MASTER

PREPRINT UCRL- 78549

CONF-760832--20

Lawrence Livermore Laboratory

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August 1976

THIS PAPER WAS PREPARED FOR SUBMISSION TO THE SPIE CONFERENCE, SAN DIEGO
AUGUST 23-27, 1976

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LASERS FOR ISOTOPE SEPARATION PROCESSES AND THEIR PROPERTIES*

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Abstract

The laser system requirements for isotope enrichment are presented in the context of an atomic uranium vapor process. Coherently pumped dye lasers using as the pump laser, either the frequency doubled Nd:YAG or copper vapor are seen to be quite promising for meeting the near term requirements of a laser isotope separation (LIS) process. The utility of electrical discharge excitation of the rare gas halogens in an LIS context is discussed.

Introduction

The strong interest in isotope enrichment using lasers can be put into perspective by considering the fact that estimates of the demand for new enrichment capacity through the late 1980's and early 1990's call for a new 9,000 tonne (1 tonne = 1,000 kg) separative work plant every 18 months. On the basis of current projections, 9 new plants of this size may be required by the year 2000, at a cost of three to four billion dollars each, using current gaseous diffusion or gas centrifuge technology. Since the Laser Isotope Separation (LIS) process has a much higher separation factor than either the gaseous diffusion or gas centrifuge process, high product assay may be achieved in a few separation stages instead of a cascade system. As a result, LIS plants have the potential to be small and relatively inexpensive. Preliminary estimates show that the construction cost of an LIS plant may be approximately three to five times less than for a gaseous diffusion plant. The potential savings through the year 2000 could be many billions of dollars in capital investment and operating costs.

The economic viability of laser photoseparation will depend upon several factors which are peripheral to the enrichment process itself. Principle among these is the development of high-power tunable lasers. Clearly numerous processes will prove to be technically feasible; however, economic feasibility will depend upon the scaling of the lasers used to drive the process.

Potentially useful uranium laser enrichment process can be configured using as the feed material uranium as a free atom or in some molecular form. In this work we shall discuss those aspects of the laser sub-system which pertain to the atomic uranium vapor process. The details of such a scheme will not be discussed here; it suffices to say that in general several photons (typically at different frequencies) are employed to selectively place the desired uranium isotope (U_{235}) in a highly excited state. This electronically excited isotope is subsequently ionized (using another laser for example) and then extracted from the interaction region in some fashion. Since the ionization potential of uranium is 6.2 eV, a multiphoton process will typically utilize lasers which span the visible portion of the spectral region.

The purpose of this paper is to outline a number of the laser requirements and to scope operating parameter ranges for some of the principle laser systems where possible. We begin with an abbreviated statement of nominal laser system requirements, followed by a review of the status of the more promising laser candidates.

Laser Systems Requirement

The average power requirement for each laser of the multi-step process, is a function of the number of LIS modules which might be present in a commercial plant. It is quite straightforward to show that the total laser power P in watts, which is required for a given plant module, assuming that it produces 0.2% tails, is given by the following relationship:

$$P = 5.67 \times 10^{14} \frac{Nh\nu}{\eta_\phi \eta_c} \qquad (1)$$

*This work was performed under the auspices of the United States Energy Research and Development Administration. Contract W-7405-Eng-48.

Here N is the module output in SWU†/yr, η_ϕ is the probability that a photon which enters the separation chamber will be absorbed by the desired isotope, η_c is the probability that an atom (or molecule) which has absorbed a photon will go on to be collected and not lost due to an exchange or undesired decay channel and $h\nu$ is the photon energy in joules. As an illustrative example; the laser requirements for a miniplant module with a throughput of 50×10^3 SWU/yr (roughly 1/200th the throughput of the present Oak Ridge facility) assuming a total utilization efficiency $\eta_\phi \eta_c = 0.1$ and a 4000 Å photon, turns out to be approximately 100 watts.

