MASTER

UCRL-5000
PREPRINT

CONF-810810-4

CONFIDENTIAL - UNCLASSIFIED
EXCLUDED FROM AUTOMATIC DOWNGRADING AND DECLASSIFICATION

R. E. Smith
D. L. Hutton
L. M. Johnson
J. C. Lister

This paper was prepared for publication
in the Journal of the Society of Nuclear
Instrumentation Engineers, 1981, by
the Instrumentation Group, Lawrence Livermore
Laboratory, California.

May 14, 1981

Lawrence
Livermore
Laboratory

This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

Radiation-to-light converters for nuclear environments:
Near-infrared emitters

F. Roeske, M. D. Rotter, and P. M. Calavan

Lawrence Livermore National Laboratory, University of California
Livermore, California 94550

S. S. Lutz

EG&G, Goleta, California 93017

Abstract

The use of fiber optics in the diagnosis of hostile nuclear environments has brought about a pressing need for the development of suitable radiation-to-light converters. These converters must meet stringent diagnostic requirements of linearity and time response while having a wavelength of emission that is compatible with transmission over relatively long lengths of optical fibers. In this paper we describe initial investigations of two, near-infrared-emitting semiconducting materials--CdTe and GaAs. Data are presented on the wavelength of emission, linearity, time response, and relative efficiency of these semiconductors. Most data were taken at 77 K because of a dramatic increase in efficiency at this temperature. Measurements show that the intensity-vs.-dose curves are linear over several decades of useful input levels. Measurements indicate that GaAs has an efficiency and time response that promise to be useful in diagnostic systems. Radiation-damage effects are briefly discussed. Finally, the future direction of these investigations and possible applications in nuclear-test diagnostic systems are reviewed.

Introduction

Scintillators have long been used as a reliable, convenient means of detecting nuclear radiation. When used to convert nuclear radiation to light suitable for transmissions over long distances (1 km) of optical fibers, conventional plastic scintillators commonly used for detection of nuclear radiation have serious shortcomings, as follows:

- Such scintillators generally have a peak emission in the blue region of the spectrum, an extremely high loss region in fiber-optic systems (Figure 1).
- For applications requiring large bandwidth, multi-mode graded-index fibers generally have an index profile that is peaked for minimum dispersion in the near-infrared region of the spectrum. Because this profile is usually narrowly peaked, bandwidth deteriorates rapidly as the wavelength is moved off this peak.
- Material dispersion for optical fibers becomes much lower as wavelength increases.
- Finally, radiation sensitivity of optical fibers generally decreases for longer wavelengths.

These considerations strongly suggest the need for a scintillator with a peak emission in the near-infrared region of the spectrum if the scintillators are to be used for the sole light source in long, fiber-optic transmission systems. There are, however, limitations on how far we can push a system into the infrared. The largest limiting factor at present is the detector response. Present photocathode technology limits the maximum useable wavelength to about 880 nm. Keeping this limitation in mind, we will discuss research directed at finding suitable radiation-to-light converters (RLC's) with peak emissions in the spectral range from 700 to 880 nm.

The desired properties of an RLC will vary considerably depending on the application. Though exceptions exist, the following list represents a reasonable set of desirable qualities for an RLC that is to be used in the nuclear testing diagnostic system at Lawrence Livermore National Laboratory (LLNL). An RLC should:

- Have a peak emission within the spectral range of 700 to 880 nm.
- Have a fast time response (≤ 2 ns).
- Be linear over at least two decades.
- Be reasonably efficient (on the order of the efficiencies of conventional blue-emitting scintillators).
- Not suffer degrading radiation damage on the time scale of the measurement.

DISCLAIMER

This document contains information which is the property of the United States Government and is loaned to your agency. It is to be used only for the purposes specified and is not to be distributed outside your agency without the express written consent of the United States Government. This document and its contents are not to be used for advertising or promotional purposes, nor for the purpose of reproducing or copying for general distribution. The views and opinions of authors included herein do not necessarily state or reflect those of the United States Government or any agency thereof.

