and aid in providing the necessary radial confinement of the ions in the trap.

Paul Trap

The geometry of the Paul trap is essentially identical to that of the Penning trap (Figure 3). The RF field was generated using an Agilent 8648C signal generator (the same unit used to drive ion motion for the Penning system) and an Amplifier Research 10LA power amplifier. The gain of the power amp ranges from 30 to 50 dB for an input frequency range of 1 to 110 MHz. This setup was able to provide RF voltages of up to 40 V (0 $\rightarrow$ peak) at frequencies of 1 to 30 MHz. Simulations conducted on this system showed that N_2^+ at thermal energies would be confined for up to 10 ms using a trapping frequency of 10 MHz at an amplitude of 20 V (0 $\rightarrow$ peak)—well within our experimental range. Ion detection was facilitated by applying a negative extraction voltage to the upper endcap to draw positive ions out of the trap. A Channeltron electron multiplier was placed in close proximity to the upper endcap and a variable bias of -900 to -1500 V was applied to the cone for ion detection (Figure 3).

Figure 3. (left) Benchtop Paul trap with (right) Channeltron electron multiplier detector

As with the Penning trap, multiple detection schemes were applied to the system and several experimental parameters were varied to obtain and optimize ion signals. The most promising method involved biasing the electron beam on (up to -60 eV) for 50 ms to create ions in the trap, then rapidly switching the beam off (~ 50 μ s to reach 0 V). The extraction voltage was then applied to the endcap 0.5 to 10 ms after the beam was off. The delays were set using a Stanford Research Systems DG535 delay generator, and bias voltages were switched on and off using Crydom solid-state relays. During the process, endcap biases were scanned, both positively and negatively (including ground), the energy of the electron beam was varied, the extraction voltage was scanned (1–60 V), and several sample gases were introduced to the system (H_2 , He, N_2 , and ambient air).

As with the Penning system, no conclusive evidence for the presence of trapped ions was acquired. Initially, the problem was believed to be due to RF crosstalk between the ring and endcap electrodes. However, this was corrected by decoupling the wires delivering the RF to the ring from those providing the DC biases to the endcaps. Nevertheless, no trapped ions were detected. It is now believed that the problem stems from the close proximity of the filament to the lower endcap. If the electric field generated when the filament is biased is penetrating into the trap through the 1-mm-diameter endcap orifice, then trapping conditions of the system could be severely destabilized. A new ionizing method has been envisioned where the filament would be moved away from the trap and placed at a 90-degree angle to the plane created by the trap and the detector. The electron beam would penetrate into the trap through an orifice that would be machined into the ring. This would serve to shield the trapping region from the electric field generated by the filament and, by moving the electron beam perpendicular to the detector, reduce the probability of ions formed outside of the trap reaching the Channeltron.

System Considerations

Efforts towards miniaturizing other system components were also performed. Of note is the development and testing of direct-air sampling components in the laboratory, which yielded successful test and evaluation results using a commercial quadrupole MS at the NTS NPTEC. Miniaturization of electronics, vacuum, and other components will require finalization of the MS design.

Conclusion

Penning- and Paul-type benchtop ion trap MS were designed and constructed. Simulations were incorporated into the design work and were used to verify and explain experimental results. Design improvements were first modeled and then implemented. Multiple experimental trapping conditions and detection schemes were used. Although no trapped ion signals were acquired with either trap, we expect our newest design modifications, once implemented, will trap and detect ions with the sensitivity and selectivity common to mass spectrometry. When ion signals are obtained, designing a smaller, field-portable ion trap MS using our new simulation capabilities should be straightforward. The direct-air sampling device and methodology, demonstrated at the NTS NPTEC, is ready to be

integrated into the miniature MS system. Additionally, this research engendered new concepts to reduce or eliminate interferences, plus interest in development of instrumentation and methods for a variety of applications. Development of portable MS sensor networks in support of ground-truth diagnostics at NPTEC and autonomous, mobile MS systems for persistent monitoring applications are only two of the many CBE applications expected.

Acknowledgments

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ELECTRONICS

INFORMATION SECURITY TACTICAL OPERATIONS

Patrick Kiley^{1,a}

The primary goal of this project was to develop a platform and software to monitor and/or prevent the use of an 802.11 wireless network. The platform is based on a small tablet portable computer (PC), as well as on many open-source Linux-based tools, integrated into one platform. This toolset can be used as a security assessment tool for use by network engineers and security professionals, as well as an offensive toolkit used to deny and monitor a targeted wireless network.

Background

Wireless networking allows anyone within the radio range of a wireless device to potentially access that device, unlike traditional networking, which requires one to have a physical connection in order to send and receive data. Consequently, wireless 802.11 networks are very easy to set up and run, but very difficult to secure. The hacking community is very aware of the faults and vulnerabilities inherent in wireless networks. Typically, the best way to properly secure a network is to attack it, thus discovering the weak spots in the network that can then be appropriately addressed. A toolkit, such as the one developed in this project, can be used to discover and address all of the radio-based risks of operating a wireless network.

For this project, it was posited that a small PDA or very small PC could be used as a “leap-off point” to eavesdrop on and deny access to a wireless network. We also wondered if a cellular data connection could be used to control such a device remotely. With this cellular link, the device could be covertly installed somewhere and then accessed remotely, keeping the operator well beyond the security perimeter of the physical location being monitored or attacked.

The current version of the National Nuclear Security Administration (NNSA) NAPs 14.2.C, which governs NNSA security policy, states that organizations operating in the Unclassified Mandatory Protection Information Group (or higher) must scan for *unauthorized* wireless devices on a quarterly basis (NNSA Policy Letters, 2008).² All organizations, regardless of whether or not they have a deployed wireless system, must scan for wireless devices once a quarter. The toolkit conceived of in this project, once developed, would make an extremely effective and very inexpensive (approximately \$2000) way for any organization to meet the requirements of this policy.

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² Note that Official Use Only Information, Personally Identifiable Information, and Unclassified Controlled Nuclear Information all fall under the Unclassified Mandatory Protection Information Group.

Project

Platform (Customization)

The primary goal of this project was to research and develop a wireless platform to monitor and deny access to an 802.11 wireless network. The platform eventually selected was engineered to meet the following criteria:

- small in size,
- lightweight,
- easy to conceal,
- a cellular link to allow remote control,
- the ability to gather GPS data in order to assist the operator in locating radio transmitters,
- an 802.11 radio that can communicate on both the 2.4- and the 5-GHz frequencies,
- a limited ability to be customized with longer range antennas and newer wireless radios, and
- functional at the 802.11 protocol level, instead of being just a wireless jammer.

An initial goal of the project was to eventually use either a cell phone or PDA as a test platform for the software. After a bit of time spent researching this possibility, it was quickly determined that PDAs and cellular phones do not have a very powerful or flexible radio. A new class of very small x86 computers has come onto the market recently: the Ultra Mobile Personal Computer (UMPC). Instead of the limited processing power and non-x86 architecture of a PDA, these units are full PCs loaded with a keyboard and laptop operating system, such as Windows XP and/or Vista. Given that the UMPC made by OQO (OQO, 2008) was only slightly larger than a PDA, we decided to focus all efforts on the UMPC platform. After researching appropriate 802.11 wireless chipsets and Linux OS requirements, an OQO model 2 was purchased (Figure 1). This platform has a full keyboard, Atheros 802.11 a/b/g chipset, Evolution Data Optimized (EVDO) cellular modem, and solid-state hard drive. This unit weighs just one pound and measures 5.6" × 3.3" × 1".³ The OQO platform also has USB connectivity. This USB port was tested with both a USB GPS receiver as well as a high-powered 802.11 radio, the Alfa AWUS036H. The radio has a Realtek chipset and can transmit at powers up to 500 mW, giving it exceptional range. The Alfa also has an external RP-SMA attachment, so different antennas, such as a directional antenna, can be attached. This capability increases the flexibility of the platform, allowing it to be located farther from the network being monitored, audited, or disrupted.

³ Recently, there have been a larger number of UMPCs released to the market. Some of these are ruggedized and would make an excellent platform for continued research in this area.

Figure 1. OQO Model 2 open, with case

Operating System

The vast majority of wireless network security testing is done using the open-source Linux⁴ operating system (OS). The ability to modify source code, as well as the open source nature of device drivers, made this the obvious choice for a base OS to use for this project.

Some of the major Linux distributions, or distros, include RedHat, Slackware, Debian, and Gentoo. In addition to distros, there are dedicated variants developed by teams that customize the Linux code to be used for a particular task. One of these releases, BackTrack, is a Slackware-based OS that can be used as either a live CD-ROM or an installed OS. BackTrack is a dedicated toolkit used by information security auditors and penetration testers (remote-exploit.org, 2008). It natively includes a large number of wireless security tools and was therefore the best choice for a baseline OS from which to do the research for this project. BackTrack versions 2 and 3 were both used to perform the actual testing of the platform's effectiveness. A description of the changes made to the base BackTrack image is attached to this report as Appendix A.

Cellular Radio

Having the ability to leave the wireless denial and audit platform in situ was determined to be an important capability. It would be a much more covert way of monitoring traffic if the device could be left hidden and either operated until the battery died, or connected to external power and then left until

⁴ "A GNU/Linux operating system is made of a Linux kernel, a set of GNU tools, an installation program, a package management system and a lot of other software components. Because all these components are free to use and to distribute, anybody can assemble and configure them according to their needs and create their very own GNU/Linux operating system. Since 1993, a lot of people and companies have been distributing Linux operating systems. These distributions made it easy for people to get and to install a working GNU/Linux system on their personal computer" (Linux Forums, 2008).

later recovered. For these reasons, a cellular data radio became an important capability of the toolkit. The OQO comes with an EVDO modem, which is a relatively high-speed cellular radio. This radio can support up to 3.1 Mbits/sec. Although the radio rarely exceeded 512 kbits/sec in practice, it was still fast enough to be used as a control channel. There were a few issues getting the EVDO modem to operate as a useful control channel and backhaul from which to send captured data. These issues centered around two problems. The first was a high variation in the latency from the cellular link. The latency for data to cross the cellular data network and return would vary from 200 to 2000 ms. In the Voice over Internet Protocol (VoIP) world this is called jitter, and it can seriously hamper real-time communications, such as voice. For data connections, it is just annoying to the user, though it does not impact the control channel (except when being used interactively). The second issue with EVDO was its implementation on Linux. The cellular providers and hardware companies have not written Linux device drivers for EVDO; the Linux community has had to reverse-engineer the connection to USB serial drivers. The USB serial drivers are rate-limited to about 200–500 kbits/sec since they were written for low-speed RS-232 connections, such as terminals and analog modems. Later in the project, the USB serial drivers were patched, which improved the performance of the connections, but still did not reach the potential speed of the link as when using the Windows device drivers.

GPS Logging

GPS information is very useful when analyzing radio signals. If a wireless capture device is able to embed GPS latitude and longitude, in addition to signal strength, it then becomes very easy to locate a radio transmitter. These tools and their built-in basic mapping capability were briefly explored in this project. After the Linux GPS device drivers were stabilized, it was easy to map wireless transmitters and locate their approximate, and sometimes even exact, location. It also became very easy to capture the track traveled by the wireless capture device and display that track on a map to verify all the important areas were covered by the track. This is important, as a security auditor would require verification that a wireless site survey has covered all areas of interest when searching for unauthorized wireless devices.

