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Nuclear Isomer Separation**

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Abstract. Pure specimens of nuclear isomers are required for gamma-ray lasers. We have selectively photoionized atoms containing isomeric nuclei of ^{197}Hg. The isomers were produced by the ^{197}Au(d,2n)^{197}Hg reaction and distilled. Three pulsed dye lasers were used to selectively ionize mercury atoms by doubly resonant three-step photoionization. Other isomer separation techniques and their limitations are discussed.

1. Introduction

Our scheme for a gamma-ray laser involves two-step pumping (Baldwin 1981, 1986), in which a long-lived nuclear isomer would be produced by a nuclear reaction, separated, and implanted into a crystalline host (for supporting the Mossbauer effect and Borrmann modes). The crystal fiber would be irradiated by a beam of photons to transfer energy (probably through the electrons) to "tickle" the nucleus into a nearby short-lived nuclear state that would then decay to the upper lasing level. First nuclear superradiance would be measured by observing the time dependence and angular distribution of emitted gamma rays from 10^{10} to 10^{14} excited nuclei implanted in a low-Z substrate about a micrometer in diameter and several millimeters long. The first step in making such a gamma-ray laser is to provide a nuclear population inversion. However, when nuclear excited states are produced by a nuclear reaction, many more ground-state nuclei are usually formed. A separation step is generally required. The work reported here demonstrates the feasibility of isomer separation; the particular nucleus for making a gamma-ray laser has not yet been chosen.

We also include here a summary of various possible isomer separation techniques, outlining advantages and disadvantages of each.

2. Resonance Ionization of ^{197m}Hg

We have demonstrated isomerically-selective photoionization of ^{197m}Hg (nuclear half-life 24 hours) via the atomic excitation sequence 6^1S_0 - 6^3P_1 - 8^1S_0 - Hg$^+$ (Dyer 1985). 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.

First, gold target foils were bombarded by deuterons at the Los Alamos tandem Van de Graaff accelerator, to generate ^{197}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 the Massachusetts Institute of Technology Laser Research Center.

At the Laser Center, optical excitation experiments were performed with two vapor cells, one containing natural mercury and the other, mercury enriched in ^{202}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, shown in Fig. 1, 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}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}Hg as a tracer, to have low tendency to adsorb mercury.

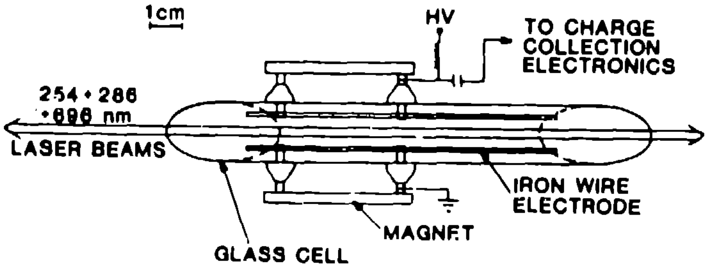

Fig. 1 Schematic diagram of the mercury vapor cell.

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 bandwidths of the 254- and 286-nm beams were about 3 and 2 GHz, respectively. For the 254-, 286- and 696-nm beams, the intensities were about 1, 10, and 400 μJ/pulse, respectively. Beam diameters were about 2 mm. The 20-nsec pulses were repeated 20 times per second. Frequency doubling was provided by KDP (286nm) and lithium formate (254 nm) crystals. The quantities of ^{197m}Hg released into the cell were of the order of 2×10^{12} atoms. Ion-collection rates were of the order of 4×10^{6} s^{-1}.

For orientation, Fig. 2 shows a rough computer simulation of the ion current as a function of the frequencies of the 254- and 286-ion beams. Figure 3 shows the measured ionization current when the 254-nm laser was fixed at the ^{197m}Hg-c peak (selecting the F = 11/2 6^3P_1 hyperfine state), the 286-nm radiation was scanned, and the ions that were created in the final transition to the continuum were collected. Figure 4 is the same, but with the 286-nm laser fixed and the 254-nm laser scanned. Peaks in the ionization current are observed at the expected positions. The combined selection by both 254- and 286-nm radiations was sufficient to achieve a clean separation of ^{197m}Hg.

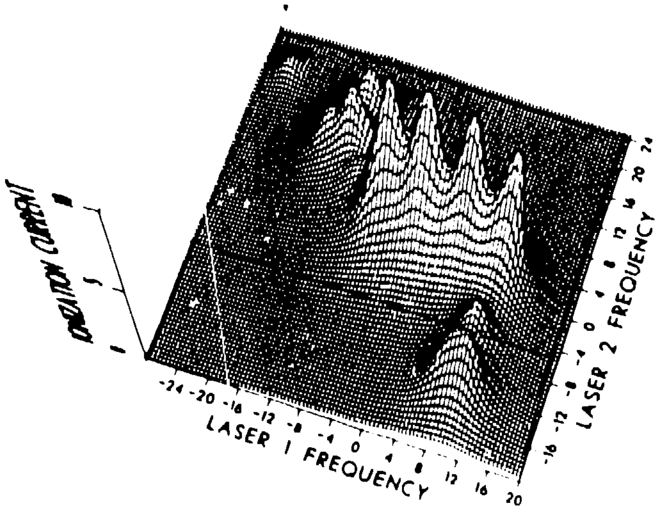

Fig. 2 Computer generated simulation of the ionization current as a function of the 254-nm laser frequency (laser 1) and the 286-nm laser frequency (laser 2). The ^{197m}Hg peak of interest is labeled. Most of the other peaks are from natural mercury contamination. Frequency scales are in GHz; ionization current is in arbitrary units.

