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TITLE: Three-Step Photoionization of Mercury for Application to Separation of Mercury Isomers

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THREE-STEP PHOTOIONIZATION OF MERCURY FOR APPLICATION TO SEPARATION OF MERCURY ISOMERS

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ABSTRACT

Development of techniques for separating isomeric nuclides is important to the investigation of schemes for gamma-ray lasers. In preparation for an experiment to separate 10^{14} atoms of the ^{197m}Hg (299 keV, $\tau_{1/2}$ = 24 hours) isomer, we report isotopically selective resonance ionization of mercury atoms. This has been accomplished by three-step excitation via the 6^3P_1 and 8^1S_0 excited states, using three collinear pulsed laser beams of 254, 286, and 532 nm wavelengths from a Nd:YAG and two dye lasers. These beams were passed through a closed mercury-vapor cell containing electrostatic plates to which the ions were drawn. Ion current and fluorescent radiation were measured as a function of laser frequency. Hyperfine structures for the 254- and 286-nm transitions were observed.

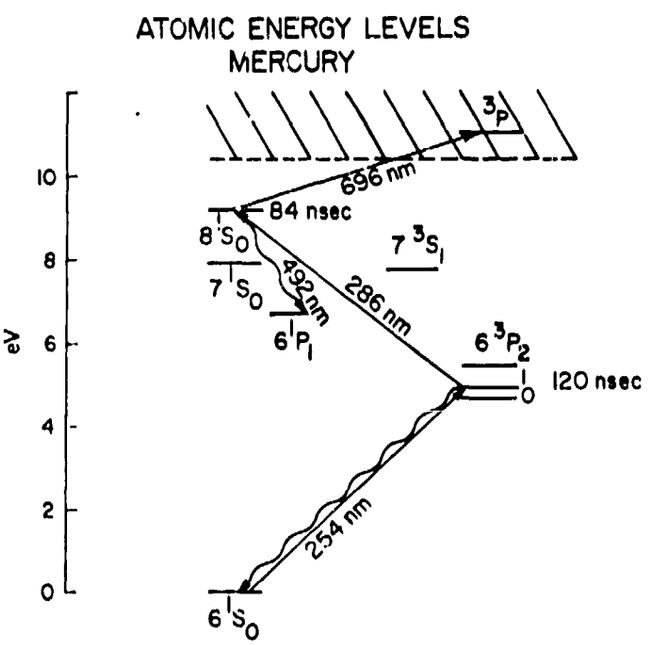

Fig. 1. Mercury atomic energy levels involved in the ionization scheme.

INTRODUCTION

Experience in enriching nuclei in isomeric states is important to the development of gamma-ray lasers.[1,2,3] As a test case for creating a nuclear population inversion, we have chosen the ^{197m}Hg isomer having a half-life of 24 hours (299 keV, I = 13/2+). The ^{197}Hg ground state (I = 1/2-) has a half-life of 64 hours. This isomer is not a gamma-ray laser candidate, but it is relatively long-lived and the hyperfine structure of the transition to the first atomic excited state has already been measured.[4]

The proposed scheme for isomer separation has three steps. In the first step, we produce 10^{14} atoms of the ^{197m}Hg isomer at an accelerator by the ^{197}Au(d,2n)^{197m}Hg reaction (cross section about 300 mb), using 3 μA of 16-MeV deuterons bombarding a gold target 2-MeV thick for 24 hours. Roughly equal quantities of ^{196}Hg, ^{197}Hg, and ^{198}Hg ground states will be produced at the same time. In the second step, the gold foil will be melted, and the mercury vapor will fill an interaction region traversed by three collinear laser beams. The mercury atoms will be resonantly ionized by three successive photon absorptions, (see Fig. 1): 1) excitation from the 6^1S_0 ground state to the 6^3P_1 first excited state (lifetime 120 ns[5]) by a 254-nm transition, 2) excitation from the 6^3P_1 state to the 8^1S_0 state (lifetime 84 ns[5]) by a 286-nm transition, and 3) ionization of the 8^1S_0 state by a photon having

