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## The laser isotope separation program at lawrence livermore laboratory

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**THE LASER ISOTOPE SEPARATION PROGRAM AT  
LAWRENCE LIVERMORE LABORATORY**

**R. C. Stern and B. B. Snavely**

**April 9, 1975**

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THE LASER ISOTOPE SEPARATION PROGRAM AT  
LAWRENCE LIVERMORE LABORATORY\*

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The projected demand for enriched uranium will exceed the projected supply from existing facilities in the mid 1980's. Beyond that point, if no improvements in technology are made, a > 3 billion dollar gaseous diffusion plant will have to be brought on line every 18 months. The development of tuneable narrow band lasers has opened the prospect of large scale laser isotope separation (LIS) at a fraction of the gaseous diffusion power and capital equipment costs.

This talk will present an overview of the Livermore LIS program as it looks today - by describing some of the research projects completed in the past year or actively contemplated, in the sense that equipment is in hand or under construction, for the next. Given the large number of photoprocesses that can potentially separate isotopes, the challenge is to assemble a data base of fundamental cross sections so that the feasible schemes can be culled from the unlikely ones.

PROCESS SURVEY

The starting point for a laser isotope separation is the selective photo-excitation by 1 or more photons to produce a population of the  $^{235}\text{U}$  species in an excited state. This process obviously depends on the level positions and their isotope shifts -  $< 1 \text{ cm}^{-1}$  due to nuclear volume effects in U atom (Figure 1) - up to a few parts in  $10^{-4}$  from vibrational reduced mass effects in molecules. The microscopic steps involved in several photoseparation processes are displayed in Figure 2. Excitation competes with spontaneous emission and, depending on the system, internal conversion, collisional quenching and near resonant exciton transfer.

The transient selectively excited population can be more permanently labeled -  
\* This work was performed under the auspices of the U. S. Energy Research Development Administration.

so that separation is possible - in several ways:

1. Direct photoionization using one or more non-selective photons.
2. Indirect photoionization using field or collision induced ionization of a high Rydberg level.
3. Dissociation of a molecular excited state.
4. Chemical reaction - ie: classical photochemistry - to produce neutral or ionic products.

Each technique depends on the parameters which define its intensity: photoabsorption, chemical reaction and photoionization cross sections, lifetimes with respect to dissociation and field ionization; parameters which define competing channels: photoemission lifetimes, cross sections for collisional relaxation or quenching; and parameters which describe the scrambling of the selective label: excitation transfer, charge exchange or reactive atom transfer cross sections. In addition, a successful process requires the design of vapor sources, of product collection techniques and the development of efficient lasers.

#### PHOTOIONIZATION OF U ATOMS

The separation of  $^{235}\text{U}$  on a microscopic scale using selective ionization was announced by the Livermore group several months ago.<sup>1</sup> The apparatus is shown in Figure 3. Here a tuneable dye laser is used for the selective excitation and an Hg-arc lamp for the ionizing photon. Ions are detected in a quadrupole mass spectrometer.

The crucial parameters which determine the intensity required from the ionizing laser in this scheme are  $\tau\sigma_i$ , the product of the intermediate state radiative lifetime and the photoionization cross section from that level. Both these quantities are under investigation.

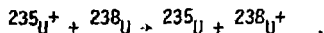
We will shortly be measuring lifetimes for a number of U atom excited states by monitoring the fluorescence decay following pulsed dye laser excitation.

Photoionization cross sections are being measured by detecting ion currents

as a function of UV wavelength in an experiment shown in Figure 4.<sup>2</sup> As may be seen in Figure 5, structure in the ionization continuum, corresponding to autoionizing levels of the atom, has been observed with peak ionization cross sections > an order of magnitude above the continuum average.

This work has benefited from a program of extended Hartree-Fock calculations on U atom which have estimated energy level locations and transition probabilities.

In any photoionization scheme, there is the possibility of losing isotopic selectivity by the near resonant charge exchange process:



The successful separation experiments permit a crude upper bound of  $\sim 500 \text{ \AA}^2$  to be deduced for this cross section. Given the psychological significance of this number, a series of U atom charge exchange measurements are currently under contract.

Since the U atom ionization potential is 6.14 eV, 2 photons with  $\lambda < 4000 \text{ \AA}$  are required to photoionize. With the current laser technology, lasers operating at longer wavelengths may well be more efficient - hence an interest in ionization with > 2 photons - one of the schemes shown in Figure 2.