We did not, however, 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}Hg, despite precautions to use clean Fe electrodes.

The isomer ^{142m}Eu has been resonantly ionized by Alkhazov et al. (1985). In this experiment, three laser beams intersected the atomic beam from a mass separator on-line to a proton synchrocyclotron. Ionization rates of 10^4 s^{-1} were achieved.

3. 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 (light-induced drift in a buffer gas) (Gel'mukhanov 1979, Werij 1984), photochemistry, radiation pressure, magnetic or electric deflection of an optically pumped atomic beam (Zhu 1985). 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 case 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.

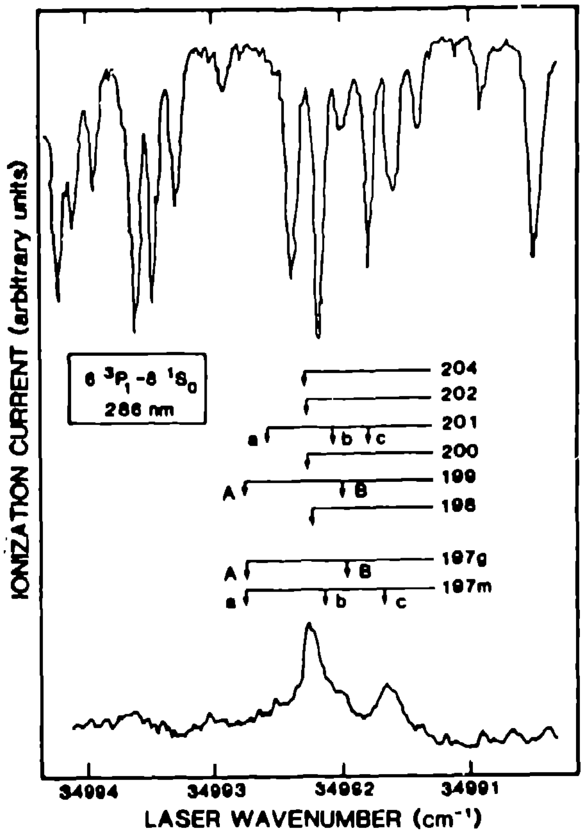

Fig. 3 Ionization current, as a function of 286-nm scanning frequency, with the 254-nm radiation fixed at the ^{197m}Hg-c hyperfine component. The larger of the two peaks is contributed primarily by stable isotopes of mercury. The smaller peak is due to ^{197m}Hg. The upper trace is an iodine comparison spectrum (at the fundamental frequency).

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

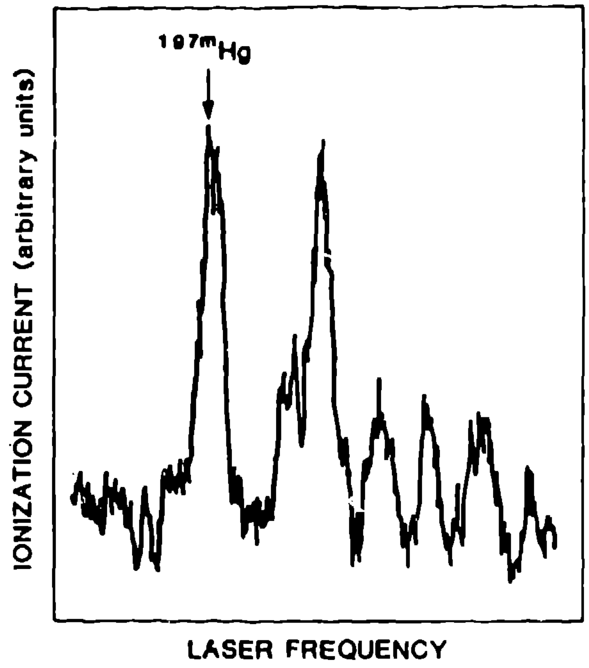

Fig. 4. Ionization current, as a function of 254-nm scanning frequency, with the 286-nm radiation fixed at the ^{197m}Hg-c hyperfine component. In this case, the largest peak is due to ^{197m}Hg.

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.

It is difficult to scale up charged-particle-induced reaction rates to produce 10^{10} or more isomers in a second. There is, however, the possibility of producing such yields with a pulsed nuclear reactor. Such devices can emit over 10^{16} neutrons into a column of about 2000 cm^3 in 30 msec. If such a reactor, the lasers for isomer separation, and the photon source for transferring the nuclei from isomeric states to nearby short-lived states, can be located in one place, it may be possible to work with isomeric states as short-lived as 1 sec for a first gamma-ray laser demonstration.

References

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Alkhazov G D, Barzakh A E, Berlovich E E, Denisov V P, Dernyatin A G, Ivanov V S, Letokhov V S, Mishin V I and Fedoseev V N, 1985 JETP Lett. 40 836
Baldwin G C, Solem J C and Goldanskii V I 1981 Rev. Mod. Phys. 53 687

Baldwin G C 1986 Proc. First International Laser Science Conference, Dallas, TX, Nov 1985, ed W Stwalley (New York: American Institute of Physics) in press

Dyer P, Baldwin G C, Sabbas A M, Kittrell C, Schweitzer E L, Abramson E, and Imre D G 1985 J. Appl. Phys. 58 2431

Gel'mukhanov F Kh and Shalagin A M 1979 JETP Lett. 29 711

Werij H G C, Woerdman J P, Beenakker J J M and Kuscer I 1984 Phys. Rev. Lett. 52 2237

Zhu X 1985 Chinese Physics 5 375