JHS

Figure 1. Typical graded-index optical-fiber attenuation as a function of wavelength. Conventional scintillators are generally found in region A. Low loss region B is the main region of interest for fiber-optics systems while region C is of interest for certain special cases.

Other laboratories¹ have strong programs for the investigation of organic scintillators as possible RLC's for nuclear-test diagnostic systems. This paper describes the initial investigation of CdTe (a II-VI semiconducting compound) and GaAs (a III-V semiconducting compound) as possible RLC candidates.

We chose semiconductors because they have their peak wavelengths of emission in a very desirable region of the near-infrared and because of reported high values for internal efficiencies.^{2,3} CdTe has a bandgap of 1.5 eV at 300 K.⁴ If radiative band-to-band transitions are the dominant recombination mechanism for excited electron-hole pairs, the photons generated will have a wavelength of 826.6 nm. Likewise GaAs has a 300 K bandgap of 1.43 eV, corresponding to a photon wavelength of 867.1 nm. Bogdankevich et al.⁵ showed that the efficiency of GaAs samples cooled to 80 K can be as high as 30%; i.e., a value very close to the theoretical maximum.

CdTe and GaAs are direct semiconductors. For direct semiconductors, the maximum in the valence band and the minimum in the conduction band are at the same value in k -space. At this point, radiative combinations can occur without involving a phonon to conserve momentum as is required by indirect transitions. The direct transition is a first-order process and has a high probability of occurrence. A high probability of transition implies that these semiconductors have a potential for very fast transitions.

Experimental

Data were taken on three different measurement systems. The first system measured spectral response, efficiency, linearity, and (within the system limits) time response. A 1.2-ns pulse of 100- to 600-keV electrons from a Model 706 Pebetron impinges on the semiconductor sample, which is located in an optical Dewar in which the sample temperature can be varied from 77 K to 300 K. The number of electrons reaching the sample is varied by adjusting a variable iris or adjusting the nitrogen pressure in a beam guide tube. The light output from the sample is focused into a 1/4-in. fiber bundle and transmitted to either a spectrometer or bandpass filter, depending on intensity and resolution considerations. The spectrally filtered light is then detected by a high-speed photomultiplier tube (PMT) and recorded on a Tektronix 7104 oscilloscope. Spectra are obtained by using a Tracor Northern Diode Array Scanning Spectrometer (DARSS) and an optical multichannel analyzer. The electron beam is monitored by a Paraday cup. Dosimetry was performed on the Pebetron system by calibrating Paraday cup voltage against

the reading of calibrated radiachromic film. After this procedure was completed, the Faraday cup voltage could be related directly to a dose reading. It should be pointed out that the quantity measured here is the dose in the radiachromic film. Because of differences in density between the film and semiconductors, different volume excitations will occur, so the numbers we give here represent only an approximation to the true dose absorbed in the semiconductor.

A second measurement system was used primarily to determine the impulse response of the sample. This system uses the EGS LINAC at Santa Barbara, California. A continuous stream of 40-ps pulses of 6-MeV electrons are used to irradiate the sample, which is located in an optical Dewar. The light output is transmitted by means of a 1-mm-diameter optical fiber to a spectrometer-PMT or filter-PMT combination. The data are recorded using a sampling system and PDP8 minicomputer.

The third system, also used primarily to determine impulse response, uses a Candella high-powered, mode-locked laser as the exciting source. A pockel cell selects a single, approximately 10-ps pulse of 605-nm light with about 20 MW peak power as the input impulse. The light output of the sample, which is once again mounted in an optical Dewar, is transmitted by a 1/4-in. fiber bundle to a SPEX model 1404 double spectrometer. This particular spectrometer was chosen for its high stray-light rejection ratio (10^{14} , at 0.5 nm from 505-nm wavelength). When using such an intense laser beam, this stray-light rejection ratio is necessary to insure that the detector will see no part of the input pulse. The spectrally filtered signal is detected by a high-speed PMT and recorded on a Tektronix 7104 oscilloscope.