An experiment directed at such processes divides a CW dye laser into 2 oppositely directed beams to measure Doppler-free 2 photon absorption in U vapor. The energy level density in U atom makes it likely that large 2-photon cross sections can be found. The technique will map odd parity levels including some at energies near the ionization limit.

An important component of all work on U systems has been the development of high temperature sources for atomic vapor production by members of the Livermore Inorganic Materials group under the direction of Oscar Krikorian. One convenient source exploits the lowered chemical activity of the compound  $\text{URe}_2$ , Figure 6. A photomicrograph of a  $\text{URe}_2$  source after extended use, Figure 7, shows only minimal attack of the tungsten boat by the alloy - as opposed to the extensive corrosion which would occur for a U in W source.

Finally, several modeling efforts are underway: component modeling on oven heat losses and the plasma physics of ion extraction by electromagnetic fields, and the third cycle of economic modeling on the system as a whole - with results that are encouraging in all cases.

#### PHOTOPHYSICAL PROCESSES IN MOLECULES

In comparison with atom based schemes, molecular processes have the advantage of working with more volatile compounds which have potentially larger isotope shifts and the disadvantage that the spectra involved are complex with low populations in any single level. The gas phase spectroscopy of most U compounds is unknown. Thus the first task has been survey spectroscopy on several U tetrahalides, oxyhalides and oxides. A sample spectrum, 1.0 to 1.3  $\mu$  absorption by gas phase  $UCl_4$  is shown in Figure 8. In one case, this spectroscopy, and the accompanying calculation of molecular level populations, has been sufficiently promising that detailed isotope shift measurements will follow. An important part of this effort is the development of sources which produce the compounds in the gas phase - essentially an experimental exercise in the complex condensed phase chemistry of U compounds.

#### COLLISIONALLY DRIVEN PROCESSES

In classical photochemistry, the excitation is converted into a permanent label by a selective chemical reaction - one that proceeds for the excited states but not for the ground states. Photochemical (as opposed to photophysical) processes have some obvious advantages. One does not have to excite into a continuum (small cross sections). The lower state hole burned by the laser

can be collisionally filled, allowing high single pass conversions. On the other hand, once collisional contact is established any excitation becomes more transient, i.e., the system relaxes to equilibrium through a greater number of channels. Extensive modeling of photochemical systems has established sets of operating conditions where photochemistry can be relatively quite efficient. A typical set of levels and rate processes are shown in Figure 9. Given the short lifetimes of U atom excited states, models of the atomic vapor chemistry optimize at high scavenger number densities, Figure 10.

Underway at present are experiments on the associative ionization of  $U^*$ . We have begun construction of a molecular beam facility for the study of gas phase neutral-neutral photochemistry of U compounds. From the thermodynamics one can predict systems with small ground state reactivity. It remains to be seen what the excited state reactivity will be.

A component of the kinetics effort is the investigation of low temperature sources of U atoms and small molecules. Possible means of doing this include pyrolytic, photolytic or discharge cracking of volatile U compounds. One fundamental problem faced by many low temperature sources is the time limit set by condensation. For example, 1 torr of U atom vapor at 1500° K is supersaturated by  $10^{27}$ . Studies of the condensation kinetics of U vapor in several carrier gases are under contract.

#### LASER DEVELOPMENT

Last but by no means least, one leg of the Livermore program is the development of lasers to drive the various separations. Areas of interest are incoherently and coherently pumped dyes and fortuitously overlapping fixed frequency lasers. Incoherent pump concepts under development include flow stabilized lamps and imaging pumped cavities using high speed transverse dye jets. The configuration used in experiments on the latter system is shown

in Figure 11. Experimental results are combined into extensive modeling efforts which enable us to evaluate the potential efficiency of classes of laser systems. The results for incoherently pumped dye lasers, Figure 12, are encouraging. Coherent pumps include high power CW ion lasers and pulsed  $N_2$  and Cu vapor lasers. The last system looks particularly promising on the basis of both efficiency and economics. A general concern is the tight control of laser center frequencies and linewidths. The laser development has and will run in close cooperation with the laser R&D industry with major joint efforts anticipated in the future.

