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ATOMIC VAPOR LASER ISOTOPE SEPARATION USING RESONANCE IONIZATION

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ABSTRACT

Atomic vapor laser isotope separation (AVLIS) is a general and powerful technique. A major present application to the enrichment of uranium for light-water power-reactor fuel has been under development for over 10 years. In June 1985, the Department of Energy announced the selection of AVLIS as the technology to meet the nation's future need for enriched uranium. Resonance photoionization is the heart of the AVLIS process. We discuss those fundamental atomic parameters that are necessary for describing isotope-selective resonant multistep photionization along with the measurement techniques that we use. We illustrate the methodology adopted with examples of other elements that are under study in our program.

1. Introduction

Atomic vapor laser isotope separation (AVLIS) is a general process for enriching an atomic-vapor stream in one or more of its isotopes. The heart of the process is the selective multistep photoionization of the isotope of interest. In order to fully describe this selective photoionization, one must measure or otherwise determine the values for several atomic parameters of the particular element and isotope. A list of these key atomic parameters includes: radiative lifetimes, energy levels and ionization potentials, quantum numbers, hyperfine structure and isotope shifts, transition strengths and ionization rates, and electric- and magnetic-field effects. A large portion of the values comprising this list can be found in literature sources and more general spectroscopic compilations such as the NBS monographs. In this paper we describe some of the techniques that we use to measure those values, including some not found in the literature, and illustrate these techniques with results from elements we are currently investigating in our program.

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2. Measurements

The measurement of the essential atomic parameters and the development of the necessary tools and techniques for these fundamental measurements have played a key role in the success of AVLIS. Ionization provides a very sensitive means of signal detection and consequently is used in many of the measurements. These ionization techniques typically use tunable pulsed dye lasers for resonant stepwise excitation to the level of interest followed by ionization by one of several possible techniques. Ions can be detected with biased parallel plates, microchannel plate detectors, or Channeltron detectors. Oftentimes these detectors are part of a mass spectrometric system employed for distinguishing individual isotopes. Using the basic technique of resonant excitation followed by ionization and adapting this technique to the particular measurement of interest, many of the atomic parameters necessary for designing an AVLIS process can be determined. Then by complementing this technique with standard methods such as absorption and fluorescence spectroscopy, the entire set of atomic parameters can be determined for the element of interest.

Perhaps the simplest measurement to make and one of the most important for the initial evaluation of a particular photoionization process is the radiative lifetimes of the various levels in the photoionization ladder One technique that we use for determining radiative lifetimes is time-delayed photoionization (Carlson 1977). The atom is excited to the level of interest with one or more laser pulses, then photoionized with a final sequence of laser pulses that is varied in time. Ion signal is plotted as a function of delay time. In a more traditional fashion we also measure fluorescence decay using photomultipliers and fast transient digitizers.

An important technique that employs ionization for signal detection and which has yielded a wealth of atomic physics information over the years is scanning laser ionization spectroscopy. In this technique the atom of interest is stepwise excited to a high-lying level with 1, 2, or 3 lasers. This excited state can then be ionized by one of several methods including collisions with neutrals or electrons, field ionization, or photoionization. All of these techniques employ a scanning dye laser for probing levels just below or slightly above the atom's ionization threshold. Figure 1 shows some data taken in dysprosium (Worden 1978) where the third-step laser was scanned from slightly below the ionization potential, up to and beyond the continuum. This scan gives the location of the onset of photoionization as well as information on different Rydberg series including level locations and principal quantum numbers, *which are found by fitting the data to standard formulas.*

In order to selectively photoionize one isotope of an element, a photoionization ladder must be chosen such that the isotope of interest is clearly separated by virtue of its isotope or hyperfine shifts with respect to the other isotopes. Consequently we must measure these shifts for any photoionization sequence of promise in a particular element. For first-step hyperfine structure, traditional spectroscopic techniques such as fluorescence or absorption are generally used. For the second- and third-step transitions in a three-step photoionization scheme, resonance ionization detection is used where the transition of interest is excited with a narrow-band (~1 MHz) cw dye laser and scanned over the bandwidth of the pulsed dye lasers, which are wide enough in frequency to excite a large portion of the hyperfine structure in a single shot. Figure 2 shows the hyperfine structure of each transition in a three-step photoionization scheme in Gd. The second- and third-step transition hyperfine spectra were obtained in the manner outlined above.

In situations where small hyperfine splittings and isotope shifts are encountered such as for the odd isotopes of gadolinium, polarization selection rules can be exploited to achieve photoselectivity. In fact, polarization selection rules in resonant photoionization provide a convenient tool for unambiguously determining the angular momentum or J quantum numbers of previously unassigned levels.