Single crystal samples of N-type CdTe with 111 orientation were obtained from II-VI Corp. The sample dimensions were 1 by 1 by 0.2 cm. CdTe samples used for laser excitation were etched in a solution of 1 part bromine to 3 parts methanol.

Undoped GaAs samples (in the 10^7 to 10^8 ohm-cm range), obtained from Metals Research, Ltd., as 0.38-mm-thick wafers, were cut into 1-cm² samples. Li-doped samples were formed by vapor depositing Li on one surface of the undoped wafers and heating for 24 hours at 250°C.

Results and discussion

The normalized spectral emission curves for CdTe and GaAs are shown in Figures 2 and 3, respectively, for different modes of excitation and different temperatures. As the temperature of a semiconductor increases, the lattice expands and the oscillations of the atoms about their equilibrium lattice points increases. This expansion leads to a change in the energy gap. This effect is evident in both the CdTe and GaAs samples with a shift of 0.074 eV and 0.068 eV, respectively.

GaAs is one of many semiconductors whose temperature dependence of the gap is given by the empirical relation⁶:

$$E_g(T) = E_g(0) - \frac{17}{T + 8}, \quad (1)$$

Figure 2. Spectral emission from CdTe peak. Amplitudes are normalized to 1.

Figure 3. Spectral emission from GaAs peak. Amplitudes are normalized to 1.

where $E_g(0)$ is the value of the energy gap at zero kelvin, and α and β are constants. Using Eq. (1) with appropriate values of α and β for GaAs,⁶ we obtain a calculated energy gap change of 0.083. It is not surprising that this value differs from our measured value because the fall-off of our system response at these wavelengths makes it difficult to obtain an accurate spectrum at 300 K. We should expect the correct spectrum for GaAs at 300 K to peak at 886 nm.

Besides shifting the band edges, the increased motion of the atoms at higher temperatures also broadens the energy levels. This effect broadens the output emission curves as can be easily seen in the case of CdTe. CdTe has a spectral FWHM of 19 nm at 300 K, whereas at 77 K the FWHM is only 10 nm. Again, it is difficult to obtain an accurate comparison of all GaAs data.

The spectral curves of Figures 2 and 3 have been normalized with respect to their own peak values. However, in reality great differences exist in peak amplitude between 300-K and 77-K data. The external efficiency has an exponential dependence⁴:

$$I_{\text{ext}} = I_0 \exp(-T/T_0) \quad (2)$$

CdTe has 10 times the peak output at 77 K as at 300 K, and GaAs increases by a factor of 12.5.

For both GaAs and CdTe, the 77-K emission spectrum for laser excitation is shifted toward higher energies than that for electron excitation. The laser excitation peak agrees very well with accepted values of the energy gap.^{7,8} This discrepancy between laser and electron excitation most probably arises because of self-absorption. Laser excitation at 605 nm will only penetrate the sample to a depth of about 1 μ m. Most of the light generated, therefore, will be able to escape with minimal attenuation. Electrons, however, penetrate the sample from several tens of micrometers to a few hundred micrometers, depending on the electron energy. A large fraction of the electron energy is deposited deep below the surface, and the light generated will be subject to a large attenuation for high-energy photons because of a large absorption coefficient. When the unattenuated spectrum is folded with the absorption curve, shifts in the peak output spectrum on the order of what is experimentally observed here can be obtained. Confirmation of this will have to await measurement of a Pebelectron energy-spectrum and of CdTe and GaAs absorption curves.

