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SEPARATION OF URANIUM ISOTOPES BY SELECTIVE PHOTOIONIZATION

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SEPARATION OF URANIUM ISOTOPES BY SELECTIVE PHOTOIONIZATION*

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Considerable progress has been made during the past year in the development of selective photoionization processes for the separation of uranium isotopes. Uranium enrichment by selective photoionization has been scaled from the microscopic level reported by Tuccio et al[1] in June 1974 to the milligram per hour rate[2]. This progress has been supported by developments in the understanding of the uranium spectrum resulting from the application of tunable dye lasers as spectroscopic tools. In this paper, recent results of experiments on the laser photoseparation of uranium isotopes are reported.

The high density of uranium energy levels in the 10,000 cm^{-1} to 50,000 cm^{-1} energy range provides the opportunity for selective photoionization of uranium by a number of different schemes. Some of these are represented in Figure 1. In Figure 1A is shown a two-step process in which the isotopically selective step, $h\nu_1$, is followed by the absorption of photons with energy $h\nu_2$ to produce ions of the desired isotope. In the diagram, $h\nu_2$ is shown terminating on an autoionization state or discrete state, above the ionization continuum of the atom to take advantage of the relatively large absorption cross section associated with these states.

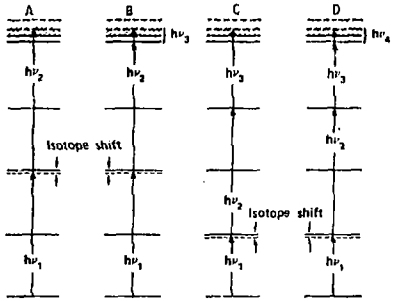

FIG. 1 Alternative Excitation Schemes for the Isotopically Selective Photoionization of Uranium

A potentially serious problem with the two-step photoionization process is that a small value of the photoionization cross section may preclude efficient utilization of the laser producing $h\nu_2$. This could seriously impair the economics of such a process. A variation of the two-step process is shown in Figure 1B. In this case, photons of energy $h\nu_2$ excite the atoms to an energy level slightly below the

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ionization continuum. Ionization is accomplished by the absorption of a third photon of energy $h\nu_3$. This scheme was suggested by Nebenzahl and Levin[3] as a means by which small photoionization cross sections could be utilized effectively in a separation system. By using an efficient infrared laser, such as a CO_2 laser, to provide the photons for the ionization step, process economics may not be affected adversely by a small photoionization cross section.

Another type of three-step process is diagrammed in Figure 1C. If the photons, or energy levels, can be properly chosen, it would be possible to ionize with photons approximately equal energy, that is:

$$h\nu_1 \cong h\nu_2 \cong h\nu_3 .$$

This places the required laser wavelengths in a spectral region which is readily accessible to presently available tunable lasers. A variation on this scheme, Figure 1D, utilizes the infrared photon $h\nu_4$ to overcome a small photoionization cross section.

In all of the diagrams, photons have been used to accomplish the ionization. If, however, the desired isotope is excited to a state which lies slightly below the ionization energy, there are alternatives for the ionization step. These include: field ionization by an electric field or by electron impact. These techniques have also been proposed as means of overcoming effectively small photoionization cross sections.

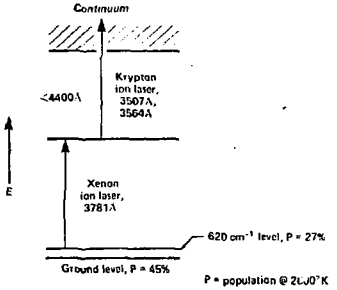

FIG. 2 Excitation Scheme Used in the Photoseparation of Small Macroscopic Quantities of Enriched Uranium

The excitation scheme used in an experiment carried on at Livermore to separate macroscopic quantities of enriched uranium is shown in Figure 2. Two lasers are used for the selective photoionization. A xenon ion laser operating 3781 Å excites uranium atoms from a metastable state at 620 cm^{-1} above the ground state, to an energy level at 27,068 cm^{-1}, approximately one-half of the ionization energy. Excited atoms

are then ionized by the ultraviolet output of a krypton ion laser operating simultaneously on two lines at 3507 Å and 3564 Å. These lines were chosen for convenience and do not necessarily carry the atom to an autoionization state. The terminal state is considerably higher in energy than any of the autoionization states which have been found to date. Therefore, the cross section for this process[1] is expected to be of the order of 10^{-17} cm^2. The xenon laser was chosen for the isotopically selective excitation step because of the fortuitous coincidence between the xenon laser line at 3781 Å and a uranium-atomic transition.