The behavior of atomic levels in the presence of external electric and magnetic fields is another area studied extensively in our program. In Fig. 3 we illustrate some dramatic Stark effects that can be observed for high-lying valence levels even at extremely modest electric fields. The high-resolution spectra were taken with a cw dye laser scanned across the $J = 0$ autoionizing resonance at 49799 cm^{-1} in ^{160}Gd using a field ionization and collection arrangement. As the electric field is increased, the line breaks up into a multitude of separate lines. The autoionizing level is thought to be coupled to a high-lying ($n^* \sim 40$) Rydberg level via the electric field. The multiple lines correspond to transitions to parabolic angular momentum states that are degenerate in a field-free environment.

Perhaps the most important spectroscopic parameters in the AVLIS process are the transition oscillator strengths or dipole moments. The resulting optical cross sections determine the laser fluences needed for effective atomic photoionization. A method used extensively in our laboratories for accurately measuring transition oscillator strengths relies on observing Rabi flopping of atomic populations. An example of data obtained for atomic gadolinium using this technique is shown in Fig. 4. The ^{160}Gd was photoionized in a stepwise fashion by three resonant laser pulses. The first- and second-step excitations were derived from pulse-amplified cw dye-laser systems allowing selective excitation and detection of ^{160}Gd without a quadrupole mass filter. The second and subsequent photoionizing steps were delayed in time from the first-step excitation pulse. In Fig. 4 the photoion signal is plotted as a function of fluence of the first-step laser. The dramatic oscillations observed in the photoion signal arise from the coherent evolution of the atom between the manifold of magnetic sublevels in the ground and first excited state. Since each magnetic sublevel has its own dipole moment, the signal is not purely sinusoidal as a function of field amplitude of the first-step laser, but is instead periodic. Since the first-step laser fluence is accurately measured, the only parameter varied to fit the data is the transition oscillator strength. This method has been used in our laboratory to measure transition oscillator strengths for bound-bound transitions originating on both low- and high-J excited states.

In addition to the above technique, which requires a very narrow-bandwidth pulse-amplified cw dye laser, oscillator strengths can be determined from the branching ratio and lifetime of the level. We have already discussed means for determining the radiative lifetime of an atomic level: we employ two methods for obtaining transition branching ratios. In the first method the branching ratio is measured using the time-delayed photoionization technique described earlier. The upper state of the transition of interest is populated with a pump laser. This level is allowed to decay radiatively for a time τ, whereupon a probe laser photoionizes the lower level. By comparing the ion signal for a τ equivalent to several lifetimes with the ion signal when the lasers are coincident, we obtain a value for the branching ratio of the transition. Another means for obtaining transition branching ratios is to perform large survey emission scans of discharge lamps containing the element of interest. These types of scans have been done at the Kitt Peak National Observatory using their high-resolution Fourier transform spectrometer.

A very accurate technique for determining relative oscillator strengths uses absorption spectroscopy in a hollow-cathode discharge. The cathode is constructed from the material of interest, usually in the form of a bar with a deep slot. The discharge is operated with a few Torr of neon or a heavier rare gas that ionizes and sputters the cathode material. By running at a high-discharge-current density, high densities of ground- and low-lying excited states can be produced. With this device we can measure the relative absorption between two transitions with a common lower level and determine the relative oscillator strengths of the levels.

The key to any promising AVLIS process is the ionization step. In some cases the final step is to an autoionizing level that has been chosen from the photoionization scans described earlier. To determine the oscillator strength of a transition to an autoionizing level, we measure the Lorentzian line width of the transition and the peak photoionization cross section. To measure line width, we simply scan the photoionizing dye laser over the transition of interest and plot ion signal vs frequency. To obtain the photoionization cross section, the fluence of the photoionizing laser is varied to yield a plot of ion signal vs laser fluence. Figure 5 shows the results for an autoionizing transition in hafnium. The points representing ion signal vs laser fluence are fit to a saturation curve that is derived from a simple rate equation model.

Knowledge of the fluence of the photoionizing laser in the region of the intersection of the atomic beam with the excitation lasers is vital for determining an accurate value for cross section. The plot on the right is the beam profile of the photoionizing laser fit to a curve representing the intensity profile of a circular Fraunhofer pattern. The curve for the saturation fit used in the left-hand plot shown in this same figure is generated by integrating the intensity-dependent photoionization rate equation over both the spatial and temporal bounds of the photoionizing laser pulse. Finally, the value for oscillator strength is determined from the product of oscillator strength and line width.

3. Conclusions

In the case of oscillator-strength measurements, as well as the other atomic parameters necessary for designing an AVLIS process, there are advantages to having more than one technique available for the measurement. By making a particular measurement using different methods, we have a cross check of our result. This philosophy ensures the highest possible accuracy in our final values. Beyond this is the flexibility afforded by having a variety of available measurement techniques. This versatility becomes more important as we attempt to expand the AVLIS process to other elements.

