1 of 1

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Laser Absorption Spectroscopy System for Vaporization Process Characterization and Control

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Abstract

In support of the Lawrence Livermore National Laboratory's (LLNL's) Uranium Atomic Vapor Laser Isotope Separation (U-AVLIS) Program, a laser atomic absorption spectroscopy (LAS) system has been developed. This multi-laser system is capable of simultaneously measuring the line densities of ^{238}U ground and metastable states, ^{235}U ground and metastable states, iron, and ions at up to nine locations within the separator vessel. Supporting enrichment experiments that last over one hundred hours, this laser spectroscopy system is employed to diagnose and optimize separator system performance, control the electron beam vaporizer and metal feed systems, and provide physics data for the validation of computer models. As a tool for spectroscopic research, vapor plume characterization, vapor deposition monitoring, and vaporizer development, LLNL's LAS laboratory with its six argon-ion-pumped ring dye lasers and recently added Ti:Sapphire and external-cavity diode-lasers has capabilities far beyond the requirements of its primary mission.

Introduction

Successful commercialization of the U-AVLIS technology is hinged upon optimized separator system designs, accurate and reliable diagnostics to monitor and control system efficiency, and a solid understanding of the process economics. Contributing in all three of these areas, the LAS systems operated in support of LLNL's AVLIS program have been, and will continue to be, paramount tools in realizing the goal of the deployment of a U-AVLIS plant.

The AVLIS program initially activated a LAS system approximately fifteen years ago. In the well over one hundred major experiments since, this diagnostic system has provided data to further our understanding of the vapor and photoionization physics of the AVLIS process and has provided the experimenters with a means of evaluating system performance. The U-AVLIS program currently has two, nearly identical LAS laboratories. For simplicity sake we will concentrate on the Separator Demonstration Facility LAS Laboratory. The goal of this laboratory is to provide a measure of the uranium and iron vaporization rates during long-duration enrichment experiments and to be flexible enough to support a large and ever-changing host of vapor physics and photoionization experiments. This LAS system has continued to evolve throughout the years and has recently been used for closed-loop control of the uranium vaporizer and feed systems and has supplied data to tune economic models of plant architectures.

The Basic Technique

Laser atomic absorption spectroscopy is a mature and widely used analytical tool in physics and chemistry research. Although there are many variations of the technique, most involve sweeping the wavelength of a narrow line-width tunable laser through an atomic absorption transition of the species of interest. The laser light is then delivered to a vessel where, after passing through an atomic vapor sample, the transmitted light is measured by a photo-detector. The resulting absorption signal is recorded along with that from a frequency standard which is employed to monitor the laser sweep. By correlating the data from the laser sweep and the detector output, information such as the density and the kinetic temperature of the vapor can be determined.

The U-AVLIS LAS Optical Systems

At the heart of the LAS laboratory is six argon-ion-pumped ring dye lasers. Four of which are configured for operation in the mid-visible portion of the spectrum and two operate in the blue. Employing R110, R6G, and R2 dyes (depending on the requirements of the experiment), the visible lasers can be used to access atomic transitions from approximately 5300 vac Å to 6400 vac Å. Using S3 dye, the blue lasers can range from 4200 vac Å to 4650 vac Å. The recently added Ti:Sapphire and external-cavity diode-laser systems expands the laboratory's capabilities into the UV and infrared portions of the spectrum allowing the probing of atomic and molecular transitions of countless materials.

Figure 1. Simplified optical system block diagram showing a typical ring dye laser configuration.

The configuration of a typical ring dye laser system, the layout of its associated diagnostics, and its position within the remainder of the optical system is depicted in figure 1. Prior to an experiment, each ring dye laser is tuned, using a scanning Michelson interferometer based wavemeter (Burleigh Instruments Inc., Model WA-20), to within 2.0 GHz of the absorption transition of the species to be measured. Once the center frequency has been verified, a sweep signal that is generated by the laboratory's computer system, and connected to the ring dye laser controllers, is enabled. This sweep signal is a 4 Hz triangle wave that causes the ring dye lasers to repetitively ramp up in wavelength for 128 ms and then ramp down in the same amount of time. To verify the linearity and width of the laser sweep, some of the light leaving the ring dye laser is split off to a Fabry-Perot interferometer. The output of this diagnostic is digitized and used by the computer to interpret the absorption waveform. One cycle of the ramp signal, the digitized output of the Fabry-Perot, and a typical absorption trace is shown in figure 2.