TABLE I. CHARACTERISTICS OF THE RADIATIONS EMPLOYED

Wavelength (nm)	Produced by	Timing (ns)	Bandwidth (GHz)	Beam Diameter (mm)	Intensity (μJ/pulse)
532	Nd:YAG-2H	20 - 40	30	6	8000
254	Nd:YAG-3H + dye	0 - 20	2.6	1	3
286	Nd:YAG-2H + dye	20 - 40	1.8	4	20

wavelength less than 1022 nm. Ions will be collected on plates by an electrostatic field. As one of the 254-nm hyperfine lines of the ^{197m}Hg atoms is known to be 10 GHz away from other ^{196},^{199},^{200}Hg lines, the bandwidth of the 254-nm laser beam can be as large as a few GHz. In the third step, the isomeric enrichment will be monitored by measuring ratios of gamma-ray yields from the ground and isomeric-state decays.

EXPERIMENTAL METHOD

In experiments thus far, we have performed the second step--resonance ionization--for natural mercury isotopes (see also the work of Ref. 7). This work represents the first isotopically selective resonance ionization of mercury.[8]

Radiations from a Nd:YAG pump laser and two pulsed tunable dye lasers (see Table I) were merged collinearly into the reaction cell. The 254- and 286-nm beams from the dye lasers were formed with the aid of frequency-doubling lithium formate and KDP crystals.

The path length in the reaction cell was 20 cm. Two Al plates, 10 x 1.3 cm, spaced 1.3 cm apart, established a 150-Vcm^{-1} collecting field for the ions. Mercury vapor was kept at a density of 7 x 10^{12} atoms/cm^3 by means of a reservoir maintained at 0°C. The background pressure was 0.05 Torr.

RESULTS

Excitation of the 6^3P_1 level was monitored by observing fluorescence with a photomultiplier tube through a quartz side window. The fluorescence signal was amplified and integrated with a boxcar integrator connected to a chart recorder. Figure 2 shows the fluorescence spectrum of the 6^3P_1 - to - 6^1S_0 transition, labeled by the isotopes known to produce each of the five most intense hyperfine lines. In addition, we have observed the fluorescent decay of the 8^1S_0 level to the 6^1P_1 level (492 nm) when the 254- and 286-nm laser beams were passed through the cell. Observation of this fluorescence required use of a condensing-lens system with a low-fluorescence UV-blocking filter placed between the lenses.

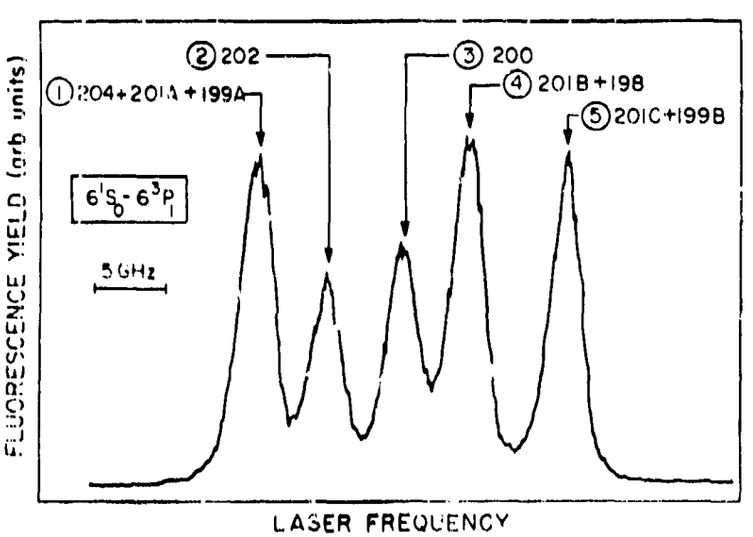

Fig. 2. Fluorescence spectrum of the transition 6^3P_1-6^1S_0 of natural mercury excited by 254-nm tunable radiation.