Linearity curves (intensity vs dose) for CdTe and GaAs are shown in Figures 4 and 5, respectively. Both samples show linear behavior over the lower two decades of measurement. Deviations from linearity occur only at the higher dose levels at the onset of stimulated emission. The magnitude of the superlinear "jump" in the curve depends on the relative position of the narrow superradiant spectrum (which is red shifted with respect to the fluorescence peak, see Figure 6) with respect to the transmission peak of the bandpass filter used in the measurement. If the filter transmission peak and the

Figure 4. Intensity vs dose for CdTe at 77 K.

Figure 5. Intensity vs dose for GaAs at 300 and at 77 K.

Figure 6. Spectral emission of GaAs in the superradiant mode.

superradiance peak overlap reasonably well, a large jump is observed. Likewise, a small jump is observed when the overlap is poor. In all cases, the output reaches a saturation level where the slope of the curve levels off or increases in a sublinear manner.

The absolute efficiency that is determined in the linear portion of the curve for CdTe is 3.0×10^{-11} J/cm²·sr·rad. The value determined for GaAs is 3.5×10^{-11} J/cm²·sr·rad taken at $\sim 10^4$ rad.

Time-response data for GaAs at 77 K is given in Figure 7. The Febetron system has a rise time of 700 ps and a FWHM of 1.5 ns. GaAs output taken from this system has a 1.0-ns rise time and a 1.6-ns FWHM (unfolded). This response is similar to that observed in p-type samples of GaAs with a hole density of $\sim 5 \times 10^{17}$ /cm³ when subjected to electron injection.⁹ If this is true, increasing the acceptor density should decrease the lifetime of the excited electron and cause a faster response. This effect was observed when a Li-doped sample was subjected to the same electron excitation (Figure 7c). The system response observed in this case indicates that we have improved the time response; however, by how much is uncertain.

To further investigate the response of undoped GaAs, we excited a sample with a ~ 10 ps laser pulse of 605-nm light. The results of this excitation can be seen in Figure 7e. Again we are system-limited in response, indicating a very fast response of GaAs. This response is indicative of a pure band-to-band transition.¹⁰

There seems to be a fundamental difference in the time response of GaAs when excited by photons and when excited by electrons. The damage threshold for displacement of Ga and As atoms is 233 and 256 keV, respectively.¹¹ Therefore, this effect might be explained by a radiation-induced perturbation in the crystal lattice. Another possibility is that we are observing a surface effect in the laser excitation, whereas the electron excitation is a bulk effect. Our data are insufficient to allow determination of the cause of the difference in response times. For this, excitation with a faster electron source and a careful time-response-vs-temperature measurement will be necessary. These measurements are planned for the near future.

Febetron excitation of CdTe at 77 K (see Figure 8) leads to a time response of about 11 ns (1/e decay time) and 51.5-ns rise time (Figure 8a). This result agrees reasonably well with the electron excitation data taken on the LINAC (Figure 9, a and d). Laser excitation data give a response of about 1 ns FWHM with a 600-ps rise time at 117 K (Figure 8b). This response is not inconsistent with the high-energy electron response obtained after several minutes bombardment (Figure 9c). The high-energy electron data indicate that we are most probably quenching the sample by introducing radiation defects that anneal at 300 K. This speculation, coupled with the reasonable agreement between the laser-excited sample and the high-energy electron-damaged sample, suggests the laser-excited response for CdTe arises from the quenching that results from surface

Figure 7. Time response of GaAs at 77 K. (a) Febetron system response 1.5-ns FWHM. (b) GaAs undoped, electron excitation, 2.2-ns FWHM. (c) GaAs, Si-doped, electron excitation, 1.5-ns FWHM. (d) Laser system response, 650-ps FWHM. (e) GaAs, undoped, laser excitation, 650-ps FWHM.

Figure 8. Time response of CdTe at 117 K. (a) Electron excitation, 1/e decay time of 11 ns. (b) Laser excitation, 1-ns FWHM.

