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Research Proposal on Certain Atomic Physics Measurements Associated with the Laser Isotope Separation Method

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W. Duewer

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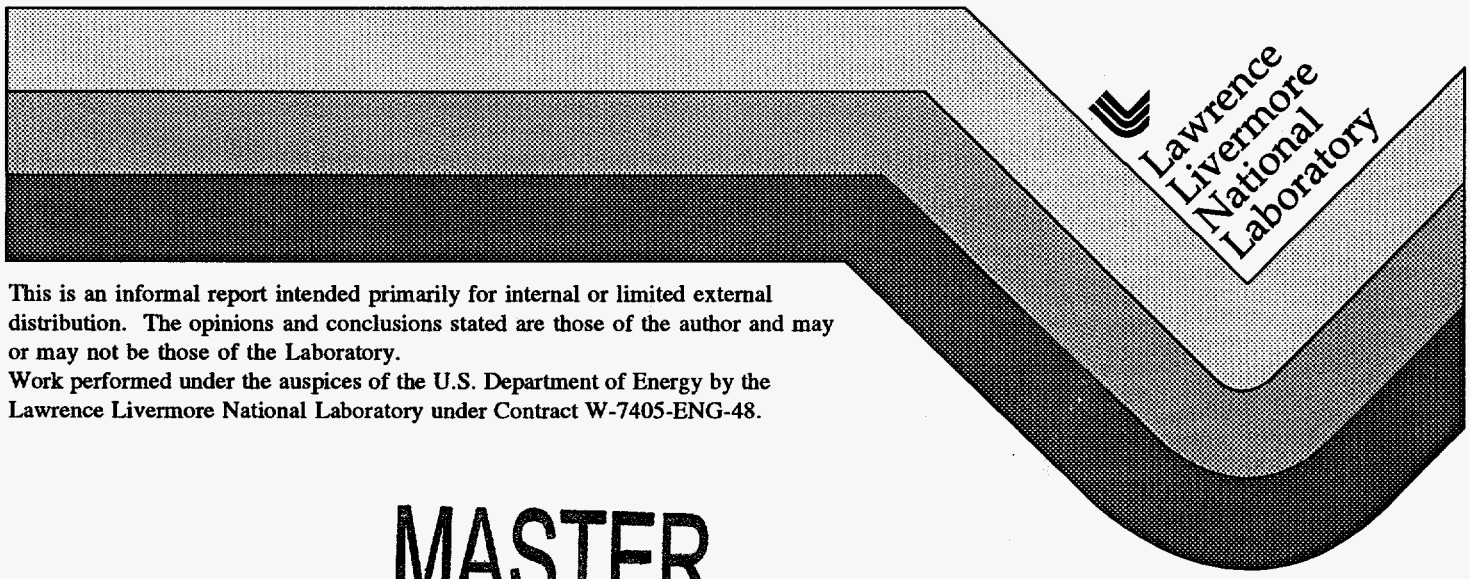

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Interdepartmental letterhead

March 6, 1973

Mail Station 404

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Classified By: [redacted]
Chemist

TO: James Kane and John Emmett

FROM: James Dubrin and William Duewer

SUBJECT: Research proposal on certain atomic physics measurements associated with the laser isotope separation method as applied to U-metal vapor.

The selective photoexcitation of U^{235}, either in the elemental or UF_6 form, has been proposed as a first step in its separation from the heavier, more abundant isotope. In a second step, the electronically excited uranium atom (U^*) is photoionized, while the vibrationally excited UF_6 is photodissociated to give either UF_5 or UF_4. Although photoionization is presently the most promising second step in the metal-atom scheme, other methods including selective surface ionization and chemiionization must also be considered. Needless to say there are many fundamental problems and uncertainties associated with the two-photon approach, in general. Our entire attention in this proposal is directed toward the technique employing U-metal as the starting material.

We have little doubt that the two photon approach will work on the research level. The real question, though, is whether it can be commercially competitive with the existing methods, and the answer to this is totally unclear to us at this stage. Its success must certainly depend on the future ability to fabricate powerful, but highly efficient and stable,

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tunable visible lasers with narrow line widths. At nearly the same level of importance is the need to know the radiative lifetimes of a number of the electronically excited states of uranium.

The studies proposed here are primarily concerned with the second step of the process. Theoretical calculations on the cross sections we wish to measure may be useful to predict general trends, but we are rather skeptical of their utility beyond that.

The modest program outlined below would generate an important fraction of the input data necessary to clearly differentiate the many possible variations of the two step process from both a standpoint of feasibility and cost.

Proposed Work

Three major experimental investigations are planned. Common to all of these is a standard, differentially pumped molecular beam apparatus consisting of a main scattering chamber, source chambers housing the neutral and ion beam sources, and finally a detector chamber enclosing a medium resolution-mass spectrometer. The vacuum requirements are minimal (10^{-6} torr) and furthermore the necessary electronics and other equipment are readily available. This very happy circumstance is a direct result of requiring only ion signal measurements for the cross section determinations. A significant problem will be the production and use of high temperature U-vapor sources. Fortunately, expertise in this area already exists at the Laboratory.