When all three (254-, 286-, and 532-nm) beams were passed through the cell, ionization current was measured; this signal was amplified and fed to the boxcar integrator. Figure 3 shows this signal, plotted as a function of the wave number of the 286-nm radiation; the numbers 1 to 5 identify the particular peaks in Fig. 2 at which the 254-nm radiation was fixed, while the 286-nm radiation was scanned. The abscissa was determined by simultaneously scanning fluorescence from an iodine cell irradiated by the 572-nm (undoubled 286-nm) radiation.

Appreciable ionization current was observed only when all three beams were present. With only the

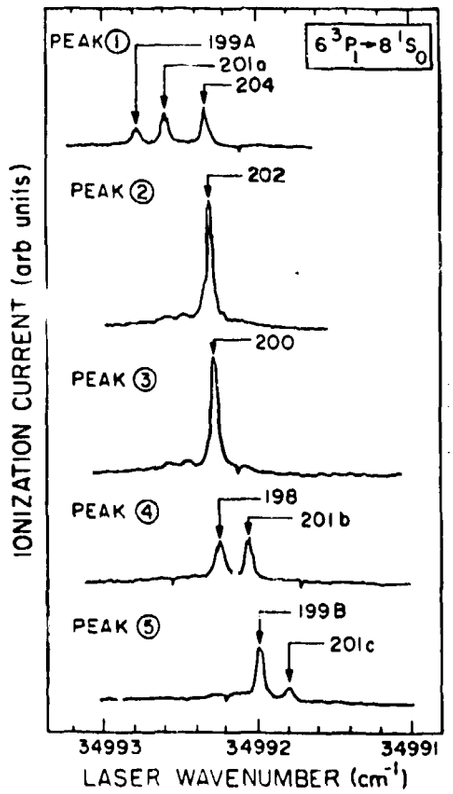

Fig. 3. Ionization current, plotted as a function of the wave number of the 286-nm 6^3P_1-8^1S_0 transition. The number (1, 2, etc.) identifies the peak of the 254 nm radiation (Fig. 2) on which the latter was fixed during that particular scan (Fig. 3), and the numbers affixed to each peak refer to the Hg isotope that contributes to it.

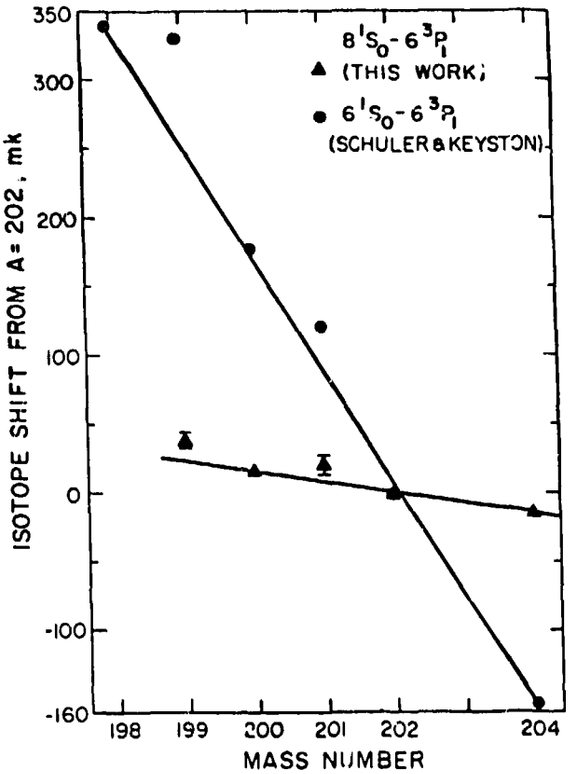

Fig. 4. Comparison of isotope shifts for the 8^1S_0 (this work) and 6^1S_0 levels (Schuler and Keyston, Ref. 9).