Wireless Radio

The chipset used on the 802.11 wireless radio was a key component to this solution. There are only a few major wireless chipset vendors, and only a portion of these have seen large-scale development by the Linux community. This is due in large part to the manner in which wireless chipset vendors share their device driver architecture with the public, as well as chipset capabilities. In order to be used as a wireless security tool, the wireless chipset must be capable of being placed into different wireless states by the driver. One of these states is called monitor mode. This mode is similar to the promiscuous mode on wired Ethernet connections. It allows the device driver to capture information seen by the firmware of the radio and then pass that data up to higher levels of the operating system. The normal mode of many drivers does not allow this, and will only pass information if the wireless firmware sees data that comes from a connected or “associated” device. In addition to monitor mode,

the drivers need to support packet injection as well as access point (AP) mode, where the wireless driver acts as an access point and accepts client connections. Packet injection is a special modification to the device driver, which allows the wireless radio to transmit packets out the radio even when not associated to an access point. Packet injection was key to our toolkit, as it even allowed modified packets that do not match the IEEE specification to be transmitted out the radio interface.

As previously mentioned, the OQO platform has an Atheros chipset radio. The Atheros chipset is very popular for wireless security since it can use the madwifi drivers (MadWiFi project, 2008). The madwifi drivers have been specially modified for BackTrack, and allow for a great amount of customization, including packet injection. There has recently been research into the capability of the Atheros chipset, through which it was discovered that it is basically a software-defined radio that can be modified to do much more than communicate via 802.11. In addition, the software can be modified to communicate on frequencies specifically forbidden by FCC regulations (d'Otreppe de Bouvette, 2008). The external radio tested on this system for longer range capabilities has a Realtek chipset. This chipset is not as flexible as the Atheros, but does support packet injection and monitor mode. This allows it to be used with the majority of tools developed on this platform.

Protocol Level

The final requirement for this toolkit was that it needed to work at the 802.11 protocol level, instead of simply acting as a jammer disrupting the wireless spectrum. Jamming the wireless spectrum is simple: a screwdriver wedged in the door of a microwave oven with the oven turned on makes an extremely effective 802.11 jammer. By working at the protocol level, the platform can be selective about its target, allowing it to not only be used as an eavesdropping device, but also as a network denial and network penetration tool.

Methodology and Effectiveness⁵

The system was tested against several different types of wireless networks: a centrally controlled enterprise network; a robust, stand-alone, AP-based enterprise network; and the ubiquitous consumer-level network. It was discovered that a blended approach, one that relied on several methods of attack, was the most effective way to disrupt a wireless network. The blended attack worked like this:

- First the toolkit deauthenticated the client(s) attached to the wireless access point. It does this by impersonating the wireless AP and sending a specially crafted packet to the client, tricking it into thinking the access point has asked it to disconnect.
- Next, the toolkit broadcasted multiple beacon frames with fake AP addresses, imitating the network the client(s) were connected to (Figure 2). This happens because 802.11 wireless networks use an identifier called an extended service set ID (ESSID) to uniquely identify them.

⁵ **Disclaimer:** All active testing done on this system was performed against targets owned by NSTec and configured in a test environment meant to simulate an actual representation of a configured wireless network.

By broadcasting multiple fake APs with the ESSID of the network under attack, the client tries multiple times to connect to different APs that do not exist. If it should find a live AP and start the connection process, then as soon as it attempts to send data, the first step will repeat, and the client is once again deauthenticated (Figure 3).

- The third step involved flooding the AP with fake association requests from fake clients. This consumes resources on the AP and is the most likely reason the consumer-level APs needed to be rebooted to restore the network. They simply have all their memory consumed to the point that they lock up and can no longer process traffic.
- The final, and optional, step was rather clever. Multiple crafted packets were sent from the radio interface that mimic a particular type of attack. This attack is aimed at confusing the wireless intrusion detection systems on the market, and it proved to be 100% effective and 100% repeatable at locking up an AirMagnet system (the one intrusion detection and analysis tool purchased for this project).

Figure 2. Screenshot of attack underway, showing large number of fake APs captured by Airmagnet tool (all APs in yellow are fake)

With these multiple attacks, the wireless networks tested were nearly always disrupted, due to the nature of the protocol itself. This will always exist as a risk that must be measured when considering deployment of a wireless network. The only mitigating factor for the attack method outlined above was an implementation of 802.11w, supported by drivers having Cisco Compatible Extensions (CCX). Also called management frame protection, 802.11w can only be deployed if all the clients on a network, plus the AP, support it. CCX-supported clients also have their own shortfalls; for example, a problem with the 802.11w protocol was discovered and published at the 2008 BlackHat and Defcon16 conference (Ahmad, 2008).

Figure 3. Screenshot of attack underway, showing only one network listed by the Windows client. If the client attempted to connect to this network, it would fail.

From the information presented in Table 1, it is obvious that the toolkit was extremely effective at disruption of wireless networks. In addition, it could be used to target and disrupt a single network as long as the toolkit's signal was within range of the appropriate receiver. For eavesdropping, the converse was true: as long as the toolkit was within range of the target of interest, it could capture traffic from it. For encrypted traffic, there were several techniques that were used to decrypt traffic. Unless the target was using a very long static Wi-Fi Protected Access (WPA) key or a properly secured enterprise deployment of Extensible Authentication Protocol-Transport Layer Security (EAP-TLS),

traffic could be decrypted by brute forcing or tricking the AP into revealing the keys, such as in the case of weaker wired equivalent privacy (WEP)–encrypted networks. Using directional antennae and an external radio, the range of this toolkit could be increased dramatically for both transmit and receive.

Table 1. Tested systems and results

Model and Brand	Software Revision	Toolkit Effectiveness
Linksys WRT600N	1.01.36, Build 3	100%; device required a reboot to restore service
Linksys WRT54GX2	1.01.22	100%; device required a reboot to restore service
D-Link DGL-4500	1.10	100%; device required a reboot to restore service
Apple Airport Extreme & Express	7.3.1	100%; device required a reboot to restore service
Cisco Outdoor Mesh 1520	4.1.192-22M	80% effective; CCX was only mode difficult to completely disrupt
Cisco 1230 a/g	12.3-8.JEA3	90% effective; easier to disrupt than controller-based unit or draft N unit; CCX was only mode difficult to completely disrupt
Cisco 1250 Draft N	12.4.10b-JA3	80% effective; CCX was only mode difficult to completely disrupt

Conclusion

This toolkit would be extremely flexible and portable. It is much easier to walk around with a 1-lb scanner than an awkward laptop when trying to scan for unauthorized wireless devices. One of the more effective ways to hide an unauthorized wireless device is to have it operate at a frequency prohibited by the FCC for use in the U.S. However, just because it is prohibited does not mean that it won't be used. All a malicious individual would have to do is download non-U.S. firmware for the wireless transmitter.

Today, this toolkit can be used with a minimum amount of training as an effective way to scan for unauthorized wireless devices, in addition to being used as a risk-assessment tool for groups considering deployment of a wireless network to support their operations. The security practice of wireless scanning, or walking around a controlled area looking for unauthorized wireless devices, is not just good cyber security practice, it is also required by NNSA policy. The toolkit can currently scan all 2.4- and 5-GHz frequencies, allowing it to detect rogue devices operating in these bands; it does this without

violating FCC regulations, since it is simply listening and not transmitting. Add in GPS information and the wireless auditor could produce some very easy-to-interpret maps showing the path traveled and the interpreted location of all detected wireless devices. Since many NNSA facilities are located in urban areas, this would be an especially important feature. Since 802.11 signals can travel some distance, it is easy to confuse the source of a wireless signal when seen inside the perimeter of an NNSA facility. For organizations needing a tool to monitor and secure their own wireless infrastructure, a toolkit such as this one could become a very valuable way to assess the actual effectiveness of security controls placed on such a system.

Further research into this toolkit could focus on expanding the platform with a mesh of very small computers (like Gumstix) connected back to a controller like the OQO. In addition, some emerging capabilities, such as the high-bandwidth 802.11n extensions, present some very interesting security capabilities that could be researched to improve the effectiveness of this toolkit as a denial and network penetration testing tool.

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Appendix A: BackTrack modifications

1. Modified X11 and KDE to get BackTrack working on the OQO display.
2. Modified drivers for Verizon EVDO modem.
3. Modified USB serial drivers for EVDO modem to get it operating, as well as improve the performance of the EVDO modem.
4. Updated Aircrack-ng to 1.0 Beta 2.
5. Modified and updated Kismet.
6. Modified GPSD to get it working. In its native state, GPSD is extremely unstable (thanks to Josh Wright, SANS Institute, for helping me modify this driver to get it working).
7. Installed Charon 1.0.
8. Installed Charon 2.0.
9. Installed MDK3 version 4, and modified for effectiveness.
10. Installed Hunter-Killer.
11. Installed lorcon (loss of radio connectivity).
12. Installed coWPAtty WPA auditing tool.

DRIVING THE ULTRAFAST STREAK CAMERA

Jonathan J. O'Connor^{1,a}

Based on previous experience in the design of streak camera systems, this research aimed to produce a subnanosecond sweep rate (yielding picosecond temporal resolution) using a 50- Ω streak tube, impedance matching network, and an avalanche pulser. The Electro-Optics Systems Engineering and Advanced Sensor Development groups located at Livermore Operations (LO) developed a prototype ultrafast streak camera. Initial testing demonstrated that the sweep rate is approximately 225 ps across 50 mm.

Background

Streak cameras are primary diagnostic tools in many large-scale DOE experiments. Cameras produced by LO have achieved streak rates of 5 ns. But in recent years, there has been a demand for streak cameras with faster and faster sweep speeds. One user community, which uses many LO-built streak cameras, is the High Explosive Application Facility (HEAF) at LLNL. One of their projects, studying the shockwave propagation through high explosives, requires picosecond temporal resolution. The new diagnostics that will be performed at the National Ignition Facility will also need streak cameras that can measure the temporal resolution (tens of picoseconds to hundred of femtoseconds with dynamic range) of the reaction histories, dynamic temperatures of targets, and others.

Several previous SDRD projects have paved the way for this capability. In an FY 2005 project, “Optimization of Streak Tube Temporal Resolution” (Shellman, 2005), Shellman replaced the inductive-capacitive deflection plates with an internal 50- Ω impedance strip line, thereby reducing the self-resonant notch from -60 dB at 930 MHz to -20 dB at 2.5 GHz. Utilizing this improvement would allow us to demonstrate subnanosecond performance. As part of the FY 2005 project, a tube with the improved sweep structure was fully assembled and was available for this year’s project.

Project

For this project we used the 50- Ω streak tube, an improved impedance matching network, and an avalanche pulser to design a prototype ultrafast streak camera that has a subnanosecond (hundreds of picoseconds) sweep speed.

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The following project milestones were established:

- Design drive circuitry, consisting of:
 - » Avalanche pulser,
 - » Matching network,
 - » Cables/high-voltage (HV) bias tees, and
 - » Negative/positive/common mode HV DC bias board.
- Bench-test the prototype using oscilloscope
- Evaluate prototype in laser lab

Prototype Bench Testing Using an Oscilloscope

Electrical tests using the oscilloscope helped determine an approximate sweep speed. The waveforms in Figures 1, 2, and 3 represent signals received by the 50- Ω streak tube deflection structure. The negative ramp waveform (Figure 1) had an amplitude of -2.53 kV with a fall-time of 450 ps. The positive ramp waveform (Figure 2) had an amplitude of 2.57 kV with a rise-time of 450 ps. We know that it takes ~ 1.25 kV to create a full deflection across the 50-mm streak tube. With this in mind, applying half the amplitude of the ramp waveform of 2.50 kV, or 1.25 kV, would reach the amplitude needed to create a full deflection; therefore, we applied half of 450 ps, or 225.0 ps (approximate sweep speed).