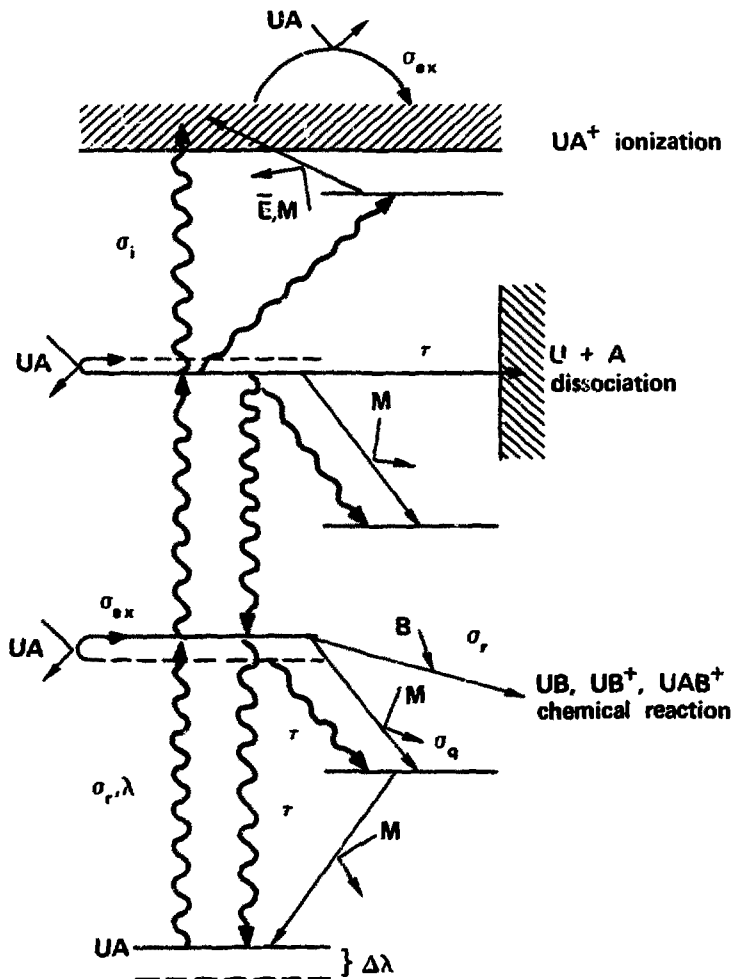
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2. R. Solarz, L. Carlson, and C. May, "Excited State Spectroscopy of Atomic Uranium", invited talk at the 1975 Spring Conference of the Optical Society of America; UCRL 76306.

