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MASTER

EXCITED STATE SPECTROSCOPY OF ATOMIC URANIUM

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and Charles May
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Stanford University
Palo Alto, California
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This concludes the experimental portion of this talk. I will next give justification for the research effort on uranium laser enrichment processes.

That there is a need to develop enrichment processes which will expand present enrichment capacities and simultaneously improve upon the economics of the gaseous diffusion plants is questioned by no one. It is generally agreed that by the early 1980's additional uranium enrichment capacity will be required in order to fuel the number of reactors expected to be in operation at that time. The promise of laser photoenrichment is obvious - which laser photoenrichment scheme will be used ultimately is, at this point, not as obvious. At present the laser enrichment work is concentrated primarily on the identification of which schemes are most promising, that is those schemes which will yield economic benefits in a scaled-up plant size operation. The identification process is hindered on two fronts. The first problem is that happily there are a near infinite number of schemes to consider. The second problem is that the data base, spectroscopic and kinetic, on atomic uranium and uranium compounds, is unhappily in a rather primitive state. Even data on vapor pressures of many uranium compounds is lacking. In an exercise where each new proposed scheme typically requires the measurement of three or four critical parameters, this is a situation which must be rapidly alleviated. The motivations for investigating selective photoionization of uranium as a method of enrichment should follow naturally from this discussion. It is the enrichment scheme for which success on a short time scale is most obvious due to both the general simplicity of atomic schemes and the comparatively advanced state of atomic uranium as opposed

* This work was performed under the auspices of the U. S. Energy Research Development Administration.

to molecular uranium spectroscopy and kinetics. It is quite probable that ultimately some atomic or molecular kinetic scheme will be of even greater use than photoionization - whether such schemes can be discovered and scaled in the required time scale is difficult to answer. Yet I now hope to demonstrate that even photoionization schemes may be economically attractive successors to gaseous diffusion or centrifuge plants (slide - Davis Table 1). Most of the analysis which I will now outline is due to Rich Davis and Mary Spaeth at Livermore.

Now that I have outlined the spectroscopic behavior of atomic uranium, I would like to discuss the impact of each of the atomic parameters on a specific separation scheme, namely photoionization of uranium vapor. The next slide outlines the spectroscopic loss mechanisms in the photoionization process. First of all, as a result of the high oven temperatures, not all the U^{235} exiting from the oven slit is in the ground (cm^{-1}) state. At 2600°K only 45% of the uranium is in the ground state with the bulk of the remainder lying in the 620 cm^{-1} level. It is clear that a three laser scheme, in which the third laser is added to bring the population of the 620 cm^{-1} level to the same excited state used for the ground level ions, is an important wrinkle in the photoionization scheme. A further loss mechanism is illustrated by the decay of atoms from state 2 to a metastable level at rate W_{21} before they undergo ionization. Decay rate W_{10} is typically much slower than W_{21} and the collective levels 1 represent an energy sink in the scheme, atoms decaying to these levels are not affected by the lasers and do not undergo ionization before they thermally migrate from the laser ionization area.

To perform an economic analysis on a large scale uranium separation unit, the first step is to identify the largest complete module which can be constructed which is:

- a. Permitted by the physics of the separation scheme and is
- b. compatible with a predefined output tails assay when combined in a series or paralleled with other modules of the same design. Oven and laser conditions resulting in minimum costs per separated U^{235} atom are then identified.

Consider the oven geometry shown in the next slide (slide - Davis Oven). The oven module of length L is used to vaporize the uranium through a slit of height and thickness H_2 and H_1 . Let us choose an oven length of 4 meters in order to minimize laser beam divergence and alignment problems as well as fabrication difficulties. H_1 is chosen to be 1 mm (the laser beam diameter) and H_2 we choose to be several millimeters. H_2 is limited by charge exchange considerations and available laser beam intensities.