References

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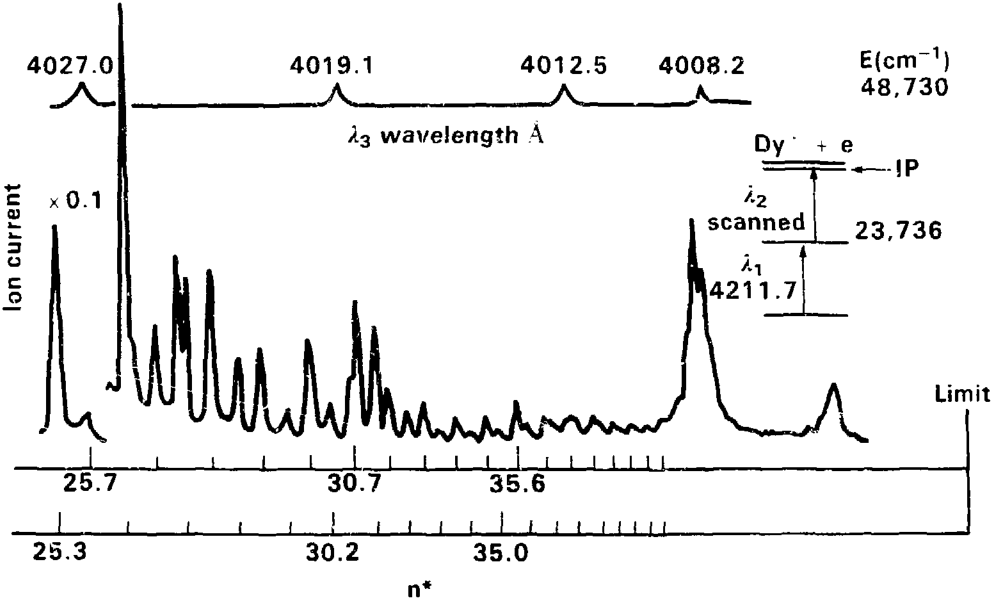

Fig. 1. Photoionization threshold of dysprosium and autoionizing Rydberg series converging to the first excited state of the ion.

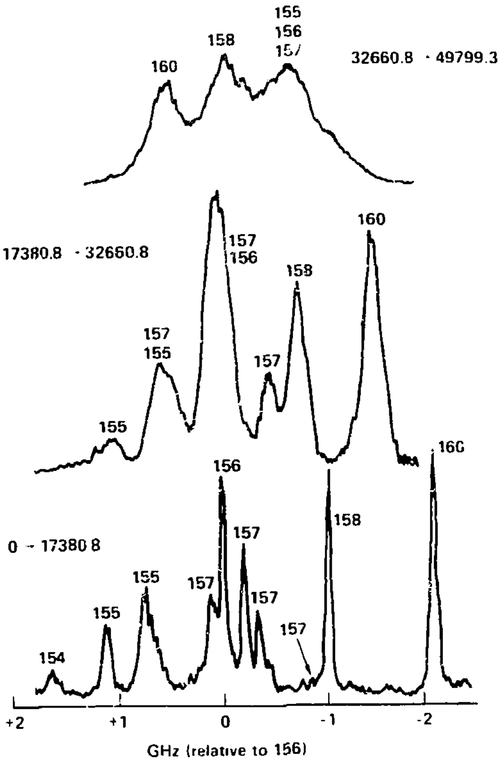

Fig. 2. Hyperfine structure for transitions of 3-step photoionization ladder in Gd.

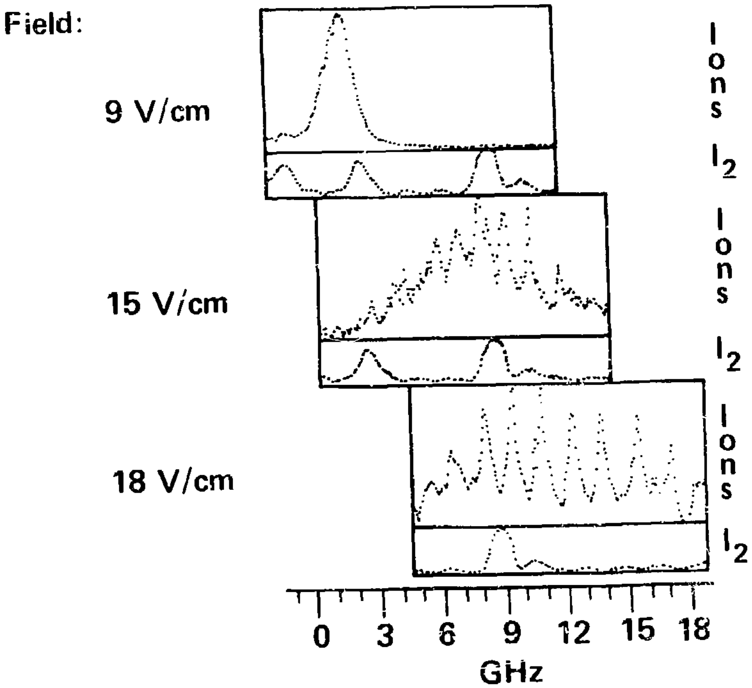

Fig. 3. DC field effect on the 32660-49799 cm^{-1} transition in ^{160}Gd.