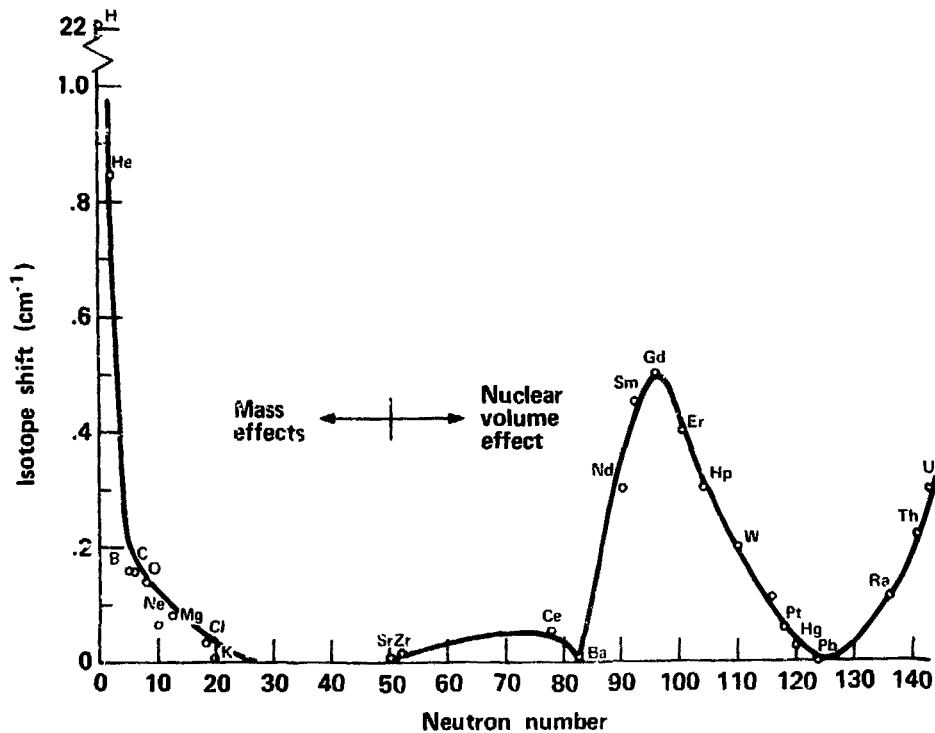


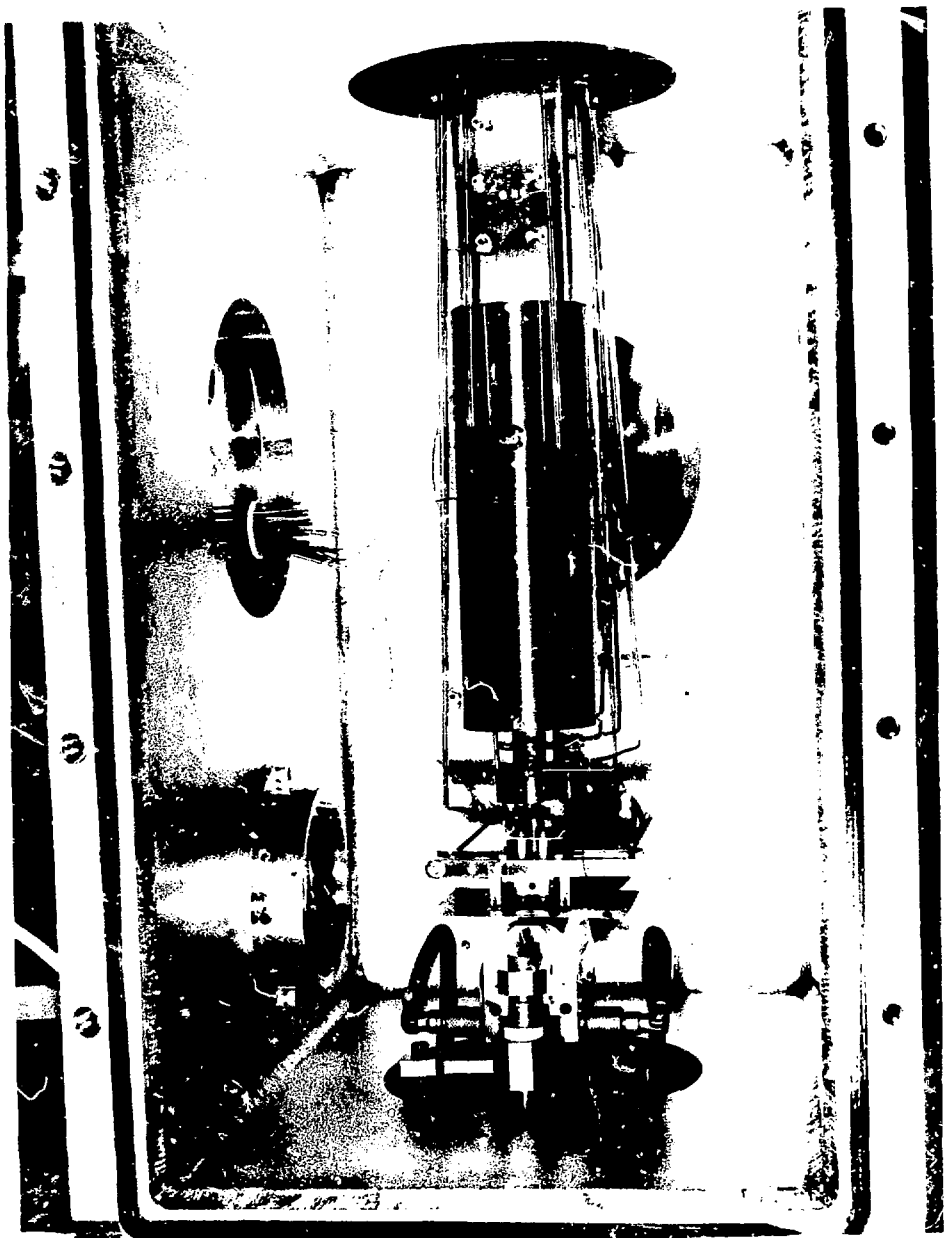
FIGURE CAPTIONS

- Fig. 1. Atomic isotope shifts.
- Fig. 2. Microscopic channels in laser isotope separation: M = inert gas, B = chemical scavenger,  $\sigma_Y$  = photoabsorption cross section,  $\sigma_q$  = quenching cross section,  $\sigma_r$  = reaction cross section,  $\sigma_{ex}$  = exchange cross section,  $\tau$  = lifetime,  $\Delta\lambda$  = isotope shift.
- Fig. 3. Apparatus for selective photoionization: U vapor oven to right (inside shields), quadrupole mass spectrometer to left.
- Fig. 4. Schematic of the apparatus for photoionization measurements.
- Fig. 5. U ion signal as a function of UV wavelength.
- Fig. 6.  $URe_2$  in the U - Re phase diagram.
- Fig. 7. Photomicrograph of a  $URe_2$  filled source after use: top, recrystallized  $URe_2$ ; bottom, W boat; center, narrow zone of alloy attack.
- Fig. 8. Survey spectrum of a nominal  $UBr_3Cl_2$  sample. Absorption is due to  $UCl_4$ .
- Fig. 9. Levels and channels for kinetic modeling of uranium photochemistry.
- Fig. 10. Model results for the time for 99% conversion of U atoms to molecules as a function of laser intensity and scavenger density.
- Fig. 11. Configuration used in experiments on flash-lamp pumped dye lasers.
- Fig. 12. Summary of experimentally calibrated modeling calculations on flash-lamp pumped dye lasers.