In the experiment, the atomic vapor source was operated at a temperature of 2600°K. At this temperature, approximately 27% of the atoms are in the 620 cm^{-1} state. The ground state contains approximately 45% of the atoms. If a selective photoionization process is to utilize the greatest fraction of the uranium atoms in the vapor stream, then it is necessary to provide a second laser for excitation from the ground level. Since the purpose of this experiment was proof of principle, and not large scale operation, a laser accessing the ground state was not used.

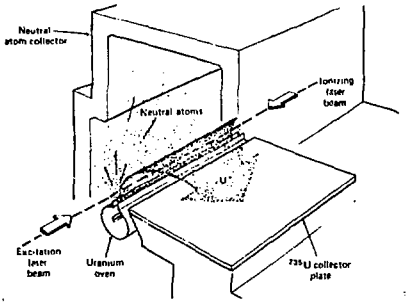

FIG. 3 Geometry of the Photoseparation System

The geometry of the separation experiment is shown in Figure 3. The uranium oven produces a stream of uranium atoms in a fan-shaped beam. The xenon laser beam, passing near the slit as shown, produces the isotopically selective excitation. Ionization of the excited atoms is then produced by the krypton laser beam traveling in the opposite direction. The ^{235}U ions thus produced, are collected on a beryllium plate by an electric field produced by maintaining the collector plate at a negative potential of 1500 volts with respect to the uranium oven. Neutral atoms are collected on a plate provided for the purpose as shown.

The xenon laser beam power in this experiment was approximately 70 mW. The

increased by placing the uranium vapor within the cavity of the krypton laser. In order to accomplish this, the output mirror of the xenon laser served as one of the reflectors for the krypton laser. The circulating power in the ionizing laser beam was between 30 and 50 watts.

The deposition pattern of uranium ions on the collector plate is shown in Figure 4 as determined from densitometer tracings of an autoradiograph of the plate. Approximately 1 mg of material enriched to slightly over 1% in ^{235}U was deposited on this plate. In subsequent experiments, approximately 4 mg of uranium enriched to between 2.5 and 3% has been obtained during a run lasting approximately 2 hours.

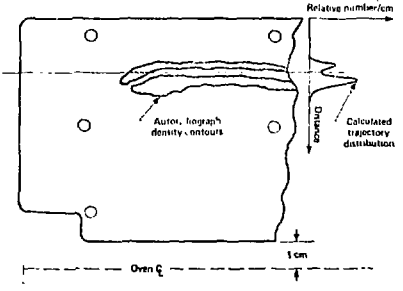

FIG. 4 Uranium Ion Distribution on the Beryllium Collector Plate

Calculations have been carried out to describe the ion trajectories in the collector system. The two-band pattern observed on the beryllium plate is predicted by these calculations and results from distortion of the electric field by the neutral atom collector structure. At the right hand side of Figure 4 is shown the calculated trajectory distribution as a function of distance across the beryllium collector plate.

The quantity and the enrichment of material collected in these experiments was determined primarily by the power of the krypton laser. The number of thermal ions produced by the hot uranium vapor source corresponds to about 0.1% of the total number of atoms in the vapor stream. With the laser powers used in this experiment, enrichment greater than approximately 3% cannot be expected. There was no provision in these experiments for suppression of thermally generated ions to improve the enrichment.

Knowledge of uranium atom spectroscopic parameters is essential in the assessment of the economic feasibility of enrichment processes based on uranium metal vapor. The value of the photoionization cross section is of particular importance

in this regard and a system has been assembled to study the photoionization spectra. In this apparatus, a uranium beam is produced by an atomic vapor source based on the alloy URe_2[1]. The uranium beam is excited to a selected energy level by the tunable output of a CW dye laser. The dye laser, with an output power of about 30 mW, produces sufficient intensity in the beam region to nearly saturate the chosen transition. In a scheme which is very similar to that used to demonstrate selective two-step photoionization, a second, ultraviolet, optical beam intercepts the excited uranium beam. The ultraviolet beam is produced by a 2500 watt mercury arc filtered by a monochromator. As the ultraviolet wavelength is scanned, the rate at which uranium ions are produced is measured. Uranium 238 ions are filtered out from spuriously produced ions of other types by a quadrupole mass filter. By measuring the ^{238}U ion current as a function of the ultraviolet photon wavelength, it is possible to determine the photoionization spectrum related to the selected state of uranium.