We also performed tests to determine if both the negative and positive ramps arrived simultaneously at the 50- Ω deflection structure. Figure 3 shows the negative ramp (brown), positive ramp (black), and the inverted (math function) negative ramp waveforms (dark yellow) arriving at the same time.

Prototype Evaluation in the Laser Lab

The prototype ultrafast streak camera system was transferred to the laser lab for optical testing and to measure the actual sweep speed. We used an EKSPLA Model PL 2143 laser, which produces a 532-nm wavelength and a 25-ps pulse width. After the laser and the prototype ultrafast streak camera were timed, we collected the streaked image in Figure 4, which shows the actual laser pulse (somewhat round circle of light) in the bottom-center of the image. We are unsure of the cause of the horizontal black and white lines in the center of the image. The CCD camera used was the SI 800 CCD, which has a $4K \times 4K$ chip and 9- μm pixel resolution. Figure 5 shows the vertical line-out of the streaked image that was used to calculate the sweep speed.

Figure 1. Negative ramp waveform shows amplitude of -2.53 kV and fall time of 450 ps

Figure 2. Positive ramp waveform shows amplitude of 2.57 kV and rise time of 450 ps

Figure 3. The negative ramp (brown), positive ramp (black), and inverted (math function) negative ramp (dark yellow) waveforms arrive at the 50-Ω deflector simultaneously

We can calculate the number of pixels and the physical size of the CCD camera as:

$$\text{Number of pixels} = 4K = 2^{22} = 2^{10} \times 2^{12} = 4096 \text{ pixels}, \quad (1)$$

and the distance as:

$$\text{Distance} = (9 \text{ } \mu\text{m} \div 1 \text{ pixel}) \times (4096 \text{ pixels}) = 36.864 \text{ mm}. \quad (2)$$

From the data in Figure 5, we calculated the FWHM of the laser pulse. Cursor 1 = 1249 and Cursor 2 = 620; therefore, the number of pixels that this laser pulse covers in 25 ps is $(1249 - 620) = 629$ pixels. Next, we extrapolated the sweep speed across the CCD camera as:

$$\text{Sweep speed across CCD} = (25 \text{ ps} \div 629 \text{ pixels}) \times 4096 \text{ pixels} = 162.80 \text{ ps}. \quad (3)$$

From this, we extrapolated the sweep speed across the 50-mm streak tube as:

$$\text{Sweep speed across 50-mm tube} = (162.80 \text{ ps} \div 36.864 \text{ mm}) \times 50 \text{ mm} = 220.81 \text{ ps}. \quad (4)$$

Our calculations show the sweep speed across the 50-mm streak tube to be ~ 220.81 ps, which, when compared to 225 ps, reasonably matches our earlier calculation from the electrical bench testing with the oscilloscope.

Figure 4. Streaked image with the laser pulse in the bottom-center of the image

Figure 5. Vertical line-out of the streak image used to calculate FWHM of pulse as 629 pixels in 25 ps

Conclusion

We developed a prototype streak camera system with a sweep speed of 225 ps. The current performance of this system is adequate for many applications; however, further work is required to produce a robust field ready camera. Recommendations would include an improved charging line and reprocessing of the streak tube for better impedance matching.

Acknowledgments

Our team members were essential in the success of this research project. We thank Robert Buckles for sharing his expertise in avalanche pulsers and impedance matching; Richard Shellman for contributing his knowledge in streak tubes; Jerry Cradick for soldering the circuit boards, assembling the prototype streak camera, and working in the laser lab; Robert Guyton for timing the streak camera and doing the optical testing in the laser lab; William Warthan for performing the optical testing in the laser lab; Jeffrey Anderson for procuring all of the necessary parts for this project; Russell Knight for providing drafting support; and Jim Mirador for laying out the circuit board.

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Sections of this technical report have been deleted because they contain technical data whose export is restricted by the Arms Export Control Act (Title 22, U.S.C., Sec. 2571, et seq.) or the Export Administration Act of 1979, as amended, Title 50, U.S.C. App. 2401 et seq. In order to obtain the full technical report, contact the author.

PHOTONICS

TERAHERTZ TIME-DOMAIN SPECTROSCOPY AND IMAGING

Robert A. Buckles^{1,a}

This effort continued work begun in FY 2007 to develop terahertz time-domain spectroscopy (THz-TDS) and imaging technology for potential Stockpile Stewardship missions and directed research opportunities. Using the system developed last year, we successfully demonstrated THz-TDS of non-hazardous, non-explosive materials obtained from Non-Explosive Security Testing and Training (NESTT) explosive material. Our success may further future endeavors in detection of improvised explosive devices (IEDs). The FY 2007 system was also a springboard for the design of a much more powerful, single-shot THz imaging system for hydrodynamic measurements. In this regard, we were marginally successful at demonstrating THz imaging and generating a laser plasma. We characterized the plasma generation with Faraday cups and a gated camera, but THz measurements were not forthcoming due to sparse availability of the High-Energy Density Physics (HEDP) Short-Pulse Laser (SPL) facility.

Background

Using a system built in FY 2007 and refined in FY 2008, we endeavored to construct a single-shot THz imaging spectrometer, utilizing the laser source in the HEDP SPL. The original concept is nearly a decade old (Shan, 2000), and pharmaceuticals and coatings industries have recently shown interest in this type of spectrometer (Yasuda, 2006). Realizing such a capability would allow unique plasma diagnostics for Defense Experimentation and Stockpile Stewardship missions, such as advanced radiography with DARHT, RITS, Cygnus, and neutron sources such as the dense plasma focus (DPF) machine, and shocked materials characterization. Understanding the detailed phenomena of high-voltage breakdown and hydrodynamic expansion on these machines will greatly benefit reliability and usefulness for the DOE Science Campaign. On another front, we would be better poised to offer domestic security solutions in the way of explosive material detection. Such a high-power THz system can provide the throughput needed for a real-time detection system.

Project

The first part of this project validated last year's development in THz spectroscopy, while making a useful measurement of high-explosive (HE) material. The spectroscopy system was optimized by stabilizing the laser, carefully aligning the beam path, and reducing system noise. A crucial step was to set up a microscope and view the laser spot on the emitter and detector chips (Figure 1). The layout was fully characterized such that the emitter and detector could be removed and the laser

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retro-reflected and aligned through the THz path, then remounted with the electrodes centered. This careful approach guaranteed spatial alignment of the THz beam with the optical probe, so a temporal scan was nearly guaranteed. The spectrometer setup with a sample holder and the beam path are shown in Figure 2. Stabilization of the room temperature at 70°F was the final key to enabling repeatable performance. More system details can be found in the FY 2007 report (Buckles, 2008).

Figure 1. (a) Emitter and (b) detector chips viewed through a powerful microscope. Electrode gaps (circled) are 50 μm and 10 μm , respectively. When the laser strikes inside the circled portion, a detectable signal is generated.

Figure 2. (a) THz optomechanics inside the purge box, showing addition of Teflon lenses, microscope mirror, Keithley triax cabling, and sample holder stage, and (b) the optical beam path to the spectrometer

Although we have access to the High-Explosives Application Facility located at Lawrence Livermore National Laboratory (LLNL), and could acquire actual HE, we decided it would be expedient to first try a less-hazardous approach with diluted, non-explosive samples. In order to make an equivalent measurement, the samples would need to be of a non-interacting matrix material, have a high-enough percentage of HE, and be finely dispersed. The NESTT² canine training aids were appropriate, containing approximately 8% by weight of HE in a petroleum jelly suspension. Five samples were obtained: *n*/TNT, *n*/RDX, *n*/PETN, *n*/KNO₃, and *n*/KClO₃; the *n*/ prefix designates a NESTT material.

Considerable research went into selecting a container and designing a holder for the samples. The aluminum cube shown in the foreground of Figure 2a has six 8.7-mm holes passing through. The holes define the probing path, and the wires keep the sample precisely located within the holder. The assembly is stepped from one path to the next when rotating samples without breaking the purge. The block is sized for nine 10-mm cuvettes, although only three side-by-side were used. The cuvettes are slightly tapered, allowing for slightly different sample probing thicknesses as the stage is moved up or down. The material, polystyrene, was chosen for its inertness to the THz spectrum; however, the 1.2-mm wall is thin enough to cause etalon pulses within the scanning range. (This is actually a good feature if a multipass Fresnel reflection analysis is applied, as it provides variable thickness.) The material samples are shown loaded in the cuvettes in Figure 3.

Figure 3. Samples in polystyrene cuvettes.
1,2 = petrolatum; 3 = empty; 4 = *n*/TNT; 5 = *n*/RDX;
6 = *n*/PETN; 7 = *n*/KNO₃; 8 = *n*/KClO₃.

It was visibly apparent that the samples might behave differently. We assumed that the different types of HE in each sample would be responsible for the variations, as opposed to some inconsistency in the petrolatum matrix. The petrolatum sample must be relatively transparent to make a good reference

² NESTT materials were developed and licensed by LLNL to the XM Division of Van Aken International, 9157 Rochester Court, Rancho Cucamonga, California, 91730.

against the HE. Figure 4 shows a relative temporal scan of the petrolatum (with polystyrene) versus the empty polystyrene cuvette. Figure 5 shows the absolute and relative transformed spectra (both amplitude and phase) of the THz-TDS scans. The petrolatum amounts to a loss of 20 dB out of 60 dB of dynamic range out to 3 THz. The relative measurements are reliable to ~ 1.5 THz, after which noise begins to influence the ratios.

Figure 4. THz-TDS scan of empty cuvette (blue) and petrolatum sample (green). The 15.5-ps delay through the petrolatum implies a nominal index of refraction of 1.46, with little absorption or dispersion.

Figures 6 to 10 show temporal (samples 1 and 2 are offset for clarity) and relative spectral measurements for the five NESTT samples. The scan time in all cases requires about 285 seconds for 1024 steps of 33.3 fs each for a scan rate of 120 fs/sec, and an effective sampling rate of 30 THz. With a real-time detection, single-pole, time-constant of 100 ms, or 1.6-Hz bandwidth, the effective bandwidth is 13.3 THz with a Nyquist limit of 15 THz. A slower scan with narrower (real-time) bandwidth will improve the dynamic measurement range.

Figure 5. Absolute transmitted spectra of petrolatum samples and the empty cuvette. The power spectral density is normalized to a 1-THz bandwidth. The time-domain amplitude is obtained by multiplying by a 13.3-THz bandwidth. The difference in phase slopes indicate the petrolatum index of refraction.

Figure 6. Temporal (offset for clarity) and relative spectral measurements for n/TNT samples

Figure 7. Temporal (offset for clarity) and relative spectral measurements for n/RDX samples

Figure 8. Temporal (offset for clarity) and relative spectral measurements for $n/PETN$ samples

Figure 9. Temporal (offset for clarity) and relative spectral measurements for n/KNO_3 samples

Figure 10. Temporal (offset for clarity) and relative spectral measurements for n/KClO_3 samples

The n/TNT and n/RDX showed a strong characteristic absorption at 1.2 THz and 0.9 THz, respectively, while the n/PETN showed a broad resonance at 0.7 THz. It is not yet clear how these measurements compare to those made by other researchers. The potassium nitrate (n/KNO_3) and potassium chlorate (n/KClO_3), though barely distinguished from the petrolatum, showed some resonances at high frequency. A more careful scan with optimized sample preparation may reveal characteristic absorption features. Nevertheless, due to the very fine phase sensitivity of this coherent technique, the dispersive properties are well-resolved.