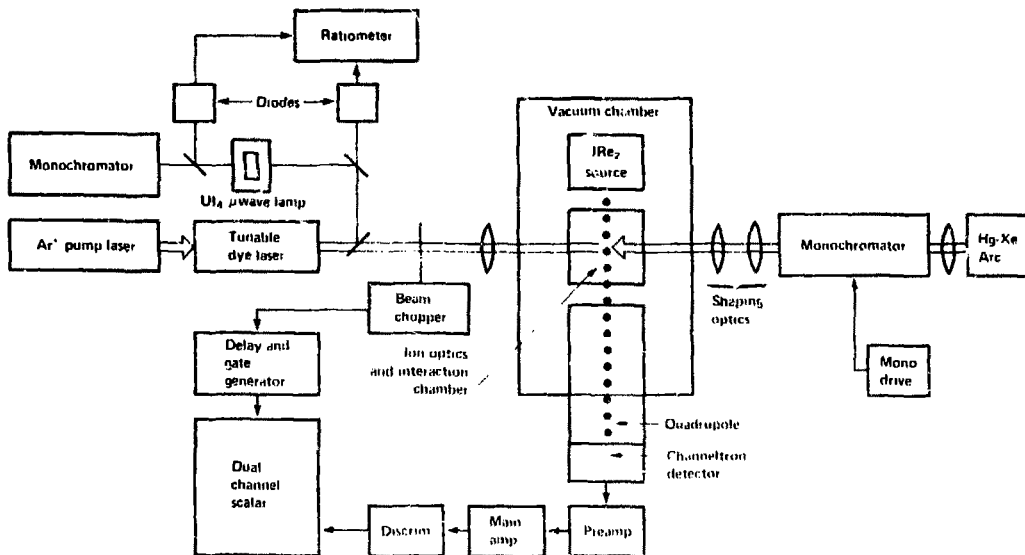


# ISOTOPE SHIFT VS NEUTRON NUMBER

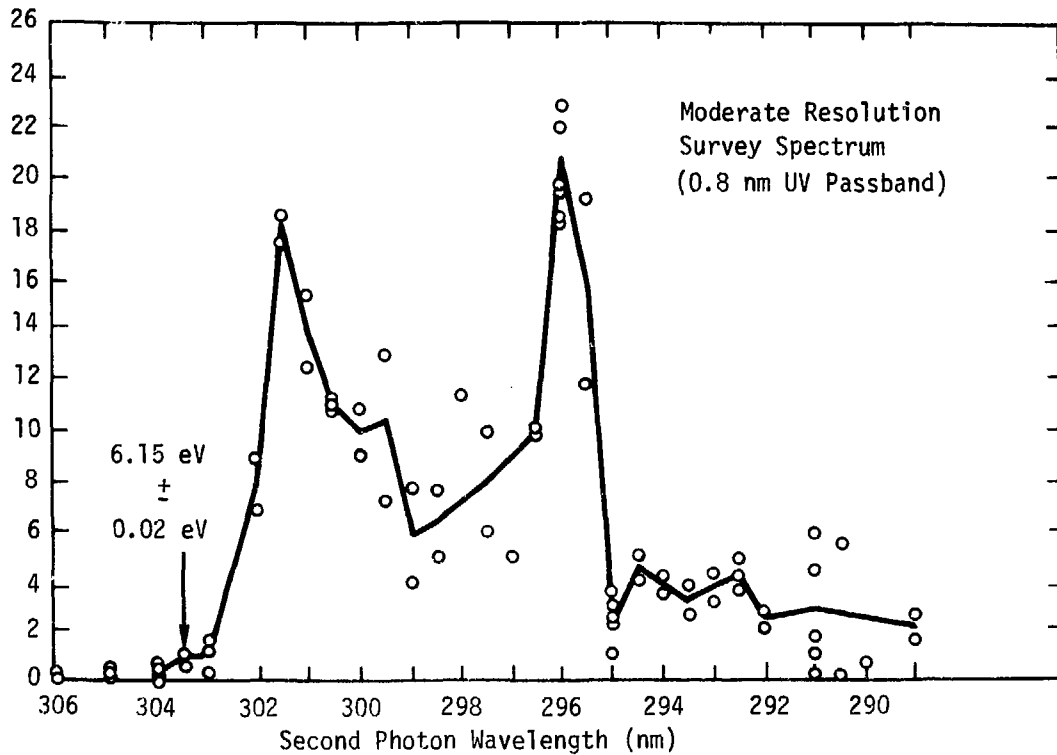




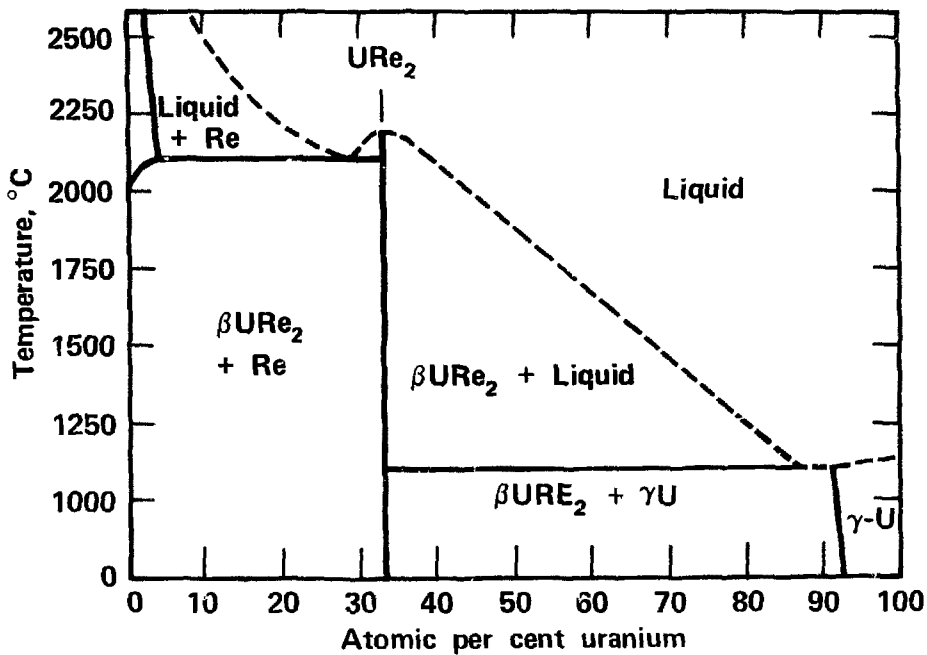