532-nm beam passing through the cell, the background (presumably multiphoton ionization) was only 10^{-3} times the on-resonance beam signal. Decreasing the intensity of the 532-nm beam reduced the ionization current by the same factor, as expected, since its intensity was much lower than that required to saturate the ionizing transition. If the 254-nm and 286-nm beams were both present, but not overlapped, there was no signal; thus, at this mercury concentration, radiation trapping was not a significant factor.

Each of the (one, two, or three) peaks observed in each 286-nm scan corresponds to one of the isotopes that contributed to the respective 254-nm peak at which the 254-nm radiation was held fixed. The isotope responsible for each peak in scans 1, 4, and 5 was identified by knowing the hyperfine-structure splitting of the odd-isotope 6^3P_1 levels[9,10] and by requiring the isotope shifts to vary smoothly with neutron number. Total line intensities, when compared with isotopic abundances, confirmed the assignments. The letter designations (199A, B and 201 a, b, c) correspond to the hyperfine states reached by 254-nm transitions in the conventional designation.[9]

Figure 4 compares isotope shifts for the 8^1S_0 level deduced from the observed hyperfine structure in Fig. 3 with the shifts for the 6^1S_0 level given in Ref. 9. As expected, the isotope shifts are much smaller for the state of higher principal quantum number. With the present (not optimized) configuration, about 20 minutes would be required to separate 10^{14} atoms. This time can be reduced considerably if we do not become limited by space-charge effects.

Resonance ionization has also been observed with a 700-nm beam from a third broadband dye laser in place of the 532-nm beam. At 696 nm, there is an 80-nm-wide $^3P_1$ autoionization resonance. We anticipate the gain in cross section to be much greater than the loss of laser beam intensity caused by adding the dye laser, but we have not yet measured the net gain.

CONCLUSION

We have thu demonstrated an iso ically selective procedure for ionizing mercury by pulsed-laser radiation. Although us of a 254-nm beam alone would not have sufficient selectivity for all of the naturally curring isotopes, becaus of overlap of some split odd-isotope lines with other lines, e use of two high-resolution beams does provide complete selectivity. This one experim nt does not suffice to demonstrate that a clean separation can be accomplished, because of the possibility of resonance-charge-exchange losses, although rough estimates suggest hat they should be negligible in the case of Hg at this density. We plan in a future exp riment to determine the isotopic composition of the collected ions by a time-of-flight measurement of the masses of the ions, using an accelerating electrode stack and a Channelton.

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REFERENCES

1. V. S. Letokhov, Zh. Eskp. Teor. Fiz. 64, 1555 (1973); Sov. Phys. - JETP 37, 787 (1973).

2. G. C. Baldwin, J. C. Solem, and V. I. Gol'danskii, Rev. Mod. Phys. 53 687 (1981).

3. P. Dyer and G. C. Baldwin, Proc. Conf. on Applications of Lasers in Nuclear Physics, Oak Ridge, TN, April 1982, C. Bemis, ed., to be published.

4. A. C. Melissinos and S. P. Davis, Phys. Rev. 115, 130 (1959).

5. G. C. King and A. Adams, J. Phys. B7, 1712 (1974).

6. G. C. King, A. Adams and D. Cvejanovic, J. Phys. B8, 365 (1975).

7. A. W. Miziolek, Anal. Chem. 53, 118 (1981).

8. P. Dyer, G. C. Baldwin, C. Kittrell, Dan G. Imre, and E. Abramson, Appl. Phys. Lett., in press.

9. H. Schuler and J. E. Keyston, Z. Physik 72, 423 (1931).

10. H. E. Gunning and O. P. Strausz, Advances in Photochemistry, W. A. Noyes, G. S. Hammond and J. N. Pitts, eds., I, 209 (Wiley Interscience, New York, 1963).