Next, we created a more energetic THz source, using the HEDP SPL, and a hydrodynamic plasma sample from a Spectra-Physics Quanta-Ray 600-mJ pulsed laser. Following the general layout of Shan (2000) and Yasuda (2006), we configured a THz imaging system that would fit inside a vacuum chamber, specifically the one used to produce the laser plasma (Figure 11). As a single-shot system would likely develop in steps, we anticipated running the experiment continuously for several minutes and integrating the signal over many plasma shots. We constructed an aluminum target to provide repeatable, fresh material at every shot, using a stepper motor and linear actuator. Most of the equipment was on-hand and only required tailoring, assembly, and the addition of rudimentary controls.

To monitor the plasma expansion and timing, we installed Faraday cups inside the chamber, facing the target (Figure 12), and collected plasma current versus time data. Two cups biased alternatively for collecting electron versus ion current were installed. A PI-MAX gated camera (Princeton Instruments) recorded plasma emission, both prompt and delayed. A 450-nm band-pass filter (40-nm BW) filtered the light and isolated the thermal components. The light coming from the target spot (Figure 13a) was clearly in the blue-UV spectrum, due to the high temperature, whereas the gaseous emission in front of the target (Figure 13b) was due to decaying excited material and background gas.

Figure 11. (a) Laser plasma experiment chamber and (b) view of aluminum target disc

Figure 12. (a) Side view of aluminum target, stepper motor, linear actuator, and (b) Faraday cups

Plasma current traces for various bias voltages (Figure 14) revealed expected behavior on only one cup: large electron signal with positive bias, small ion signal at negative bias. The other Faraday cup appeared to have about half as much current. We theorized that the target was not normal to the laser beam; therefore, most of the plume was going toward one cup. Various bias voltages were tried on one cup, and an I-V characteristic could be made, but this was a low priority and will be resolved when necessary. Overall, the laser plasma chamber was very successful, and we are indebted to the exceptional contributions from the HEDP group.

The eventual goal was to move the laser chamber into the SPL to perform THz spectroscopy on the plasma. The design in Figure 15 fit the THz components inside the vacuum system, thereby eliminating water vapor and atmospheric absorption. We used 2-mm-thick ZnTe crystals for optical rectification generation and electro-optical detection (Winnewiser, 1997; Jiang 1999). We carefully measured

Figure 13. (a) Laser spot and plasma emission unfiltered and (b) filtered with 450 nm bandpass filter

Figure 14. Faraday cup traces at alternating bias polarities showing electron version ion collection

the damage threshold to be 1 mJ for the 150-fs, 6-mm beam, which agreed well with previous figures (Soileau, 1982). With only a few days to assemble the optics and establish the pump-probe timing synchronization, progress was very difficult, as one can never be quite sure what is happening with the beam until detection is initially established. Only a very meticulous beam prealignment could have helped set the temporal delay line accurately with a parallel input beam to guarantee spatial alignment during the time delay. Still, we achieved good temporal alignment with a fast photodiode. By placing the photodiode at the pump-probe interaction point, we could block either arm sequentially and measure the histogram of the laser pulses. The delay line was adjusted to achieve overlap of the histogram peaks with picosecond accuracy. This narrowed the parameter space in which to hunt for the interaction to within half a millimeter (placement error of the photodiode).

Figure 15. Design of the THz beam generation and probe paths inside the vacuum system (a) and realization on the benchtop (b) showing additional beam routing, delay, and electro-optical detection components.

To increase sensitivity, we obtained an EG&G 162/165 Boxcar averager mainframe and gated integrator plug-in with 115 and 181 current preamplifiers. By using balanced detection and monitoring the difference in photocurrent on the probe beam, we achieved a very sensitive nanoampere detection limit. Near the expected temporal overlap, we observed a very small coherent component buried in the much larger (low-frequency) noise component. It's difficult to be quantitative with a 10-Hz repetition rate, but it seems that we found the THz interaction. Unfortunately, there was no opportunity to improve the signal.

On further review, we strongly suggest using a collection lens on the THz generation crystal. Despite using a collimated input beam, the output THz beam had a much shorter wavelength and expands, as if it had a 2-mm waist. A special collection lens (Pax, Ltd.) was used successfully by Yasuda (2006).

A suitable lens would need to be optically transparent, as well as THz-transparent. Our research indicates that Tsurupica lenses are available from Microtech at a premium cost. We would endeavor to obtain a Tsurupica lens given the opportunity to repeat this exercise.

Conclusion

We have presented THz-TDS data on NESTT materials in the range of 100 GHz to 1.5 THz, showing characteristic absorption and dispersion. Although the spectral resolution could be improved with longer acquisition time (the petrolatum matrix is more absorbent at higher frequency), the investigation deserves some comparison with actual HE. We have assembled a laser plasma system and a THz imaging system to measure hydrodynamic plasma behavior. There was not sufficient beam time to improve the signal nor to investigate the plasma behavior. We will continue to propose future development to the U.S. Department of Homeland Security on the use of THz spectroscopy for IED detection. Further improvement in the THz imaging optics could yield an image. A laser with a higher pulse rate also seems a necessary component to success, and if this work continues, we will continue to lobby to obtain a dedicated high-powered research laser.

Acknowledgments

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MANY-POINT VELOCIMETRY USING HETERODYNE TECHNIQUES

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Expansion of photonic Doppler velocimetry (PDV) techniques into a many-point PDV (MPDV) system was investigated via laboratory experimentation and computer modeling. Selected experiments within the Stockpile Stewardship program that require many PDV data channels and other experiments that historically use line-VISAR (velocity interferometer system for any reflector) diagnostics are the target applications. We investigated the feasibility of techniques and hardware configurations that could multiplex and/or frequency-shift PDV data channels in order to make use of available bandwidth within the recording system frequency domain. The research included evaluating candidate multiplexing techniques (wavelength division, polarization, and coherency) through laboratory demonstration of specific MPDV configurations, evaluating acousto-optic frequency shifting (heterodyne) techniques, and analyzing harmonic suppression within these techniques. Our MPDV configurations are applicable to frequency-based analysis methods. Results of laboratory experiments indicate several techniques can successfully exploit the available bandwidth within a PDV recording system without generating harmonics detrimental to data fidelity.

Background

PDV-based velocimetry has become a widely used diagnostic, both within and outside of the DOE community. Diagnostic groups typically possess capability to measure data channels (probes) numbering from a couple to as many as twenty. However, there exist two categories of experiments that require many more channels than are typically available. The first category involves experimental geometries (and corresponding velocimetric behavior) with well-defined symmetries, and the second involves geometries with a significant degree of uniformity. Diagnosing these experiments may require a high channel count that either cannot currently be met or may require alternative diagnostics such as line-VISAR. We were motivated to expand a conventional PDV system into an MPDV system to potentially leverage existing detection and recording hardware by multiplexing data channels onto a single recording channel. The technical aspect of the existing PDV system we wished to exploit was the large amount of unused bandwidth, within the recording system frequency domain, which exists for a typical data record. By “filling” this available bandwidth (frequency space) with additional data channels, one could maximize the use of high-value digitizers while maintaining data fidelity and system portability.

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To make use of the available bandwidth within a PDV data record, two areas were investigated. First, we sought to determine the best (if any) method for optically multiplexing data channels together without the generation of harmonic interference that could potentially corrupt data fidelity. Second, we investigated whether there was a suitable method for frequency shifting multiple data channels. Frequency shifting is necessary for experiments that exhibit a significant degree of uniformity because the velocity profiles are very similar and multiplexing is only possible if the profiles are separated—“un-stacked”—in frequency space. Therefore, we investigated configurations of frequency shifting data channels relative to each other while simultaneously multiplexing them.

Initially we evaluated potential multiplexing and frequency-shifting techniques through computer and mathematical modeling, and then followed up with laboratory benchtop measurements. This report deals primarily with the results of our benchtop measurements. The laboratory investigations sought to evaluate three multiplexing techniques: wavelength division multiplexing (WDM), polarization multiplexing (PM), and coherency-based multiplexing (CBM). We also wished to evaluate frequency-shifting techniques, sometimes referred to as up-shifting or heterodyne mixing, generated with acousto-optical (A-O) modulation and stand-alone lasers. Evaluation of the candidate techniques and configurations is based on (1) data fidelity and extent of harmonic suppression, (2) degree of data assurance—a dependable and robust approach, and (3) cost effectiveness. It should be emphasized, however, that all of the multiplexing techniques discussed in this report are valid *only* for PDV data analyzed by frequency domain techniques such as Fourier transform.

Project

Benchtop experiments were designed to test specific multiplexing and frequency-shifting configurations. Candidate configurations were assembled and tested using commercially available hardware: 2-W, 1550-nm, single-mode erbium continuous wave (CW) lasers (IPG Photonics); a 20-GHz optical receiver (Miteq SCMR-100K20G); a 12-GHz digitizer (Tektronix 6124c); and fiber-optic circulators and splitter/combiners (both polarizing and non-polarizing) from various vendors. Optical probes were commercially available (Oz Optics) with back-reflection values ranging from 20 to 60 dB. Two methods of generating a PDV beat frequency, often referred to as the “local” and “remote” oscillator approaches, were evaluated. One approach uses the optical probe to generate reference (or un-Doppler shifted) light (local), while the other uses a fiber-optic pick-off near the laser output (remote).

To provide signal generation (probe data channels) due to surface motion, we employed audio speakers as probe targets, modified by affixing retro-reflective tape. Each speaker was driven by a frequency generator operating at a fixed frequency ranging from 40 to 90 Hz. Speaker surface motion is typically several tenths of a meter per second, which generates PDV beat frequencies of several hundred kilohertz. The speaker-driven data channels proved to be very stable, producing repeatable sinusoidal signals. Speaker and probe test fixtures, along with a typical PDV data record, are shown in Figure 1.

Figure 1. Test box used for benchtop measurements with a typical velocity time history record (inset). Up to four audio speakers and eight probes were employed. Note the sinusoidal speaker motion is always positive going, i.e., absolute value of sine. This is because a “conventional” PDV measures the absolute beat frequency and is unable to discern direction-of-travel, i.e., discern blue shifted (“forward”-traveling) speaker motion from red shifted (“backward”-traveling) speaker motion.

It is worth noting that while PDV is often capable of measuring velocities up to several kilometers per second on explosively driven experiments, it is also capable of clearly resolving the very small velocities generated for these measurements.

An important aspect of these investigations is the ability of candidate MPDV configurations to suppress the generation of undesirable beat frequencies, referred to here as “harmonics.” A single PDV data channel will return two optical signals, one affected by surface motion and the other unaffected. The electric fields from these two intrachannel signals will manifest themselves at the photo detector as a beat frequency that we record. We define a harmonic as the beat frequency generated from interactions between multiple probes (interprobe) via electric field interactions at the photo receiver. Our goal was to optically combine multiple probes at the photo detector, but to allow *only* beat frequencies

from each probe, and to suppress (or disallow) interactions between differing probes. To demonstrate the potential for confusion due to harmonics generated within a multichannel PDV data record, we multiplexed four data channels without harmonic suppression, permitting all interactions. Figure 2 illustrates this situation and the potential difficulty one would have in discerning real velocity profiles from an interprobe harmonic.

Figure 2. PDV velocity record of four optically combined data channels. Channels were driven from a common laser and combined by a four-to-one fiber-optic combiner/splitter. This method does not provide harmonic suppression and demonstrates the similarity of inter- and intraprobe beat signals.

Of the three multiplexing techniques investigated, WDM, PM, and CBM (Mujat, 2004), the WDM and PM appear to be most promising. The CBM approach is problematic because it requires the laser state of polarization to be completely unpolarized (a statistically stationary beam approximating “natural” light) with a degree of polarization (DOP) equal to zero. Commercially available lasers for PDV operate at a fixed polarization state with a DOP = 1, i.e., they are completely polarized, and the output beam is not readily depolarized. The remainder of this report addresses experimental results of WDM and PM as well as A-O frequency shifting.