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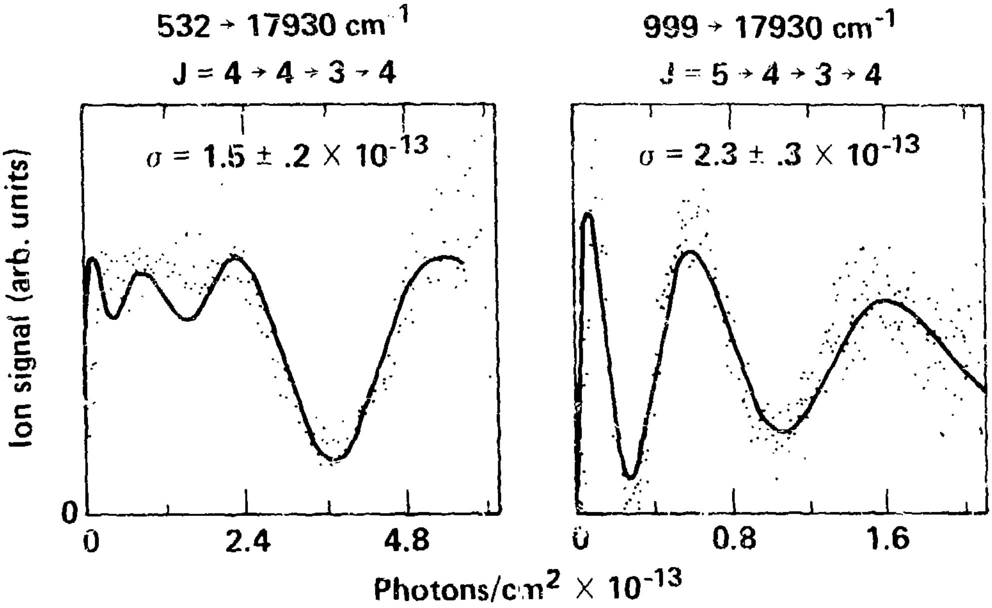

Fig. 4. Cross-section measurement using Rabi oscillations in ^{160}Gd.

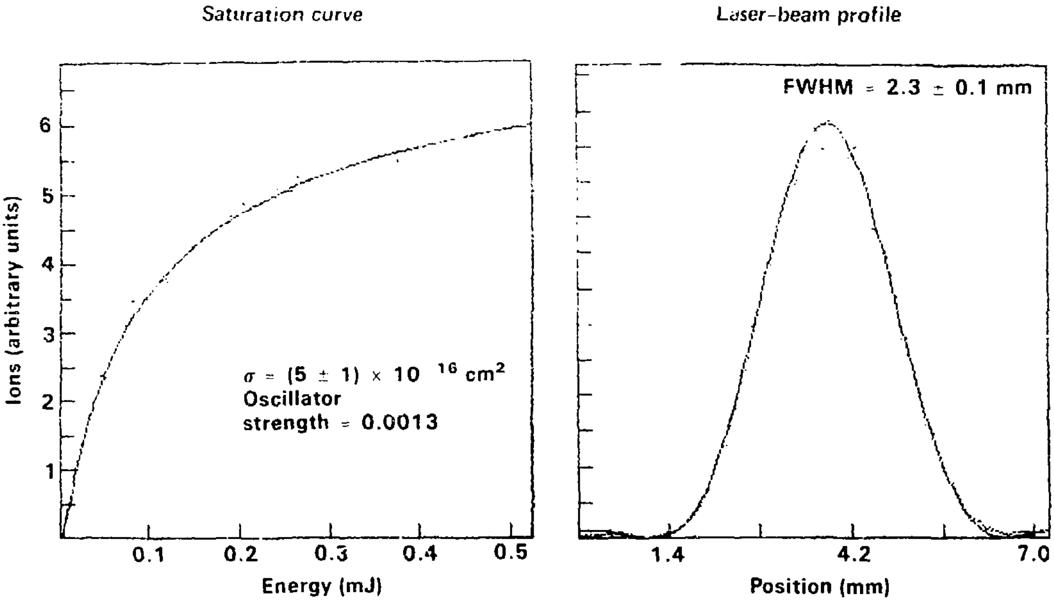

Fig. 5. Autoionization cross-section measurement of the 34991.64 → 55190 cm^{-1} transition of Hf.

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