Another parameter of importance is the overall laser efficiency denoted by η_L. This quantity is intimately related to the efficiency with which the laser isotope separation process uses the available photons and to the maximal energy cost per step per separated atom E_ϕ which can be economically tolerated. The relationship is written in the following fashion:

$$E_\phi = \frac{h\nu}{\eta_L \eta_\phi \eta_c}. \tag{2}$$

To be competitive with other isotope separation processes E_ϕ should lie nominally at 30 keV per separated atom (e.g. gaseous diffusion is ~ 3 MeV per separated atom). From our previous example η_L turns out to be about 10^{-3}.

While we have obtained in quite general terms several of the important laser system requirements, many more remain. Strictly speaking however, these remaining laser requirements such as beam quality, spectral distribution, pulse width, repetition rate, etc., depend upon the actual point design (for example source and extractor configurations) chosen for the LIS process. Table I quantifies some of these parameters of interest for an atomic uranium vapor process. The viability of the particular LIS process chosen is based on the employment of pulsed lasers. Under such conditions the upper limit on the pulse width is roughly set by the lifetimes of the levels which are involved in the excitation processes.

It is important to note that with the possible exception of the rare gas halogen lasers, the probability of using fixed wavelength lasers for selective excitations is poor. One then turns to the use of tunable dye lasers. One dye laser option centers around flashlamp-pumped lasers. While the efficiency projections, particularly for large flashlamp-pumped devices are acceptable, the prognosis for simultaneous high repetition rate operation (> 1 kHz) with acceptable efficiency without multiplexing is questionable. In addition the flashlamp lifetime and photochemical degradation of the dye media are also areas of concern. For these and other reasons the use of coherent pumping of dye media appears more viable.

Table I. Desired Laser Requirements for a Multiphoton Atomic Uranium Vapor LIS Process

wavelength	visible
average power	> 100 W
laser efficiency	> 0.1%
pulse width	$\lesssim$ 100 ns
repetition rate	> 10^4 Hz
bandwidth	~ 1 GHz
beam quality	diffraction limited
lifetime	> 4000 hrs

Table II summarizes the lasers that were considered as coherent pumps for dye lasers. Of those listed only two were identified as having the projected characteristics which lay within the acceptable parameter space; the copper vapor and the frequency double Nd:YAG laser systems. One can of course envision using a rare gas halogen laser to pump the dye media; however, at this time we feel that the major utility of these systems lie in their intrinsic tunability thus obviating the need of the dye laser entirely. In the next sections we discuss some aspects of the copper vapor and rare gas halogen laser systems.

Table II. Summary of Potential Characteristics of Coherent Pumps for Dye Lasers.

Laser	Max. Eff.	AV P, W	Rep. Rate
Nitrogen glow discharge	6×10^{-4}	~100	7×10^3
Ar N_2 (coaxial E-beam)	1×10^{-2}	?	5×10^2
Ar ion (all visible lines)	1×10^{-3}	200	DC
Ar ion (all uv lines)	2.5×10^{-4}	50	DC
Copper vapor	2×10^{-2}	1000	$>2 \times 10^4$
Nd:YAG (cw pump, rep Q switch)	2×10^{-2}	>100	$>1 \times 10^4$
Nd:YAG (frequency doubled)	5×10^{-3}	>25	$>1 \times 10^4$

†Separative work is a value balance between feed, product, and tails. The definition of separative work states that it is a measure of the net increase or change in value for a given feed amount separated into product or tails. Separative work is commonly measured in kilograms of uranium and termed separative work units, or SWU. As an example, a product output of 1 kg of uranium at 3.2% assay would require 5.871 kg of uranium feed at normal assay (0.71%) and would involve performing 4.746 kg of separative work. Of course, to achieve the product output for this example, the tails or waste stream produced would be 4.871 kg of uranium at 0.2% ^{235}U content. See ref. 1.