Wavelength Division Multiplexing and Polarization Multiplexing

The two multiplexing techniques that proved successful, wavelength division and polarization, are based on the following principles.

1. **Wavelength Division Multiplexing:** WDM employs multiple lasers with each operating at a different frequency (laser wavelength), and every probe multiplexed onto a given detector/digitizer channel is driven from a separate laser. The separation in frequency of any combination of these lasers must be much greater than the recording system bandwidth. In this way, harmonics generated by interactions of the various lasers will lie outside the PDV recording bandwidth and be of little concern. An example of four data channels multiplexed in this manner is shown in Figure 3. While WDM is relatively straightforward, it should be noted that higher-order harmonics (beat frequencies interacting with themselves and other sources) do exist, but are typically reduced in intensity by 20 to 30 dB.
2. **Polarization Multiplexing:** PM is based on the principle that orthogonal polarization components will not produce a beat frequency because they do not interfere. To confirm this, we combined two data channels (probes) from a laser using a polarizing fiber-optic splitter/combiner so that their polarization states were orthogonal to each other upon combination (Figure 4). The mutually orthogonal polarization states are preserved within the fiber-optics, effectively suppressing intraprobe harmonics. There is, however, a significant risk to data fidelity from this approach if the state of polarization of either beam is orthogonal (or nearly so) to the polarizing fiber-optic combiner. In this case, the combiner will not transmit and data loss can occur.

Figure 3. Four data channels multiplexed by WDM. Each of the four velocity time histories contained within this record is generated by a separate laser-probe-speaker combination. All fiber-optic components are polarization independent.

Figure 4. Two data channels multiplexed by PM. Signals were generated by a common laser, with two probe/speaker combinations operating at different frequencies. Within the inset sketch: F/O = non-polarizing fiber-optic splitter/combiner, VA = variable fiber-optic attenuator, S/P = polarization dependent, fiber-optic combiner/splitter, and probe back-reflection value is listed on the probe.

Frequency Shifting

For experiments that provide physical symmetries, multiple PDV target locations may often be planned such that velocity profiles occur at different times and with substantially different velocities. For these experiments, frequency shifting may not be required since the experiment, in essence, does that work for you. However, for other experiments that provide geometric/velocimetric uniformity, the data channels might be expected to have very similar profiles—velocity histories on top of each other. In this case, frequency shifting or some other means of signal separation (e.g., time delay) is essential.

We investigated the use of fiber-optically coupled A-O modulators called Bragg cells or frequency shifters, as well as the use of an independent laser as the source of reference light. This reference light was combined with the affected light returning from the probe in order to generate a heterodyne beat frequency. We acquired two A-O modulators (Brimrose Corporation of America and EM4, Inc.) and both functioned reliably and to specification, providing a known frequency shift. The A-O approach is advantageous since the Doppler shift imparted is exactly equal to the acoustic drive frequency, and can be measured to very high precision and accuracy. An A-O employed in this fashion uses light

from the same laser used by the probe; thus, the A-O follows any drift in the laser frequency, which can be as much as several hundred megahertz. Use of an independent laser proved to be less attractive because of this same potential for frequency drift occurring on the time-frame of the experiment. Another advantage of an A-O modulator is that it may be used to generate a beat frequency even when no surface motion exists; this provides a gauge for the experimenter to determine signal strength.

A significant by-product of frequency shifting is the ability to determine direction of travel. A full description of this topic is beyond the scope of this report; however, to demonstrate, we have provided a comparison of a conventional PDV data channel with the same data simultaneously recorded after frequency shifting by an A-O modulator. This is shown in Figure 5.

Figure 5. Direction of travel demonstration by a single probe/data channel, both frequency shifted and unshifted (conventional) recorded simultaneously. The conventional PDV record displays typical positive-going behavior, which cannot, without additional information, discern “forward” travel from “reverse.” The frequency-shifted signal provides direction of travel; the signal beneath the horizontal “up-shift” line is due to forward motion (towards the probe) and the signal above the line is due to backward motion of the speaker (away from the probe).

Multiplexing and Frequency Shifting

Finally, we combined frequency shifting and multiplexing via both WDM and PM. We believe the most straightforward approach is to use WDM since the additional laser power is required in any case to drive the additional probes, as shown in Figure 6.

Figure 6. Demonstration of WDM and frequency shifting. Three probes were combined, each driven from a separate laser. The first probe was not frequency shifted, the second probe was up-shifted by 165 MHz, and the third was up-shifted by 425 MHz. We also placed an optical chopper in the beam path of one probe to test for effects of signal discontinuity within the analysis software. A notional matrix is provided (inset) as an example of how 16 data channels could be multiplexed onto a 4-channel digitizer.

Conclusion

We successfully conducted laboratory investigations of various methods to multiplex PDV data channels leading to candidate MPDV configurations. Wavelength- and polarization-based multiplexing techniques proved feasible; however, WDM is the preferred candidate since additional laser power

is necessary for the additional probes, and it also avoids the risk to data fidelity due to orthogonal polarization states. In conjunction with multiplexing, we investigated the use of A-O modulators to provide frequency shifting and have found them to be a reliable and robust method for stacking data channels in frequency space. The extent to which one could multiplex PDV data will probably be determined by photodetector dynamic range, economy, and tolerance to data risk more than bandwidth limitations. We estimate that 4-to-1 multiplexing of four PDV probes per digitizer channel would reduce the cost per probe by a factor of three and still maintain data fidelity. An 8-to-1 MPDV arrangement would reduce the cost per probe by approximately $4\frac{1}{2}$ with a moderate risk to data fidelity. We demonstrated the feasibility of multiplexing and frequency-shifting techniques and provided tools with which an experimenter may consider multiplexing and frequency shifting options. We believe that construction of an MPDV field demonstration system would be appropriate to validate these techniques on explosively driven experiments. If validated, an MPDV system could prove to be a valuable tool to diagnosticians both within and outside of the DOE community.

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A MINIATURE DUAL-USE STREAKING CAMERA AND PHOTODETECTOR

Joseph D. Delash,^{1,a} Al Shellman,^a John Yuhas,^a Don Pellinen^a (former employee), and Don Ng^a (former employee)

This project investigated the feasibility of a design for a miniature streaking camera/photodetector of greatly reduced size and weight that can be mass produced at a low cost. We also designed, manufactured, and used tooling fixtures to manufacture these streak tube assemblies. We designed a miniature tube and built six streak tubes. We were not able to perform tests on the streak tubes because of a procedure problem in the assembly of the tubes. This problem can be resolved by performing the gold plating process before the glass fritting process.

Background

Streak tubes are used in several core diagnostic systems. Conventional streak tubes and cameras are very large, heavy, and costly to produce. The smaller, modular, traveling-wave deflection system of the miniature dual-use streaking camera and photodetector that we have designed can be built using integrated-circuit, hermetic sealing techniques and will allow multiple cameras to be deployed where one camera is currently being deployed. Besides being more compact, this design is less expensive to manufacture in quantity, has faster sweep speeds, and provides electron deflection comparable to standard tubes but at lower voltages. The electrodes are more precisely spaced than on conventional tubes because of more accurate manufacturing techniques, smaller dimensions, and the fact that components would be rigidly fastened on both ends rather than cantilevered as is now done. This device can be used to measure x-rays directly, or UV or visible light. Outfitted with filters, it can be used as a spectrometer. The miniature streaking tube can be used for two applications with or without spectroscopic components: (1) with optics as an imaging streaking tube, or (2) without optics as a photodiode self-readout photodiode.

Our tube design takes advantage of the traveling-wave deflection concept that was patented by Rudolph Komptner in 1942 and incorporated into oscilloscopes built by Tektronix in the 1950s. A traveling-wave tube has an electrode structure that is designed as a constant impedance transmission line where the electric wave is timed such that the voltage arrives at successive deflection electrodes at the same time as the electrons to be deflected. The same voltage deflects electrons at multiple stations and is finally terminated to eliminate reflections. The improved frequency response or rise time comes from the constant impedance of the electrodes. The voltage does not “rattle” back and forth as it does in existing, unterminated, variable-impedance electrodes. When work at the National Ignition Facility (NIF) and/or the Sandia Z machine begins developing projected fusion yields, these miniature tubes

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could prove very useful, as they would be disposable due to their ease of manufacture and low cost. This would eliminate the safety concerns and cost associated with recovering and decontaminating the reusable tubes.

Project

The miniature traveling-wave streak tubes we built feature:

- Windowless front surface photocathode,
- Three electron acceleration stages,
- Gating electrode positioned before deflection electrode,
- Impedance-matched traveling-wave deflection stage, and
- Capability of being mass produced; damaged or radioactively contaminated units may be disposed of safely.

Two views of the tube are shown in Figure 1. The left view shows the tube with its end plate (with slit), while the right view shows the tube without its end plate (with slit). Photons, entering the top at a grazing incidence of ~ 10 degrees, strike the photocathode, which is biased at a negative voltage. Some of the electrons accelerating toward the anode pass through the first anode slit, where they are accelerated to and through the next anode slit, and accelerated to and through the gating plates to the offset plates (see Figures 2 and 3). The offset plates are designed as a constant-impedance, parallel plate transmission line. The offset ramp waveform is injected from a coaxial transmission line on one

Figure 1. Mini streak tube, shown with alignment frame (left) with end plate and (right) without end plate

side and carried off on the other side by another cable to terminate the signal. Using the constant-impedance line, it is possible to avoid the resonances and oscillations common on existing National Security Technologies, LLC, (NSTec) tubes. In this design, the slits are approximately 0.3 mm wide, and the centerline spacing for leads is approximately 6.7 mm.

Figure 2. Depicted is a cross section of the mini streak tube assembly, showing the photon path

Figure 3. Completed (alignment frames removed) mini streak tube assembly

Interior parts for the traveling-wave streak tube utilize ceramic insulators, while outside metal forms are identical to the parts developed for the disposable x-ray diode developed in an FY 2006 SDRD project (Pellinen, 2007). Figure 4 shows the interior part with the photocathode, the second accelerator stage on the top of the gating plate, and one conductor of the offset transmission line. Figure 5 shows another view of the plates. The parts in Figures 4, 5, and 6 were etched out of a flat sheet of 0.005-in. alloy 42.

Figure 4. Model of the alignment frame for the upper electrodes showing the photocathode, second anode plate after bending, upper gating electrode, and offset upper plates

Figure 5. Model of the alignment frame for lower electrodes showing the anode plate after bending, lower gating plate, and lower offset plates

Figure 6. Etched alloy 42 metal parts, ceramic parts, and an assembly held together with glass fritting and the vehicle/carrier material known as Alfa Terpeneol

After the design was completed, we procured and received all of the etched alloy 42 metal parts, the ceramic parts, the glass fritting, and the vehicle/carrier material (Alfa Terpeneol) required to fabricate the prototypes. Preliminary experiments in bonding the ceramic parts to each other and the alloy 42 parts to the ceramic parts were conducted. We had excellent results in bonding the ceramic parts to each other, good results in bonding alloy 42 parts between two ceramic parts, and poor results bonding the alloy 42 parts at the ends where they attach to the ceramic parts on one side only. We also drilled small holes through the alloy 42 end pieces and applied glass frit through the holes and into the ceramic (forming glass rivets); this technique produced much better results.

The photocathode must be bent ~65 degrees and the anodes ~45 degrees to the sheet. We designed tooling to accomplish the bends in one stroke. The vertical stroke will come down and press and meet the horizontal die. The vertical and horizontal stroke tools are shown in Figures 7 and 8, respectively. The bending fixture is shown in Figure 9.