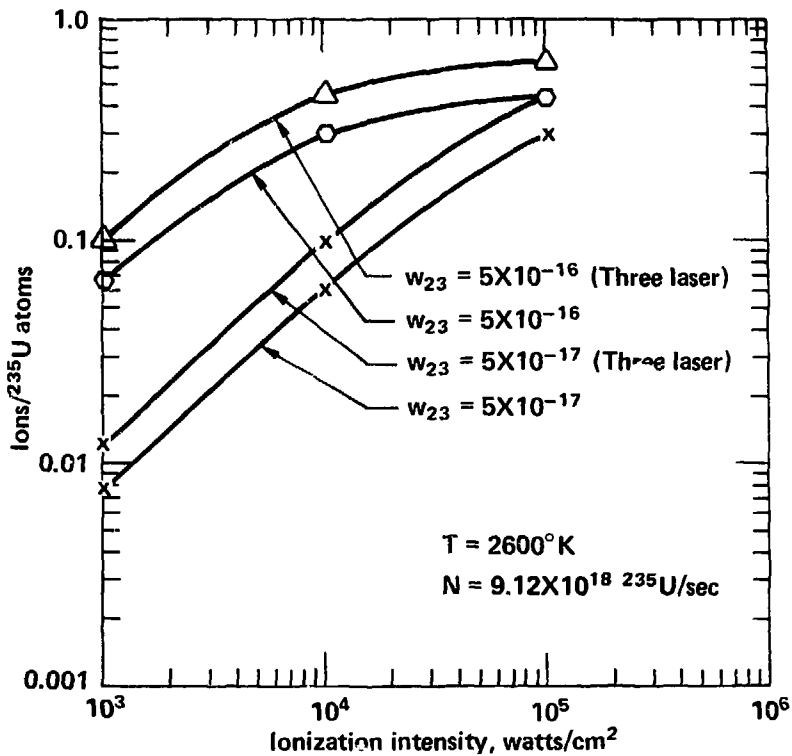
Figure three is the oven energy loss divided by the number of U^{235} atoms exiting the slit. To convert this to energy loss per separated ^{235}U atom, the percentage of ^{235}U atoms ionized and not subsequently undergoing charge exchange must also be accounted for. Note the diminishing importance of oven considerations with increasing temperature and not the importance of the 620 cm^{-1} levels to the overall process. Figure four shows the fraction of ^{235}U atoms ionized as a function of ionization cross sections and separation scheme. With an ionization cross section of 10^{-16} cm^2 and a three laser scheme roughly $3 \times 10^4\text{ watts/cm}^2$ are required to ionize 50% of the U^{235} atoms. Figure five shows the oven and laser energy expenditure at 2800°K per separated atom as a function of ionization laser intensity and ionization cross section. The major assumptions in this figure are 0.1% laser efficiency, a factor of 10 enhancement in laser intensity as a result of optics in the ion chamber and an excitation intensity of 100 W/cm^2 when $W_{23} = 5 \times 10^{-17}\text{ cm}^2$ and $T = 2800^\circ\text{K}$. It is abundantly clear that good ionization cross sections and good laser efficiencies are crucial to the overall separation. I wish to emphasize that this is only a two photon separation scheme. The last slide is a comparison of separative work cost for several isotope separation schemes (the Becker nozzle is not included). It shows that when capital equipment, energy consumption, and construction funds are all totaled, even the two photon photoionization scheme becomes competitive although only marginally so. Laser efficiencies, autoionization and charge exchange cross sections

are all of vital importance. Clearly the development of a low temperature source would be beneficial, although the present sources do not present an insurmountable problem. The point is that given existing equipment and technology, the photoionization process is clearly worth pursuing. Also, breakthroughs in any of a number of areas could result in further improvements.

