

LA-UR -86-137

CONF-851185 --15

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LA-UR--86-137

DE96 006014

TITLE NUCLEAR ISOMER SEPARATION

AUTHOR(S) P. Dyer

SUBMITTED TO International Laser Science Conference in Dallas, TX  
November 18-22, 1985.

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**Los Alamos** Los Alamos National Laboratory  
Los Alamos, New Mexico 87545

## NUCLEAR ISOMER SEPARATION\*

P. Dyer

Physics Division, Los Alamos National Laboratory  
Los Alamos, New Mexico 87545

### ABSTRACT

We report experiments on selective photoionization of atoms containing isomeric nuclei of  $^{197}\text{Hg}$ . Other isomer separation techniques and their limitations are discussed.

### INTRODUCTION

To produce a nuclear population inversion for a gamma-ray laser, isomer separation is generally required, as nuclear reactions usually produce greater quantities of ground-state nuclei than excited-state nuclei. The laser isomer separation techniques discussed here are the same as those of isotope separation, but the technical problems are much more difficult, as the sample size is many orders of magnitude smaller and the nuclear states are short-lived. Here we discuss a resonance ionization demonstration experiment with  $^{197}\text{Hg}$  and initial measurements of the optical piston with sodium. We conclude with a comparison of techniques.

### RESONANCE IONIZATION OF $^{197\text{m}}\text{Hg}$

We have demonstrated isomerically-selective photoionization of  $^{197\text{m}}\text{Hg}$  (nuclear half-life 24 hours) via the atomic excitation sequence  $6^1\text{S}_0 - 6^3\text{P}_1 - 8^1\text{S}_0 - \text{Hg}^+$ . Three collinear pulsed dye laser beams were used: 254, 286, and 696 nm, selectively exciting the first two transitions and ionizing through an autoionization state in the continuum.

Gold target foils were bombarded by deuterons at the Los Alamos tandem Van de Graaff accelerator, to generate  $^{197}\text{Hg}$  by the (d,2n) reaction. These target foils were then heated in vacuum to distill mercury onto a second gold "catcher" foil, which was then sealed in a shielded capsule for transportation to MIT.

Meanwhile, at the MIT Laser Center, optical excitation experiments were performed with two vapor cells, one containing natural mercury and the other, mercury enriched in  $^{202}\text{Hg}$ , for adjustment and calibration of the apparatus. Upon arrival of the radioactive sample at the Laser Center (sixteen hours after the end of bombardment), the active catcher foils were introduced into a clean irradiation cell and heated to expel mercury. The Pyrex irradiation cells were 12-cm long and 15-mm in diameter, with fused-silica Brewster windows at each end. No materials that had been exposed to natural mercury were used in constructing the  $^{197}\text{Hg}$  cell. Other materials to which mercury was exposed in the chamber were limited to Teflon, Viton O-rings, ceramic adhesive and clean iron; all had been previously found, using  $^{197}\text{Hg}$  as a tracer, to have low tendency to adsorb mercury.

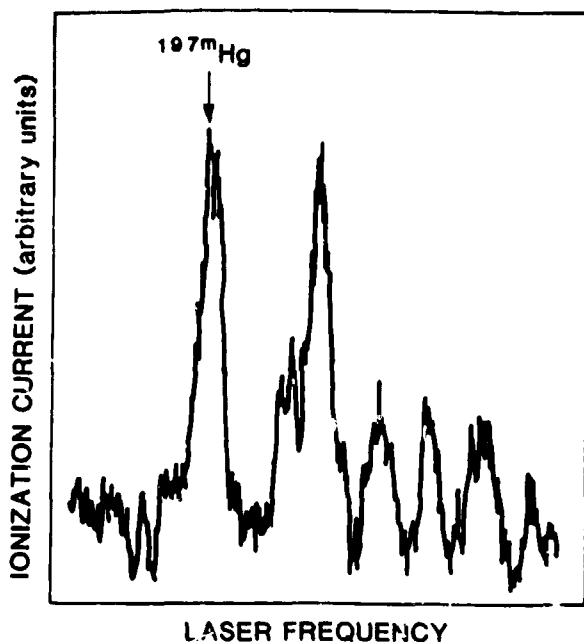


Fig 1. Ionization current, as a function of 254-nm scanning frequency, with 286-nm radiation fixed at the  $^{197\text{m}}\text{Hg}$ -c hyperfine component.

excitation by both 254- and 286-nm radiations was sufficient to achieve clean separation of  $^{197\text{m}}\text{Hg}$ .

However, we did not obtain an enriched sample outside the cell. An attempt to measure enrichment by counting gamma rays from the positive and negative electrodes failed. There was no significant difference in isomeric enrichment between the two electrodes. Moreover, the total number of radioactive atoms on the wire exceeded nearly 100-fold the number of ions collected, estimated from the ionization current and collection time. Presumably, the selectively ionized and collected portion was greatly diluted by nonselective adsorption of  $^{197}\text{Hg}$ , despite precautions to use clean Fe electrodes.