# AUTOIONIZATION APPARATUS



AUTOIONIZATION FROM  $7M_7$  AT 591.5 nm IN ATOMIC URANIUM VAPOR



# PHASE DIAGRAM FOR U-Re

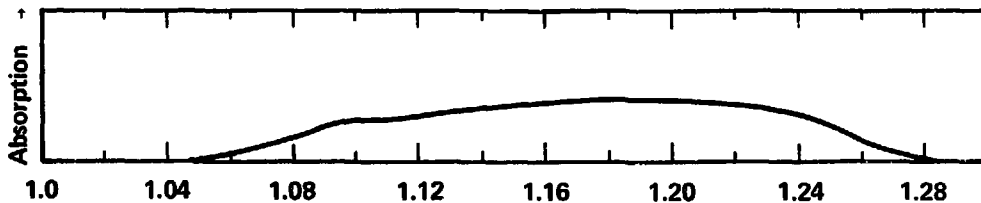






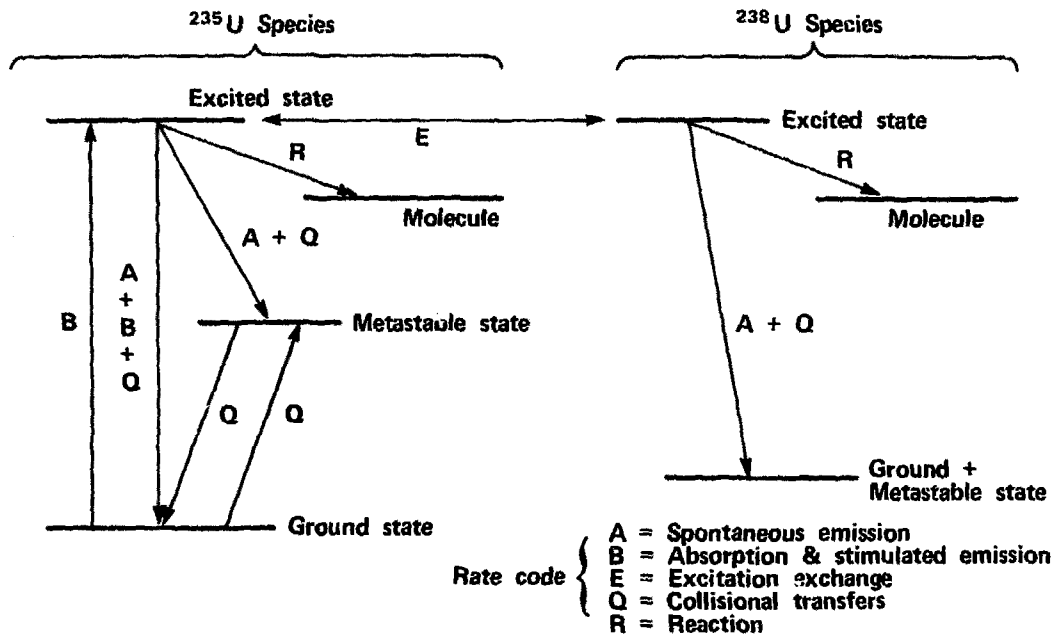