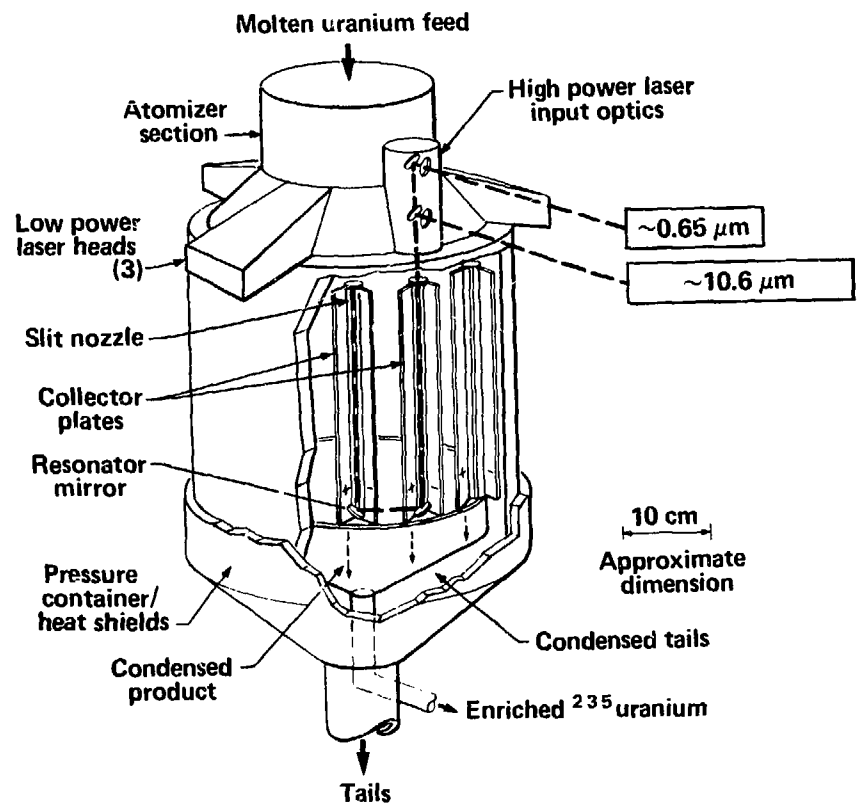
To summarize, laboratories investigating laser photoenrichment of uranium are still in the midst of sorting through the numerous proposed separation schemes. *Photoionization is among the separation schemes which have received the most attention in the laboratory and a realistic economic analysis indicates that even this obvious scheme may be economically competitive with more conventional separation methods.* Furthermore, the first cut economic analysis is a pessimistic one and the addition of one or more lasers to the two photon scheme will add considerably to its attractiveness. Progress in laser development and accelerated programs in uranium spectroscopy and kinetics in the next few years will be crucial in determining whether or not laser photoenrichment schemes evolve into full scale enrichment plants.

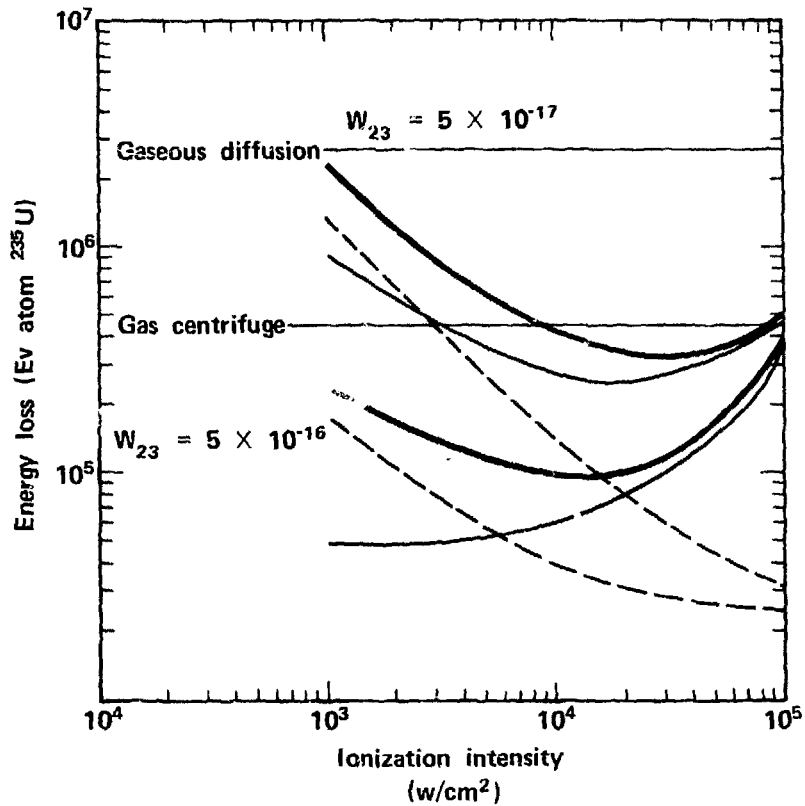
UTILIZATION RATIO VS IONIZATION INTENSITY