Figure 10 shows a 2-assembly holding fixture that was designed, constructed, and used to assemble and bake out fritted parts in the Advanced Sensor Development oven located at Livermore Operations. Figure 11 is a model of a 10-assembly holding tray that was designed for mass production of the miniature streak tube. Ten 10-assembly trays were fabricated to allow for the production of 100 assemblies at a time.

Figure 12 shows the initial test parts and assemblies that were shipped to a vendor for acid cleaning and electroplating with nickel and 5 μm of gold. The arrows on the part are pointing to bars that will be used to attach the electrodes utilized during the gold-plating process. The miniature streak tubes

were assembled and fritted together prior to being gold-plated. The acid-cleaning process degraded the glass frit to a point that it flaked away from the tube. This occurrence caused a short circuit across the streak tube leads and thus rendered them unusable.

Figure 7. Vertical stroke-forming die bends anodes to ~45 degrees and photocathode to ~65 degrees

Figure 8. Vertical die comes down to meet horizontal die and creates bends

Figure 9. Bending fixture

Figure 10. A 2-assembly holding fixture

Figure 11. A 10-assembly holding tray

Figure 12. Initial test parts and assemblies

Figure 13 is a model of the mini streak tube setup in the existing Manson chamber that was to be used to supply x-rays to the photocathode. The current generated (projected to be a nanoamp) would have been measured using a photodiode.

Figure 13. Mini streak tube mounted to the existing Manson chamber

Conclusion

A miniature streak tube and the tooling required to manufacture them in small and large quantities were designed. Methods of successfully fritting the etched metal parts to the ceramics were developed. We successfully exercised techniques of glass fritting the etched metal parts to the ceramics. However, we were unable to perform tests on the assembled mini streak tubes because the frit was damaged during the gold-plating and acid-cleaning process. Because the lead oxide in the frit was broken down, the electrodes short-circuited. This experience led us to conclude that the cathode should be gold plated prior to assembly and the fritting process. This would result in better tubes capable of tests.

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Pellinen, D., J. Yuhas, "Disposable x-ray diode for fusion shots," *Nevada Test Site-Directed Research and Development*, FY 2006, National Security Technologies, LLC, Las Vegas, Nevada, 2007, 233–239.

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DYNAMIC OPTICAL ADJUSTMENT OF A PHOTONIC DOPPLER VELOCIMETRY SIGNAL IN REAL TIME

Heather Leffler,^{1,a} Adam Iverson,^a Araceli Rutkowski,^a Wendi Dreesen,^a and Jason Young^a

This project sought to develop a new control system to dynamically adjust the optical power of a photonic Doppler velocimetry (PDV) return signal in real time. When the beat amplitude drops below the noise floor of the detector, frequency (particle velocity) data are lost. This project considered analog and digital solutions to this problem. Commercial off-the-shelf (COTS) evaluation boards were used for the designs. First, we modified a semiconductor optical amplifier (SOA) driver evaluation board to improve its response time. Then, two analog automatic gain control (AGC) circuits and one digital AGC circuit were made using COTS evaluation boards. Both prototypes were tested with lab-simulated PDV data; we demonstrated that amplitude correction in the time domain did correspond to an increase in the signal-to-noise ratio (SNR) in the frequency domain of 15 to 20 dB. The analog design was also tested with dynamic data obtained on an in-house slapper system with limited success. It was difficult to slow the slapper down enough for our prototype to be able to adjust the gain; we could see the SOA turn on, but the data record was too short (20 μ s) to make data analysis worthwhile.

Background

PDV, a type of high-speed velocimetry used for measuring particle velocities, was originally developed by Strand (2006) at Lawrence Livermore National Laboratory (LLNL). Two wavelengths of light are heterodyned together producing a beat frequency that is the difference in frequency between the two waves. The “beats” are caused by the constructive interference, or combined amplitude, of two different waves as they pass the same point. PDV detects the beat frequency from two slightly different frequencies of light by shining laser light at a 1550-nm wavelength (in the infrared spectrum) on the surface of a moving target. The Doppler-shifted laser light reflected off the target surface is collected and sent to a detector. At the same time, some of the original light is sent back to the same detector. The incident laser light has a frequency of 1.93×10^{14} Hz, or 193,414.49 GHz. If the target is moving at 1000 m/sec, the Doppler-shifted laser light will have a frequency of 193,415.78 GHz. Both frequencies are so rapid that no existing detector can directly measure them. However, high-speed detectors can measure the difference in frequencies—the beat frequency—because it is much slower than either the original laser light or the Doppler-shifted light. By applying a fast Fourier transform (FFT) routine to raw PDV data, we obtain beat frequency as a function of time, which a conversion factor then scales to velocity. When PDV is fielded on some dynamic experiments, the

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return signal from the probe can drop very low, typically due to dynamic speckle or surface tilting. When this happens, there is a corresponding loss in particle velocity information when the frequency domain data are analyzed (Figure 1).

Figure 1. The PDV signal problem illustrated. When the signal drops very low in the time domain, there is a corresponding loss of data in the frequency domain velocity spectrogram.

The purpose of this SDRD project was to increase the amount of data returned by PDV when the signal drops very low. Additionally, constraining the range of the PDV signal would allow PDV to be fielded with only one high-end (\$60–\$120K) oscilloscope per probe, instead of the customary two used to cover two different amplitude ranges. The two oscilloscopes are currently needed because the PDV return signal's voltage level cannot be known with certainty. Therefore, each oscilloscope is set to a different voltage scale to double the chances of getting good data back from a dynamic experiment. If AGC PDV could guarantee a specific output voltage range, then only one oscilloscope would be necessary, halving the cost per PDV probe.

This research is important because PDV applications allow experimenters to see multiple velocities (the particle velocity as well as the underlying free surface velocity). The potential application of

AGC PDV is as a diagnostic for use on many different types of dynamic experiments. AGC PDV could replace normal PDV in applications where signal drop-out is a concern. Currently, when the PDV signal drops low, experimenters extrapolate the missing data. Increasing the data returned from these types of experiments is highly desirable.

Project

Conceptual Design

Our design concept uses a telecommunications device called an SOA that provides both optical amplification and attenuation at very high speeds, <100 ps. Erbium-doped fiber amplifiers (EDFAs) have been used to amplify a PDV optical return signal. But EDFAs, having a response time of ~1 μ s, are not as fast as the SOA and, unfortunately, spectral hole-burning destroys the return signal. Spectral hole-burning occurs when two or more wavelengths of light are mixed together and the wavelength with the highest power level steals all of the EDFA's available gain, leaving no gain for the wavelength at the lowest power level. This characteristic of EDFAs is unacceptable with standard one-fiber PDV probes, since the unshifted and shifted signals are mixed together and the smallest signal is the signal of interest to experimenters. Figure 2 illustrates the SOA gain profile.

Figure 2. The SOA provides 15 dB of attenuation, as well as 20 dB of gain, depending on the amount of drive current

In our design, the SOA would gain up the PDV optical return signal if its return amplitude were too low and would attenuate the signal if it were too high, creating a system of AGC. Figure 3, a simplified drawing of our proposed solution in the time domain, also shows a corrected signal.

Figure 3. (a) Proposed solution in the time domain. The SOA is used for both gain and attenuation to level out the resulting shifted signal. (b) Proposed solution in the frequency domain shows reduced data drop-outs in the velocity spectrogram.

Figure 4. Existing PDV system block diagram

An existing PDV system is illustrated in Figure 4. The prototype PDV system with AGC (Figure 5) differs from the current PDV system in Figure 4 in several ways. The enhanced PDV system uses a new optical detector, a Terahertz Technologies, Inc. TIA-525. We needed to use this detector in its high-gain mode, which unfortunately limited its bandwidth to DC to 35 MHz. The SOA and its driver circuit and the AGC block were added. Finally, by including an optical filter centered at 1550 nm, we could block the SOA's broadband emissions.

Figure 5. Prototype PDV system block diagram with AGC

Prototype AGCs

To meet time and resource limits, we decided to use COTS evaluation boards in lieu of designing and fabricating custom PCBs. We obtained an SOA and a driver board and thoroughly evaluated them. Initially we were concerned that the SOA might wavelength-shift at higher gain levels, but tests showed that, at the power levels in the PDV's shifted leg, it did not wavelength-shift. (Further testing would be required if we were to move the SOA to the unshifted leg of the PDV system where it would receive higher input power.)

Next, the evaluation board was modified to increase its response time as much as possible. Finally, we developed AGC circuits using the best-fit COTS evaluation boards available. The digital and analog AGC circuits were developed in parallel.

Digital AGC

The digital AGC uses a COTS field-programmable gate array (FPGA) evaluation board with an on-board analog-to-digital converter (ADC) and a digital-to-analog converter (DAC). We found that the available selection of COTS FPGA evaluation boards with an on-board, high-speed ADC and DAC were too generalized for our needs; the voltage levels were not quite right, and the speeds were not high enough. The board that came closest to meeting our requirements was an Altera DSP Development Board, Statix II Edition with on-board 125 MSPS ADC and 165 MSPS DAC.

We thoroughly characterized the Altera board and proved that a digital AGC could work given careful component selection, electrical interfacing, and ADC/DAC matching. For a bench test, a signal was read by the ADC and piped into the FPGA using low-voltage differential signaling (LVDS), run

Figure 6. System setup used to do a side-by-side comparison of normal PDV with the analog AGC PDV

through a ROM lookup table, and output to the DAC via LVDS. System latency measured <160 ns using 100-MHz clocking. Since higher-speed FPGAs, ADCs, and DACs are available, this latency could definitely be improved. We originally aimed to correct the optical signal in real time within 10 ns, but we quickly realized that for the most prevalent type of signal drop-out, dynamic speckle events, a much slower correction of ~ 100 ns was more than adequate.

Analog AGC

We developed two analog AGC circuits using two operational amplifier evaluation boards to create an inverting amplifier, which we used to drive the modified SOA evaluation board. The higher-speed version of the analog AGC board had a response time of tens of microseconds with a gain of 10 dB. The lower-speed version had an increased gain response of 16 dB with a response time of 1 or 2 μ s. We tested the AGC first with good results and then went on to do a side-by-side comparison of normal PDV with AGC PDV. The test setup shown in Figure 6 was used for the side-by-side comparison of normal PDV with the prototype lower-speed, higher-gain analog AGC PDV.

Figure 7 shows the results of the side-by-side test in the time domain. Note that when the normal PDV (green) signal became small, the AGC PDV (blue) amplified the signal (with some overshoot due to its slow speed). When the normal PDV signal was high, AGC PDV attenuated the signal.

Figure 7. Time domain results of normal PDV (green) signal and AGC PDV (blue)

Figure 8 shows the time domain traces in Figure 7 aligned with their corresponding frequency domain velocity spectrograms below them. They are shown in the same scale. Note that when normal PDV drops very low in the time domain, its corresponding frequency domain velocity spectrogram (directly beneath it) shows data drop-outs. Note the black vertical bands and, most importantly, the gap in the lower red/orange sinusoidal signal, indicating loss of particle velocity data. While referring to the AGC PDV charts on the right, note that the upper green time domain plot does not show very small signals. Its corresponding velocity spectrogram shows blue vertical bands instead of the black bands evident on the normal PDV signal. The blue bands are the amplified broadband emissions from the SOA. These do not negatively affect the data, as they are easily removed with the in-house PDV analysis tool, pTool. Most importantly, note that there are no gaps in the lowest sinusoidal red/orange signal. All particle velocities are preserved. It should also be noted that the sinusoid in the AGC PDV resulted from the fact that the IPG laser was not optically well-isolated. The upper trace is a harmonic, thus the lower sinusoid is our signal of interest.