Figure 9. Effect of radiation on CdTe time response at 77 K. (a) Virgin sample, 1/e decay time of 5 ns. (b) After several minutes bombardment with 6-MeV electrons at 16-A beam current, 1/e decay time of 3.4 ns. (c) After several more minutes bombardment, 1.4-ns FWHM. (d) After annealing at 300 K for 24 hours, 1/e decay of 8.5 ns.

recombination or some other surface effect. Again, present data are insufficient to confirm this.

Conclusions

The peak wavelength of emission of GaAs is excellent for use in fiber-optic systems. The time response suggests a potential for use in very fast measurements (≤ 300 ps) and certainly is adequate for moderately fast signals (22 ns). Further work will be done to understand and maximize the time response with respect to the type and level of excitation, the amount (if any) of doping, and the sample temperature.

Although the GaAs intensity-vs-dose response is linear over several decades, the absolute output falls short of what we would like. Calculations show that for lens coupling into a 50- μ m graded-index fiber and with a dose of 10^4 rads, on the order of 1 μ W would reach an uphole detector. This is approximately 2% of the power received when using Cerenkov-generated light in a fiber.¹² For GaAs to be useful in fast nuclear-test diagnostic systems, 100 to 1000 times as much light will have to be transmitted to the detector. We believe there is a good probability of achieving this by developing more efficient samples, improving sample geometry, and optimizing coupling to the fiber. Work in all of these areas is presently underway.

In systems requiring only moderately good time resolution and accuracy, GaAs could be useful as it is. Cooling the sample to 77 K in the field will be annoying but not prohibitive. An obvious goal is to increase the amount of light coupled into the fiber to the degree that room-temperature operation is acceptable.

CdTe exhibits an excellent peak wavelength of emission and good linearity characteristics. The time response we have observed of the crystals under electron excitation is too slow to be of much interest. For CdTe crystals, subjected to laser excitation and for electron-damaged CdTe crystals subjected to electron excitation, time responses are fast enough to be of interest. Possibly CdTe doped with a large acceptor concentration, or the use of purer CdTe or CdTe with fewer defects, dislocations and the like, will prove to be much faster. We intend to investigate these possibilities.

The absolute power output for CdTe exceeds that of GaAs by a factor of 17. CdTe at this value is beginning to approach a range of usefulness. Improvement of the time response of CdTe and an increase in the output power coupled into an optical fiber would make CdTe a very useful RLC.

Acknowledgment

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

References

1. EG&G/LANL Support Program Project Summary, EGG 1183-2418, p. 7 (Jan. 1981).
2. O. V. Bogdankevich, N. A. Borisov, I. V. Kryvkova, and B. M. Havroshin, Soviet Physics-Semiconductors, **2**, p. 845 (Jan. 1969).
3. V. S. Vavilov, E. L. Nolle, and S. N. Makimovskii, Soviet Physics-Solid State, **7**, p. 1253 (Nov. 1965).
4. J. I. Pankove, Optical Processes in Semiconductors (Prentice-Hall, Englewood Cliffs, N.J., 1971), p. 413.
5. S. M. Sze, Physics of Semiconductor Devices (Wiley, New York, 1969), p. 30.
6. Y. P. Varshni, Physica, **34**, 149 (1967).
7. V. S. Vavilov, E. L. Nolle, V. D. Egorov, and S. I. Vintovkin, Soviet Physics-Solid State, **6**, 1099 (1964).
8. N. Holonyak, Jr., M. R. Johnson, J. A. Rossi, and W. O. Groves, Applied Physics Letters, **12**, 151 (1968).
9. H. Rieck, Semiconductor Lasers (Macdonald, London, 1970) p. 43.
10. W. P. Duke, Physics Review, **132**, 1998 (1963).
11. R. Baulelein, Radiation Damage in Solids (Academic Press, New York, 1962) p. 358.
12. P. B. Lyons, Los Alamos National Laboratory, private communication (1981).

DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial products, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government thereof, and shall not be used for advertising or product endorsement purposes.