Figure 2. Shown is the laser sweep, an idealized Fabry-Perot interferometer output, and a typical absorption trace which is generated by taking the ratio of the signal and reference.

Before being delivered to the mixing table, via fiber optic cable, the output of each ring dye laser is amplitude modulated by acousto-optic (AO) modulators. This modulation facilitates lock-in amplifier detection techniques and is used for frequency multiplexing the light from each laser system. The first order beam from the AO is sent to the fiber launch and ultimately to the mixing table where it is optically combined with the outputs of the other laser systems. The zero order beam is sent to an optogalvanic (OG) cell. By monitoring the current draw of this device, using a lock-in amplifier, the center wavelength of the ring dye laser can be verified during the experiment when the laser is in a swept mode. This waveform is also digitized and stored by the laboratory's computer system.

At the mixing table, light from all the laser systems is combined and launched into a fiber for transport to the separator vessel. Sending units split this light into the required number of shot locations and generate a reference signal using a photo-diode based detector. Identical detectors measure the light after it passes through the vessel and any vapor within.

Figure 3. Simplified instrumentation system block diagram.

The U-AVLIS LAS Instrumentation System

The optical detector used to measure the light transmitted through the vapor is a hybrid device that incorporates a silicon photo-diode and an integral JFET operational amplifier. This detector/amplifier is then housed with an additional stage of signal amplification in an electromagnetic noise resistant package. The output of each detector package is then sent to a series of micro-processor controlled lock-in amplifiers (Stanford Research Systems model SR510). The U-AVLIS LAS laboratory employs a bank of over forty lock-in amplifiers for signal detection. Taking advantage of the fact that each laser is amplitude modulated at a different frequency, these devices are used to de-multiplex the signals generated by each of the probe laser systems. This permits the simultaneous detection of many different atomic or molecular species at a single location within the vessel. In addition, signal-to-noise ratio's are dramatically improved with the lock-in technique. The configuration of the detectors, lock-in amplifiers, and the entirety of the instrumentation system is shown in figure 3.

The output of each lock-in amplifier is digitized by a track-and-hold digitizer system that samples synchronously with the laser sweep. To compensate for any fluctuations in laser power during the sweep or those caused by the beam transport system, the computer calculates the ratio of each shot to its appropriate reference channel. The resulting waveform, V_S/V_R , is then used as an indication of the Doppler-broadened absorption. Refer to figure 2 for a depiction of a typical

absorption trace. By correlating this ratio with the data from the Fabry-Perot interferometer and applying Lambert's law, a line integrated density can be calculated.

The computer system gathers data from up to 96 channels during the laser sweep. 1024 points per channel are stored during this 256 ms. Ratios of signals to references are calculated, stored, and then plotted on workstation screens. Baselines for these ratios are determined, the logarithms and integrals are calculated, and physical parameters of the vapor plume such as densities, vapor rates, internal temperature, and alloy mole-fraction are quantified and plotted for the operators. Throughout all stages of the calculations, data integrity is checked. Warnings and error messages are output if the computer determines that an absorption trace is corrupted in any way or if a calculation can not be performed. A real-time plotter/trender permits graphical representation of any raw or calculated data. Network communications allows operators to access data from any one of five workstations (Hewlett Packard Apollo Series 400 and 700 systems). With disk storage capacity of over 2.4 G Bytes, all data from a long duration experiment can be archived for post-run analysis. One of the most powerful features of this data acquisition system is the flexibility provided by its table-driven configuration files. Changing the requirements of the system or experiment requires minimal effort and time and does not require alteration of the actual code.

Summary

A LAS laboratory has been described. This mature but continually updated facility is critical to the eventual commercialization of U-AVLIS technology. Vapor plume characterization, vapor physics, photo-ionization physics, vapor rate monitoring, deposition rate monitoring, and vaporizer development have all been supported by this system. In addition, this system could have application in the semiconductor, the refractory and composite metals, and the environmental monitoring industries. It could also be applied as a research tool for general spectroscopy, atmospheric studies, combustion engine development, and chemical process control. With its proven reliability in long duration experiments, its flexibility, the wide range of wavelengths it can access, and its powerful signal processing and computer systems, the capabilities of this laboratory are broad and ever increasing.

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