Figure 8. (upper traces) Time domain data are lined up and to scale with the (lower traces) frequency domain data for normal and AGC PDV data. The (right) AGC PDV amplified or attenuated the (left) normal PDV signal, restoring all velocity data.

Finally, we investigated whether or not we had actually improved the SNR in the frequency domain. The analysis yielded an SNR increase of approximately 15 to 20 dB. The noise floor of the AGC PDV data had risen 10–15 dB, while the peak rose 30 dB, yielding an overall increase of 15–20 dB in SNR.

The noise floor for PDV data is roughly –85 dB, and –70 to –75 dB for the AGC data. Figure 9, plots of the peaks, clearly shows a gain of >30 dB (32 dB, actually).

Figure 9. Velocity trace extracted from the two spectrograms utilizing the following method: Data were low-pass filtered at 150 MHz and resampled at 3.2 ns. The FFT spectrogram was computed from 750 to 780 μ s using an extended Hamming window of 4096 points. The lineout was extracted at the same time, 760.089 μ s.

Customization of the SOA driver circuit would enable meeting the target speed of 100 ns and would also improve the SOA's linearity, thus reducing the output signal variation. The Terahertz Technologies optical detector was not sensitive below –30 dBm. We would recommend using an APD for future work; for example, an APD similar to the Newport AD50APDir could be used. The dynamic laser slapper test met with limited success. It was difficult to slow the slapper down enough to give our circuit time to respond. At a slapper speed of <500 m/s, the SOA turned on but the data record

was only 20 μ s long, as opposed to the 4-ms data record used for lab-simulated data testing; therefore, there was not enough data to make the analysis worthwhile. Future work may include developing a much faster field-ready prototype to stabilize the SNR as well as to constrain the dynamic range.

Conclusion

We successfully demonstrated AGC of a PDV signal in real time using an analog circuit. A 15- to 20-dB increase in the SNR of the frequency domain data was achieved. We determined that the digital solution requires a custom PCB.

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FARADAY SENSORS USING FIBER BRAGG GRATINGS

Bruce Marshall^{1,a}

Fiber-optic sensors based on Faraday rotation are used in the weapons test community to measure large pulsed currents and large pulsed magnetic fields. They have generally been fielded in a one-fiber/one-data-point configuration. In this work, we demonstrated the use of fiber Bragg gratings (FBGs) to make *spatially resolved* measurements of magnetic fields using a single fiber. These sensors may find application in B-field symmetry measurements in high-current, pulsed-power applications such as Phoenix, and in high magnetic field measurements where the compact size and spatial resolution would be advantageous.

Background

Fiber-optic sensors based on Faraday rotation are generally capable of making two types of measurements: current and magnetic field. For current measurements, the fiber serves as the Faraday medium with a loop of fiber placed around the current, as shown in Figure 1 (Cernosek, 1994). The plane of polarization of linearly polarized light injected into the fiber is rotated through an angle given by $V_d \int B \cdot dl$, where V_d is the Verdet constant, B is the magnetic field, and l is the optical path length. The cumulative Faraday rotation is independent of the distribution of the current inside the loop, and may include the effects of fringe fields on the downloads outside the loop. If the measurement could resolve the magnetic field distribution around the loop, the current distribution could be determined to some extent, and the contribution of the fringe fields could be eliminated from the calculation.

Figure 1. Faraday effect fiber-optic current measurement setup

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Measurements of high magnetic fields—for example, on magnetic flux-compression generators—are usually made with a multimode, fiber-coupled, bulk optic probe (Figure 2). For megagauss fields, a low Verdet constant material such as fused silica gives appropriate sensitivity. Use of mode-filled, multimode step-index fiber eliminates Faraday rotation in the downleads. Polarizers define the active sensing region, and a short rod of fused silica or other material provides the rotation medium. These sensors have typically been 2 or 3 mm in diameter and about 30 mm long. Although relatively small, these sensors are nevertheless too large in some situations. The large diameter is required to allow collimation of the light so it can traverse the rod and be coupled efficiently into the return fiber. A sensor composed simply of single-mode fiber with no structure around it would occupy considerably less of the valuable high-field volume. If that sensor could also resolve the spatial distribution of the field, it could considerably improve the usefulness of the measurement.

Figure 2. Faraday effect magnetic field probe

An FBG consists of an optical fiber with a periodic variation in the refractive index (Kashyap, 1999). By choosing the index pattern appropriately, an FBG can be made that functions as a very-narrow-band reflector with very high efficiency. Wavelengths of light outside the reflection band are transmitted efficiently. A series or array of gratings with spectra centered at differing wavelengths and spaced sequentially along a fiber allows one to probe different sections of the fiber by selecting the appropriate wavelength. Because the Faraday effect integrates the rotation along the fiber, taking the difference of two measurements reflected from different locations gives a measure of the average magnetic field over the corresponding segment of the fiber. Grating arrays are available in arbitrary lengths and with arbitrary spacings between the gratings, and could be designed for measuring the magnetic field along a fiber loop around a current as shown in Figure 3, resolving quadrants or octants of the loop. One added benefit of such a probe is that birefringence in the loop can be measured by looking at the polarization state of light reflected from each grating.

Figure 3. Fiber loop with FBG array

In a similar manner, a magnetic field probe can be constructed by using two FBGs at different wavelengths to define the sensing region, as shown in Figure 4.

Figure 4. Magnetic field sensor formed by two FBGs

If, instead of multiple discrete gratings, a single grating is made in which the spacing varies as a function of position, a “chirped” grating is formed in which the location of reflection varies continuously with wavelength (Figure 5). Using a chirped grating, Faraday rotation occurs over the portion of the fiber traversed by a particular wavelength. The magnetic field can then be measured by differentiating

the Faraday rotation with respect to wavelength, with spatial resolution determined by the grating chirp and by the spectral resolution of the recording and analyzing system. The equivalent of a conventional bulk optic probe could be realized by using two wavelengths to define the sensing region.

Figure 5. Magnetic field sensor based on chirped grating

Project

To test the practicality of the idea of spatially resolved magnetic field measurements using FBGs, we purchased gratings and grating arrays of various types from several manufacturers (QPS Photonics, OFS Fitel, and O/E Land) and set up a system to generate and measure magnetic field distributions. The work was done in the C band (1535–1565 nm) to take advantage of equipment and gratings available in that region. Optical fiber is inherently an insensitive medium for Faraday rotation, and the λ^{-2} dependence of the Verdet constant of single-mode fiber presents a challenge for operation in the 1550-nm region. For a single fiber loop, approximately 4.4 MA are required to produce one fringe. To overcome this, we operated in the fractional fringe regime and used a solenoid driven sinusoidally at 200 Hz, combined with a differential receiver and curve fitting to obtain good quality data. Operation at a shorter wavelength could improve the sensitivity significantly, but components such as narrow-linewidth tunable lasers, broadband single-mode light sources, very narrowband filters, and fiber Bragg gratings are either unavailable or very much more expensive at short wavelengths.

The system used for the measurements is diagrammed in Figure 6. Light from a broadband amplified stimulated emission (ASE) source is filtered using an Echelle grating monochromator with 0.1-nm linewidth, and amplified to deliver about 10 mW to the system. A polarized circulator assures delivery of linear-polarized light, and allows us to use the return light to monitor the wavelength with an Ando AQ6330 Optical Spectrum Analyzer. Light from the middle port of the circulator is collimated, polarized, passed through a polarizing beam splitter (PBS), a non-polarizing beam splitter (NPBS), and a half-wave retarder, and focused into the FBG. On the main signal path, return light from the FBG passes through the half-wave retarder and NPBS to a quarter-wave retarder, a half-wave retarder, and a Glan-Thompson beam-splitting polarizer, which splits the light into two orthogonal states for delivery to a differential receiver. In the NPBS half of the return light is sent toward the PBS, which directs the orthogonal state to a power meter for polarization monitoring. Nulling this signal ensures a linear polarization state at the FBG.

Figure 6. System diagram for fiber Bragg grating Faraday sensor benchtop measurements

Polarization

As mentioned above, although many field applications would involve high magnetic fields and would thus produce multiple fringes, we made these measurements in the small-signal, fractional-fringe regime, for practical reasons. In a fractional-fringe Faraday rotation sensor, the polarization state is more critical than in a multiple-fringe system, where the data can be normalized using the local minima and maxima. Ideally, in a fiber Faraday sensor the circular states are aligned at the source, sensor, and receiver, thus ensuring that all linear states correspond to linear states at all three locations.

One way to achieve this would be to wavelength-multiplex a circularly polarized beam for monitoring polarization. This light would be circular at the launch. The return signal would be monitored and the system tuned to maintain it in the orthogonal circular state, thus ensuring a circular state at the sensor. To simplify the present system, however, we used the measurement light itself to monitor the polarization. The state immediately before the fiber is linear, with azimuth adjustable via a half-wave retarder. Since there are always two intersections of two great circles on the Poincaré sphere, there will always be two orientations of the linear launch state that correspond to linear states at the sensor. This condition is indicated when the orthogonal component of the return state, as monitored by the power meter, is minimized. After passing through the NPBS, the return state is converted to circular by the quarter-wave retarder, and the orientation of the two detector states is set by rotating the half-wave retarder to maximize the Faraday signal. This approach would not be practical in the field, where a Faraday modulation signal is not available before the experiment. For a multiple fringe system, the polarization control should align the circular states at the fiber input, FBG, and output beam-splitting polarizer, thereby ensuring optimum modulation depth and allowing the initial phase to be set as desired.

In operation, the polarization was very stable, and the half-wave retarder needed only minor adjustments of less than 5 degrees as the wavelength was scanned to interrogate different positions in the sensor. For multiple-fringe operation, this amount would be negligible, and it actually affected the data in the fractional-fringe measurements by a very small amount. The polarization state is affected primarily by birefringence in the sensor downlead, which can be minimized by twisting the fiber. The fibers in the present experiment were not twisted. Birefringence in the chirped grating was very small and in the grating array was somewhat larger, but was still small enough not to present a problem.

The magnetic field was generated by a coil with 400 turns on a 0.5-in.-diameter spool 0.5-in. long, driven by a sine-wave burst at 200 Hz. The duty cycle was minimized to reduce heating, and the drive current was monitored and recorded. The coil was mounted on a stepper motor translation stage under computer control. The magnetic field was measured as a function of position using an F.W. Bell 5180 Hall-effect gaussmeter while the magnet was moved along the stage.

The location of reflection in the grating and grating array as a function of wavelength was measured interferometrically by adding a reference path to the existing optical system and replacing the Echelle grating with a broader tunable bandpass filter (Figure 7). The variable delay (General Photonics VDL001) has 50- μm resolution on a Vernier scale. The peak wavelength was measured using an Ando spectrum analyzer.

For reference, Faraday rotation was measured at 1550 nm using an unchirped grating while the magnet was moved a distance of 130 mm along the fiber. This same measurement was repeated with the chirped grating, with very similar results, thus demonstrating that, at least for weak fields, the chirped grating does not affect the effective Verdet constant. The magnet was then placed near the center of the grating or grating array, and measurements were made at series of wavelengths to interrogate the

corresponding locations. The total useful length of the chirped grating was only 16 mm, so the magnet was moved and the measurement repeated several times to trace out more of the field distribution and give some idea of the measurement's repeatability. The grating array was on 20-mm centers and only a few points lay within the main part of the field. The Faraday rotation angles versus position are shown in Figure 8.