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The Copper Vapor Laser

Laser action in copper metal vapor discharges has been studied by many investigators.[2-6] As can be seen from the energy level diagram in Fig. 1, this laser is basically a three level system. The ^2P upper laser levels are strongly connected to the $^2S_{1/2}$ ground state by resonantly radiative transitions at 3258 and 3247 Å. The untrapped lifetimes for these transitions is ~ 10 ns. The ^2D lower laser levels are metastable states and are not connected optically to the ground state. It is interesting to note that the actual laser transitions at 5105 and 5782 Å constitutes a double electron jump 4p to 4s ($\Delta\ell$ = 1) and 3d to 4s ($\Delta\ell$ = 2) with natural radiative lifetimes of 780 ns and 370 ns respectively.

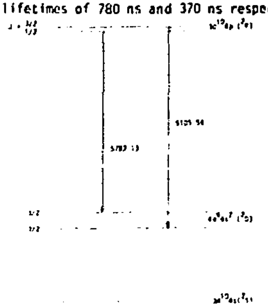

FIG. 1. Energy levels for the lasing transitions in copper. The isotope shifts for copper and hyperfine components are not shown.

Generally speaking this system exhibits the potential for efficient production of transient inversions in high voltage discharges; the intrinsic quantum efficiency being of the order of 60%. The actual overall device efficiency is considerably lower than this due to several basic kinetic processes which are operative. For example, since the peak value of the electron impact cross section for pumping the lower state (i.e., the $^2S \rightarrow {}^2D$ cross section) is only about a factor of three less than that for pumping the upper ^2P state, a reasonable amount of energy is expended in populating the lower laser level. This loss process is particularly detrimental in that it also markedly reduces the net gain in the system. There have been several kinetic models developed for the copper vapor laser. In his model Leonard[4] showed computationally that the upper level pumping rate dominated the others for values of the electron temperature in excess of 2 eV. This indicates that large E/p values, where E is the applied electric field and p the reduced pressure, are necessary for reasonable inversion densities.

While many different discharge device configurations[7-10] and buffer gas additive studies[11] have been done, the configuration that is presently employed for the LLL isotope separation experiments is a General Electric design.[6,12,13] This laser consists of an alumina tube with concentric electrodes mounted at each end. The entire assembly is mounted in a thermally insulated enclosure and the device electrically isolated using a metal-ceramic seal. After placing metallic copper at several locations within the tube bore and filling with 10-30 Torr of buffer gas (He, A or He) several kilowatts (average) of pulsed electrical power is applied to the device. The "waste" energy from the discharge heats the device to operating temperature (~ 1600° C) and pulsed outputs of several watts (average power) have been obtained.

Table III summarized the results of the copper vapor laser characteristics for such a device. The average power and efficiency of this device scale with increasing electrical input power and pulse repetition frequency. Development of plasma tubes and thyratrons capable of handling higher average power should lead to higher power outputs.

Table III. Optimized Characteristics for a General Electric Copper Metal Vapor Laser.

	Operating characteristics as a function of input power and repetition rate		
	1300 W 6.1 kHz	1326 W 11.0 kHz	1450 W 6.1 kHz
Average output power (W)	1.75	2.30	2.45
at 5106 Å	1.25	1.70	1.70
Single pulse energy (μJ)	287	209	400
at 5106 Å	205	155	279
Pulse width FWHM (ns)	27	42	27
Average peak power (kW)	10.6	5.0	14.8
at 5106 Å	7.6	3.7	10.3
Specific energy (J/cm^3)	13	9.5	18
Efficiency (d.c. input to total laser output)(%)	0.135	0.17	0.17
Tube lifetime (hr)	7	7	
Far field divergence (mrad)	2.4†		

†This corresponds to 35 times the diffraction limit for a gaussian beam.