The isomer  $^{142\text{m}}\text{Eu}$  has been resonantly ionized by Alkhazov et al.<sup>2</sup> In this experiment, three laser beams intersected the atomic beam from a mass separator on-line to a proton synchrotron.

#### THE OPTICAL PISTON

An alternative possible technique for laser isomer separation is light-induced drift.<sup>3,4</sup> Consider a laser beam incident along the axis of a capillary cell containing the atoms of interest, e.g. sodium, in a buffer gas. When the laser frequency is tuned just below the center of a Doppler-broadened absorption line, only those sodium atoms moving toward the laser beam will be excited. As the velocity-changing collision cross section for excited sodium atoms

The collecting electrodes were a pair of magnetically supported Fe wires on opposite sides of the laser beam. A collecting potential of 400 volts was applied to the electrodes. Currents from a phototube and from the ion-collector were amplified, passed to a boxcar integrator, and registered on a chart recorder. The quantities of  $^{197\text{m}}\text{Hg}$  released into the cell were of the order of  $2 \times 10^{12}$  atoms. Ion-collection rates were of the order of  $4 \times 10^6 \text{ s}^{-1}$ .

Figure 1 shows the chart record of the ionization current when the 286-nm laser was fixed at the  $^{197\text{m}}\text{Hg}$ -c peak, the 254-nm radiation was scanned, and the ions that were created in the final transition to the continuum were collected. A large peak in the ionization current is observed at the expected position. The combined

colliding with the buffer gas is greater than that for ground-state atoms, there will be a drift of sodium atoms along the direction of the laser beam, toward the downstream end of the capillary. If the sodium is optically dense, the "optical piston" is manifested as a front moving along the cell, as observed by Werij et al.<sup>5</sup>

We have begun measurements to repeat those of Werij et al. with sodium. Thus far a front movement of about 2 cm has been observed. We hope to extend these measurements to an element for which the piston effect is not limited by interaction of the active atoms with the cell walls. Enrichments for isotope separation, and subsequently isomer separation, will then be measured.

### COMPARISON OF TECHNIQUES

A number of laser techniques are available for isomer separation: resonance ionization (where the ionization step may be performed by a laser or by electric fields or collisions acting on Rydberg states), the optical piston, photochemistry, radiation pressure, magnetic or electric deflection of an optically pumped atomic beam. The optimum choice will depend on atomic state energies, hyperfine structure, vapor pressure, chemistry (especially surface), nuclear state lifetime, isomer production rate, and initial enrichment factors. Various factors limit the efficiency of the separation, the enrichment achieved, and the time required to perform the separation. In general, separations in cells have high efficiency, but low resolution, whereas the opposite is true for separations in atomic beams.

Formation of an atomic beam is necessarily an inefficient process. In a cell there is the potential for a given atom to pass through the laser beam many times, but to achieve this, there must be little loss of the material to adsorption on the walls. A further factor limiting the efficiency for resonance ionization, particularly in cells, is space charge.

If the separation is performed in a cell, and the Doppler width is greater than the hyperfine splitting of the lines involved in discrete transitions, loss of enrichment results. Collisions in a cell also limit enrichment. In the case of resonance ionization, resonance charge exchange results in non-specific collection of ions. In the cases of the optical piston or of photochemistry, inelastic collisions at high sample densities dilute the enrichment. Multiphoton ionization is also a source of non-selective background in the case of resonance ionization by photons, whether in a cell or an atomic beam.

There are various considerations involved in the choice of pulsed versus CW lasers. CW lasers offer narrower bandwidth and high duty factors. Pulsed lasers are more suitable for producing UV wavelengths by frequency doubling. They are also better suited to multistep processes, such as resonance ionization with the ionization step performed by photons.

The most widely applicable technique thus far appears to be that of resonance ionization. In the case of isomer separation for a gamma-ray laser, this technique offers the advantage that implantation into a crystal may be performed by the field that

collects the ions. If far UV wavelengths are not required, if ionization is performed by electric fields rather than photons, and if the time constraints are not too severe, CW lasers may be used. Otherwise pulsed laser excitation is necessary. The time to separate the required number of isomers for a gamma-ray laser, once the sample is in the laser beam, can be much shorter than a second. Thus the entire process of producing implanted isomers will be more limited by the time to produce the isomers in nuclear reactions and to transfer them from the reaction target to the laser beam, than it is by the laser ionization time.

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\*Work supported in part by the Division of Advanced Energy Projects of the United States Department of Energy. Part of this work was performed while the author was a Visiting Scientist at the MIT Laser Research Center, which is a National Science Foundation Regional Instrumentation Facility.

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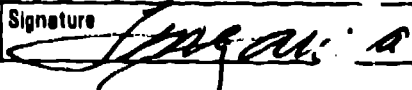
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