Figure 7. System setup with interferometer for measuring grating locations

Figure 8. Integrated Faraday rotation vs position

The best estimate of the Verdet constant for the optical fiber used in the FBGs is obtained from the NIST2 model for the Verdet constant in germanium-doped silica fibers (Equation 1) (Rose, 1997) as:

$$\mu V (\mu\text{rad} / A) = V_H v^2 + V_{H2} v^4, \quad (1)$$

where

$$V_H = 1.887 \times 10^{-35} \mu\text{rad} / (A * \text{Hz}^2),$$

$$V_{H2} = 7.80 \times 10^{-66} \mu\text{rad} / (A * \text{Hz}^4),$$

and v is the optical frequency.

At $\lambda = 1550 \text{ nm}$, $\mu V = 0.72 \mu\text{rad} / A$ and $V = 57.1 \mu\text{rad} / \text{meter-Gauss}$.

Figure 9 shows the integrated B-field calculated from the Faraday rotation using the NIST2 model for the Verdet constant.

The Hall-effect gaussmeter measurements were integrated and are also plotted in Figure 9. They are about 6% lower than the optical measurements. Figure 10 shows the magnetic field obtained by differentiating the results in Figure 9.

Figure 9. Integrated magnetic field vs position

Figure 10. Magnetic field vs position

As can be seen from the data, the wavelength-scanned measurements are very consistent with the single-wavelength, physically scanned measurement and are in reasonable agreement with the Hall-effect measurement. The ~6% discrepancy may arise in part from the nonuniform magnetic field being sampled by the relatively large cross section of the Hall probe relative to the 9- μm optical fiber core.

Practical Systems and Recording Schemes

The FBG Faraday sensors use wavelength multiplexing to measure the magnetic field integrated over multiple segments of a single fiber. The recording system must therefore either acquire continuous records of multiple channels simultaneously, or else time-multiplex the data onto a single recording channel. A spectrometer-coupled streak camera would be well-suited if the spectrum lies within the streak camera response range. Use of short wavelengths would have the advantage of better sensitivity due to the higher Verdet constant, but gratings and light sources may be more difficult to obtain. In the C band, the straightforward solution would be to use multiple diode lasers for the source, and a Dense Wavelength-Division Multiplexing (DWDM) demultiplexer connected to multiple detectors and oscilloscope channels (Figure 11). The number of channels would depend on the spatial resolution required. Single-channel, wavelength-to-time-multiplexed recording schemes are also possible, but require a tradeoff between sampling density in the time and space domains.

Figure 11. Multichannel recording system with demultiplexer

Another challenge involves matching the grating array spacing to the application. For example, if the measurement is to resolve quadrants in a fiber loop, five gratings would be required to define the four quadrants, and the overall length of the array would have to match the circumference of the loop fairly well. Grating locations must be carefully measured.

There is also some concern about the effect of high magnetic fields on FBGs. The most likely effects would be change in grating reflectivity and a polarization-dependent wavelength shift of the grating's reflection spectrum. These are probably avoidable by refraining from using spectrally narrow gratings. In a chirped grating, there may be polarization rotation due to the opposing refractive index shifts induced by the magnetic field and coupled with the chirp. Depending on the sign of the chirp, this chirp-dependent rotation would add to or subtract from the ordinary rotation seen in the bulk of the sensor. Our study was inconclusive on this question, indicating an effect that should be obvious, even at low fields, but we were unable to find evidence of any such effect experimentally.

Conclusion

We have shown that it is possible to make spatially resolved measurements of a magnetic field using FBGs and grating arrays. Measurements of a magnetic field using different types of gratings are very consistent and also agree well with conventional magnetometer measurements. FBGs and the system optical components are commercially available, although custom gratings may be required for specific applications. There is some question about the effect of very high magnetic fields on FBGs that would be best answered through further study. Overall, it appears that these sensors are well worth considering for current and magnetic field measurements where spatial resolution and compactness are important.

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MICROWAVE DIAGNOSTIC

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The use of microwave and millimeter-wave probes in shock physics experiments offers the potential to probe quasi-DC conductivity of a material on relevant timescales. In addition to potentially offering valuable conductivity information, these probes are well-suited to dynamic experiments and sample surfaces behind a dielectric window. We sought to determine the window of utility for microwave reflectance as a phase-change diagnostic using benchtop experiments on tin passing through the liquid-solid phase transition. The results were encouraging, with changes noted in the reflectance at approximately 10 and 16 GHz of approximately 5% on melting.

Background

The study of metals under shock is of great importance to the NNSA. A variety of probe technologies are available to determine velocity, temperature, pressure, and other properties of shocked metallic samples. A notoriously difficult property to monitor during dynamic experiments is the phase of the material, required for accurate phase diagram construction of the material under study. Phase changes, such as solid-liquid (melting) or solid-solid, are almost always accompanied by changes in other properties that could potentially be measured, such as acoustic impedance, density, optical reflectance, or quasi-DC conductivity.

Quasi-DC conductivity is best probed at frequencies below a few terahertz, at which point vibrational signatures can start to manifest and confound the measurement. For this reason, microwave reflectometry was investigated as a phase-change diagnostic potentially well-suited to shock physics. To date, microwave measurements have typically used interferometry to determine position and/or velocity of the shocked sample (Luther, 1993; Maron, 1980).

Project

The test apparatus was designed with an eye toward the requirements of dynamic experimentation, where it is important to physically separate expensive recording equipment from the event itself by using long coaxial signal cables (those over 6 ft long) or fiber-optic links. The fiber-optic links are most useful at frequencies above a few GHz, where attenuation along long cable runs can be prohibitive.

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This consideration led to the use of a comb-pulse generator as a source. We selected the Picosecond Pulse Labs model 7124. The comb generator is driven by a 2-GHz sine wave and generates a train of impulses at the repetition rate of the input, but with many overtones. The input signal can be generated with readily available sources and amplifiers, and the output of the comb has frequency content to beyond 50 GHz. The penalty for this wide-band signal generation using a low-frequency input is higher phase and amplitude noise than one would have from a single-frequency source. For this experiment, though, the noise was acceptable. For future work, as will be shown below, single-frequency sources may be a better choice, due to noise and antenna optimization considerations.

The antenna design was guided by the need for (a) versatility over the frequency range generated by the source, (b) directionality, and (c) the desirability of an inexpensive, consumable antenna that could ultimately be used in a dynamic experiment. The only suitable design that satisfied all of these criteria appeared to be a discone antenna (Kandoian, 1943) with a parabolic reflector to collimate the radiation. The discone antenna is widely used for wide-band signal transmission and reception. With an approximate 5:1 frequency window, it fell somewhat short of the “ultra-wide-band” classification, but was acceptable for the frequency-domain applications.

The antennas were designed and fabricated to operate from 10 to 50 GHz, with the longest wavelength being $\lambda_{\text{max}} = c/10 \text{ GHz} = 3 \text{ cm}$. Each antenna comprised a 0.375-in.-diameter brass disc soldered to the center conductor of a 0.085-in. solid copper coaxial cable. The solid brass cone had a bottom diameter of 0.375 in., and a height of 0.375 in. to the top of the hole from which the coaxial insulator protruded. Net half-angle was approximately 22 degrees. Brass was chosen because it can be soldered directly to the copper of the coaxial cable. One of the antennas is shown in Figure 1.

Figure 1. The brass discone on the end of a short length of copper coaxial cable, with a female SMA connector on the left end

Parabolic reflectors of 2-in.-diameter were selected, in the hope they would be large enough to give a well-collimated beam from the discone antenna, yet be compact enough to be fielded on an actual dynamic experiment. The antenna/reflector configuration is shown in Figure 2. During the setup and data collection, though, the collimation was determined to be less than ideal, likely because the small-source approximation was not well-satisfied by the discone centered in the 2-in. reflector. Nonetheless, cross talk between transmit and receive antennas was below the dynamic detection limit, and stray reflections were easily absorbed with microwave-absorbing foam.

Figure 2. The brass disc antenna mounted in the 2-in. parabolic reflector used for the benchtop experiment

For the experiments, the received signal was mixed down to frequencies of a few gigahertz for transmission using longer cables and for recording. The specific harmonics used were the 5th and 8th of the fundamental, typically at 10.4 and 16.6 GHz. The selection of these harmonics minimized the amount of new equipment required to be purchased. Critical to the operation of the system was the incorporation of band-pass filters to select the harmonics, since intermodulation products in the mixer will create significant cross talk between adjacent harmonics.

The transmission and receiving circuitry are shown schematically in Figure 3. Generally, modules connected by short coaxial cables were used.

Figure 3. Schematic diagram of the transmit (TX) and receive (RX) electronics for the benchtop experiment

The change in microwave reflectance upon melt was measured using tin, which melts at 232°C , and hence is well-suited for benchtop work. The tin sample was mounted in an aluminum block that was heated by cartridge heaters, and the temperature was monitored by a thermocouple and logged using a datalogging multimeter. The antennas were mounted above the sample and aligned to maximize the collected signal when the sample area was masked with a piece of microwave-absorbing foam with a hole cut in it. A schematic diagram of the experiment is shown in Figure 4.

Figure 4. Schematic diagram of the benchtop experiment. The tin sample is confined to a 2-in.-diameter recess in the aluminum block, and the aluminum is masked by RF-absorbing foam.

The melt experiment was conducted with no window over the tin surface. The sample was heated to above its melting point, then the surface was “skimmed” to remove highly oxidized material. After skimming, the foam mask was replaced and the measurement started with a cooling cycle. The temperature was ramped up and down several times while temperature and oscilloscope records were logged.

Reflectance data were collected with a Tektronix 6124C oscilloscope, using the full 40-GS/s sample rate and 12-GHz bandwidth. The scope was triggered approximately every two seconds by an acquisition computer, which then downloaded the record, Fourier-transformed the data, and measured the area under the two frequency peaks as a measure of the intensity of the signal at that frequency. These two area values are the values logged for that time. Data were taken in this way to best simulate

the record on a dynamic experiment, where the fast Fourier transform window can be dynamically adjusted during post-processing. Ultimately, however, the desired measurement is the time at which the material changes phase, which would be determined using a spectrogram of the scope record to measure amplitude as a function of time.

Three successive iterations of this experiment are shown in the plots in Figure 5. The gray horizontal lines show the melt temperature, as recorded with the thermocouple; there was an offset between the recorded melt temperature and the accepted melt temperature for tin of 232°C , possibly due to the fact that the thermocouple was near the edge of the aluminum block, away from the heating cartridges. The phase changes are marked by a temporary decrease in the slope magnitude of the temperature versus time curve, indicating the latent heat of melting.

Figure 5. Plots of three successive experiments, cycling the tin through solidification and melting while monitoring the reflected microwave signal at two frequencies

For both the 10.4- and the 16.6-GHz signals, there appeared to be change in the measured reflectivity as the sample was melting and freezing. In both cases, the reflectivity of the liquid phase appeared to be lower than that in the solid phase, as one would expect in the case of a disordered (liquid) metal, compared to a more well-ordered solid metal with higher DC conductivity.

Experiments using a 1-in.-diameter lithium fluoride window over the sample gave inconclusive results, possibly because of bubble formation and scattering under the window, or because of scattering from the edges of the window, which were not themselves masked by microwave-absorbing foam.

Conclusion

The potential utility of microwave reflectometry as a phase diagnostic for shock physics was investigated, and effects on the order of 5% changes in reflectance were measured during the melting of tin on the benchtop. Future work could focus on single-frequency or few-frequency techniques, rather than the wide-band techniques investigated here. Compact, directional antennas can readily be printed on circuit boards to achieve high gain in a one-time-use antenna.

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