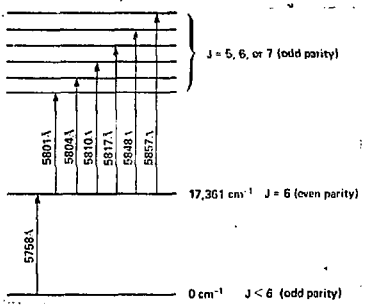

FIG. 5 High Lying Odd Parity Levels of Uranium

A photoionization spectrum obtained at a resolution of 8 Å is shown in Figure 5. This spectrum represents photoionization from a state at 16906 cm^{-1} of the $^7M_7$ level. This is an even-parity state reached from the odd parity $^5L^\circ_6$ ground state by the tunable CW dye laser. The $^7M_7$ level is a level of the f^3dsp electron configuration. From this state, it is expected that autoionization states can be reached corresponding to the electron configuration f^3dp^2.

Several distinct peaks are seen in the photoionization spectrum. In the experiments reported in Reference 1, an average cross section for the region from 3200 Å to 2100 Å of about 10^{-17} cm^2 was deduced for photoionization transitions from the same state. From the spectrum of Figure 5, it appears that the peak cross section for transitions to autoionization states from the $^7M_7$ level is approximately

10 times larger than the background cross section at short wavelengths. The implied value of $\sim 10^{-16}$ cm^2 is large enough to be used in large scale uranium separation systems.

Subsequent measurements at higher resolution have been obtained and it has been found that the peaks shown in Figure 5 have a half width (FWHM) of approximately 3 Å. Photoionization spectra such as that shown in Figure 5 yield quite accurate values of ionization potential of the uranium atom. The value deduced from these experiments is 6.15 ± .02 eV.

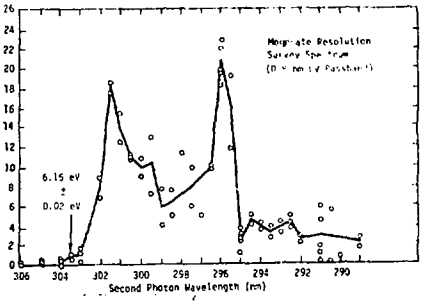

FIG. 6 Autoionization Spectrum from the $^7M_7$ Level at 16,904 cm^{-1} in Uranium

One possible excitation scheme for selective photoionization involves a two-quantum transition to an isotopically selected state. There are practical advantages for such a process in that the effective linewidth of the uranium transition could be narrowed to less than the doppler linewidth[4]. This, in turn, could result in more efficient utilization of the excitation laser. To take advantage of such an excitation scheme, odd-parity levels of the uranium atom lying between 30,000 cm^{-1} and 40,000 cm^{-1} would be required. Very few odd-parity terms in this energy region have been observed and experiments have been undertaken to identify such levels[5]. The experimental arrangement for these experiments was similar to that used in studying the photoionization spectrum. In this case, however, a tunable nitrogen-laser-pumped dye-laser was used for the excitation step. In one set of experiments an even-parity level at 17,360 cm^{-1} was excited using the CW dye laser. The pulsed dye laser operating at over a wavelength range of approximately 5800 to 5960 Å was used in the second and subsequent ionization step excitations. Transitions to odd parity states are readily observed since the same photons which

populate the odd parity levels can also produce ionizing transitions from the odd parity states. Assuming relatively little structure in the photoionization spectrum in the energy region accessed by the photons used, the ion current produced as a function of pulsed laser wavelength should show peaks corresponding to the odd-parity terms. Six of the odd parity levels found by transitions from the 17,360 cm^{-1} level are shown in Figure 6. Utilizing this information, experiments are now underway to measure the two-photon cross sections for transitions from the ground state to these high lying odd-parity states.

Several experiments of importance in the assessment of the feasibility of metal vapor processes for laser photoseparation of uranium isotopes have been described. At this point, there are many additional experiments which need to be carried out before the problems of scaling and economic feasibility of metal vapor processes can be fully addressed.

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