Two-photon process



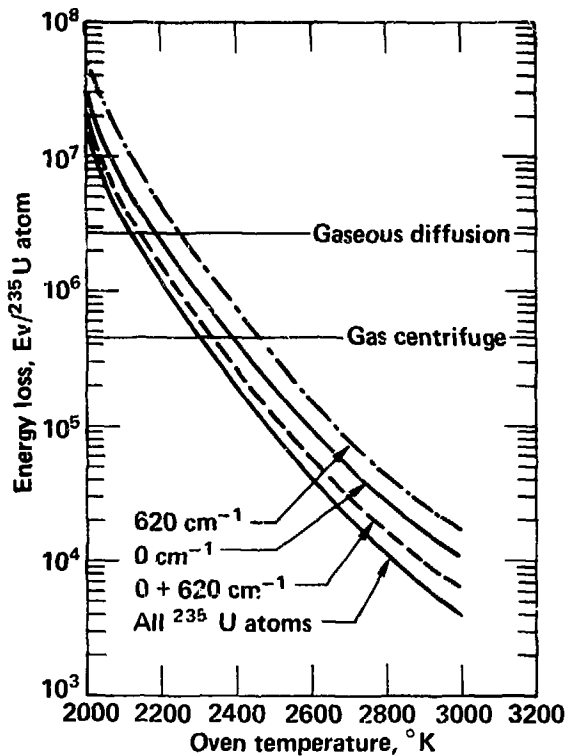
LASER ISOTOPE SEPARATION MODULE



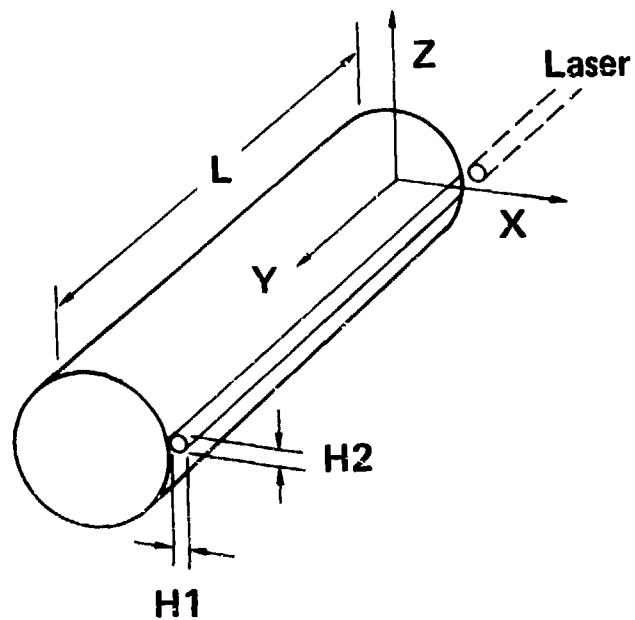


RADIATIVE ENERGY LOSS

Two-photon process



BASELINE OVEN GEOMETRY



SEPARATIVE WORK COST COMPARISON



	<u>Gaseous</u> <u>diffusion</u>	<u>Gas centrifuge</u> <u>US</u>	<u>European</u>	<u>Laser</u> <u>photoseparation</u> <u>N-photon</u>
Capital cost \$M	1050	1180	1500	162
Total power MW	2000	350	260	210
Depreciation \$	38	64	80	9
Power \$	97	16	12	10
Operating \$	8	24	40	16
Interest \$	18	—	—	9
Contingency \$	24	16	20	6
Total (\$/kgu product)	185	120	152	50
Total (\$/kg-swu)	39	25	32	11