A. Measurement of Photoionization Cross Sections Near Threshold For The Excited States of U.

Photoionization cross sections of ground state atoms are typically two to three orders of magnitude less than the characteristic gas

kinetic value, 3×10^{-15} cm^2. However, atoms populated in certain excited states can have cross sections considerably larger than the ground state values. Moreover, near threshold, these cross sections can be expected to display rather sharp resonances corresponding to absorption into autoionizing states, and the maximum cross section for such a transition may be orders of magnitude larger than the off-resonant background (continuum absorption).

To establish these cross sections selected, excited states of uranium, formed by pumping a U-beam with a dye laser, are subjected to variable monochromatic radiation (bandpass $\sim$ 0.1eV) from a high intensity lamp. By simultaneously monitoring the visible photons from the decay of U^*, and using the radiative lifetimes of this state from separate measurements, the photoionization cross section follows directly. Quite conservative feasibility calculations indicate that cross sections ca. 10^{-18} cm^2 can be measured without difficulty.

B. Measurement of the Symmetric Charge Exchange Cross Section: U^+-U.

At low relative kinetic energies, charge transfer cross sections of the above type are far greater than the geometric or gas kinetic value, especially if the atom-ionization potential is small as is the case for U. This naturally represents a serious problem in the two photon method as applied to U vapor, for its occurence will effectively destroy the selectivity achieved in the first step. To illustrate: If the charge exchange cross section were 5×10^{-14} cm^2 at 0.3eV(ca. 2000°C), a value which would not surprise us, about 98% of the U^{235+} would exchange with the U^{238} in traversing a 2 cm-path of U vapor at a pressure of 10^{-2} torr! The experimental determination of the charge transfer cross sections will be achieved using the conventional technique

of counting the slow ions produced in the encounter. The neutral beam intensity in all likelihood will be monitored with a surface ionization detector, consisting of an "oxygenated" tungsten wire. A limited amount of time will be devoted to calculating the cross sections with the aid of simple theoretical models.

C. Measurements of Chemiionization Cross Sections for Electronically Excited U Atoms.

An alternative to photoionization as the second step would be the removal of the U^{235*} through reaction with another species to give an ion, e.g., $U^* + O_2 \rightarrow UO_2^+ + e^-$. Of course the rate of chemiionization of the ground state uranium must be much slower. There are three advantages that this chemical reaction scheme holds over photoionization: (1) The cost per "particle" is considerably less for molecules (e.g., O_2) than photons; (2) Asymmetric charge transfer cross sections normally are markedly smaller than the symmetric cases (UO_2^+ vs. U^+); (3) Chemiionization cross sections for exoergic reactions can be as high as 2×10^{-14} cm^2. The very major disadvantage is that the upper limit on the molecular flux, set by vacuum requirements, is substantially less than the photon flux attainable by present day lasers.

Chemiionization cross sections for both the ground and various excited states of U will be measured employing almost the exact same procedure and analysis as outlined under A. The trivial modification is the substitution of the photoionizing light beam by a molecular beam. Since there is documented evidence†, that uranium

† W. L. Fite and P. Irving, J. Chem. Phys., 56, 4227 (1972). [No absolute cross sections were obtained, and in fact it is unclear whether U or U^* was the major precursor to the observed product ion, UO_2^+]

atoms undergo chemiionization with molecular oxygen, we would initially concentrate on the $U(U^*)$-O_2 collision systems. Other candidates such as N_2O, NO_2 and NO would also be examined.

Support and Personnel

On the basis of past experience in molecular beams, we estimate that 75% of the overall effort will be devoted to the construction of the apparatus and the remainder to data acquisition. (Thus for example the elimination of single study (A, B, or C) will not materially reduce the cost or time).*

Personnel

1.	J. Dubrin	1 man years
2.	W. Duewer	1 " "
3.	Technician[a]	1 " "
4.	Expert 1[b]	1/8 " "
5.	Expert 2[c]	1/4 " "
6	Shop Support[d]	1-1/8 "

a. To aid in: Apparatus assembly, Major component testing, Data acquisition.

b. U-beam sources

c. Dye lasers

d. Machine, glass, and electronic

*Much of the same apparatus can be used in a measurement of the radiative lifetimes.

Estimated Costs of Major Components

1.	Vacuum system	$ 5,000
2.	Laser and beam monitor*	25,000
3.	High intensity light sources, filtering systems and detectors	4,000
4.	Fluorescence monitoring system, including counting system	5,000
5.	Mass spectrometer,* including counting system	20,000
6.	Signal averaging electronics	10,000

* May be available

James W. Dubrin

William H. Duewer

JWD:WHD:ob

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