The potential device limitations imposed due to the required high temperature operation has stimulated an effort to find another source for the copper atoms. Liu et. al., have obtained super-radiant emission at 5106, 5700 and 5782 Å[14] in pulsed copper iodide discharges. Here the source for the copper atoms is obtained by the dissocation of the CuI molecule. Other copper halide compounds can also be employed.[15] Their major advantage appears to be the lower operating temperature. For example at 1600° C the vapor pressure of pure copper is about 1 Torr whereas at ~ 600° C the vapor pressure of CuI is 1-10 Torr. The alteration of the plasma and kinetic processes because of the presence of the halogen can be either a bonus or a detriment.[16] Work is presently underway to clarify this point.

It is interesting to note that despite the early discovery of the copper vapor laser, both a detailed physical understanding of the device characteristics and a quantitative data base of laser parameters is still lacking. This situation is due in part to the lack of basic kinetic data for copper, in part to experimental difficulties associated with working at very high temperatures and, in part due to the limited interest in the low overall system efficiencies which were achievable in the past with externally-heated metal vapor generator techniques. The demonstration of a 15 watt,[b] 1% efficient pure metal vapor laser, which utilized the waste heat from the electrical discharge to generate the vapor, put this system in an entirely new perspective. Table IV illustrates a comparison between various published copper vapor laser characteristics.

Table IV. Comparison of Demonstrated Copper Vapor Laser Characteristics

	Type	Tube Dia. (cm)	Tube Length (cm)	Average Power (W)	Pulsing Frequency (kHz)	Efficiency (%)	Energy Density (μJ/cm³)
Isaev[6]	Cu metal	0.8	70	6	18	0.35	9.5
		1.5	70	15	18	1.0	6.7
G. E.[13]	Cu metal	0.85	8.5	1.3	6.8	--	39
LLL	Cu metal	0.8	44	2.5	6.1	0.17	18
Walter[17]	Cu metal	2	80	11	15	1.8[a]	29
JPL[18]	Cu chloride	1	30		20	1	35

[a]Cu metal generated by external heat source.

Unfortunately until more precise knowledge of the basic kinetics of copper lasers is obtained, one is limited to the development of scaling relationships on the basis of experimentally determined output power for specific experimental situations. The energy density limitation of copper vapor is not known or well understood at the present time. The best figures reported to date are approximately 15-40 μJ/cm³ for both pure metal and halide devices. Evidence points to possible metastable quenching effects at the tube wall as a dominant scaling limit. If this fact is borne out by further investigations, then scaling of the conventional copper vapor laser can be accomplished by scaling the tube length. Under these severe conditions while 100 watt lasers seem eminently achievable, power levels in the multihundred watt to kilowatt regimes may not be realistically feasible.

The Dye Laser

The wavelengths of the pulsed copper vapor laser do not overlap any useful bound-bound transitions in uranium; one must turn therefore to employing dye media and associated hardware to shift the laser frequency into a useful spectral region. Depending upon the specific process used the wavelength region of interest for tunable dye lasers in a uranium multiphoton photoionization LIS process spans the spectral region from approximately 5600 Å to 6600 Å. While the wavelength region centered near 6000 Å is adequately covered by the rhodamine family, the region out near 6500 Å has traditionally lacked dyes with good lasing properties which can be pumped with the green copper vapor or argon ion lines.

A survey of dyes and solvents was performed at LLL by P. Hammond using the copper vapor laser operating on the 5106 and 5782 Å band. Figure 2 summarizes the best systems combinations compatible with the copper vapor laser pump. Of special note is the availability now of the new class of DCM dyes which appear to be stable and efficient lasing sources out near 6500 Å.

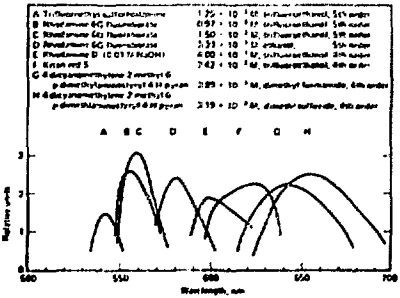

FIG. 2. Optimized dye media for use with the copper vapor laser.