**ABSORPTION OBSERVED WITH  $\text{UBr}_3\text{Cl}$  SAMPLE**

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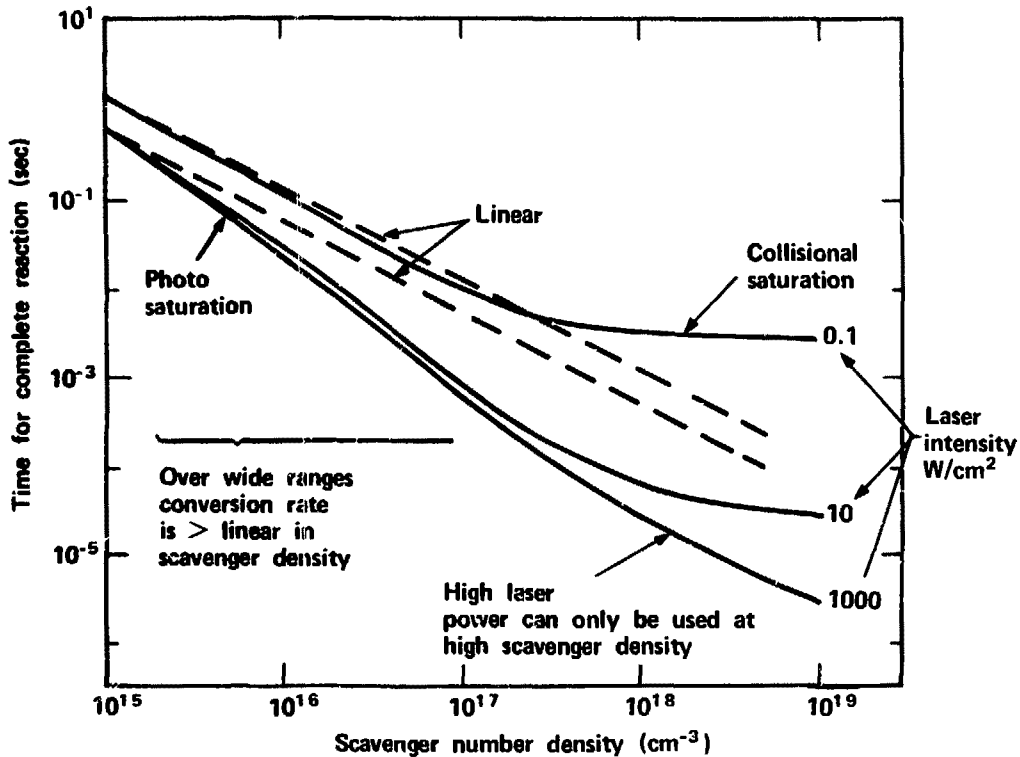


Cell temperature = 600°C  
Cell length = 60 cm  
Scan rate = 100 Å/min  
Phase sensitive detection used  
Absorption due to  $\text{UCl}_4$

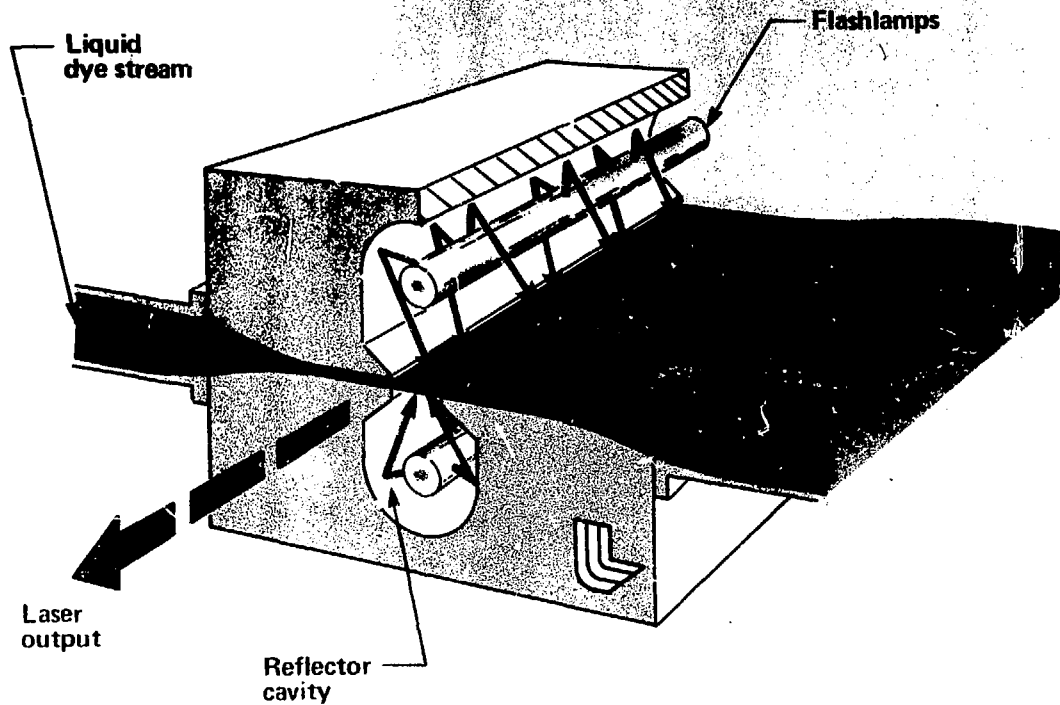
# 7-LEVEL KINETICS WITH EXCITATION EXCHANGE



# CONVERSION TIMES VS SCAVENGER DENSITY



# DEVELOPMENTAL DYE LASER



## EFFICIENCY ESTIMATE FOR INCOHERENTLY PUMPED LASER

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● Dye quantum yield	0.95
● Dye efficiency, as a laser	0.20
● Laser mode coupling	
Hard aperture direction	0.38
Soft aperture direction	0.80
● Excitation absorption	0.75
● Stokes shift	0.60
● Lamp efficiency	0.15
● Pump cavity coupling	0.50
○ Overall efficiency estimate	$\approx 0.001$