High power dye lasers for LIS applications will, in general, be configured as oscillator-amplifier systems in which the oscillator provides the necessary precise frequency-bandwidth characteristics which is then boosted to the desired final power by successive amplifier stages. Among the several dye laser parameters required for LIS, the one characteristic which is unique to the uranium metal vapor process is the requirement of a laser line-width power density distribution which will effect the uniform saturation of the inhomogeneously-broadened uranium absorption line across its nominal ~ 1 GHz (fwhm) Doppler width. This requirement is dictated by the need to achieve efficient atom utilization.

At LLL, we have developed a copper vapor pumped linear dye amplifier which exhibits a 20-30% conversion efficiency. Using known staging techniques such as a device appears to be scalable up to the desired power levels for LIS applications. However, at the present time we have not yet developed an oscillator which has the desired temporal and bandwidth format; work is continuing in this particular area.

The Rare Gas Halogen Laser

Quite recently considerable work is being devoted at LLL and elsewhere, to discharge excitation of several of the rare gas halides. These systems hold the promise at least, of meeting both the repetition rate and scalable power requirements of an atomic uranium vapor LIS process. In addition they are intrinsically tunable over a wide frequency range thus abrogating the need for using the dye laser. Before proceeding further it is useful to discuss the major kinetic processes which are responsible for the population in the upper radiative state in these systems. However, one must bear in mind that at this time there does not exist a great deal of data and analysis so that what is presented here is a best estimate of the important processes. We shall concentrate most of our discussion on the XeF rare gas halogen system, the electronic structure of which is shown in Fig. 3.

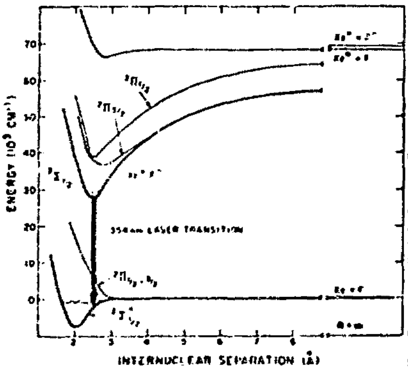

Fig. 3. Estimated potential energy curves for xenon fluoride. From Ref. 19.

The excited states of the rare gas halides are predominately ionic in character and show a strong similarity to the excited states of the alkali halides.[20] The lowest lying excited states can be correlated at infinite internuclear separation, with $Xe^+ + F^-$. As these particles approach one another they attract along a very long range coulomb curve which crosses in a diabatic sense, the excited states of both Xe and F. These curve crossings then provide for an efficient collisional channeling of energy residing in the excited state manifolds, into the lowest-lying molecular excited states (namely the $^2\Sigma^+_{1/2}$ state). The ground states are split into Σ and π branches; the $^2\pi_{1/2,3/2}$ is repulsive in character (unbound) whereas the $^2\Sigma_{1/2}$ exhibits a binding, the magnitude of which depends upon the species under consideration. For example, the $^2\Sigma_{1/2}$ ground state is unbound or only slightly bound in XeI but more strongly bound in XeF. The radiation emitted, therefore, may have a bound-bound as well as bound-free spectral character.

At this point it is useful to discuss briefly the utility of the rare gas halogen systems in an LIS context. Figure 4 shows the extent of the flourescent spectra for the rare gas halogens. One sees that for a given molecular system the spectra is comprised of at least one narrow bandwidth emission corresponding to the $^2\Sigma_{1/2} \rightarrow {}^2\Sigma_{1/2}$ transition and a broadband emission which corresponds to the $^2\Sigma_{1/2} \rightarrow {}^2\pi_{1/2,3/2}$ transitions. In actual fact these spectra are somewhat more complicated than what is shown due to the fact that at infinite internuclear separation the excited states are correlated with both the $J = 3/2$ and $1/2$ rare gas ionic ground states.[21] The useful spectral range spanned by these molecules is from roughly 1900 to 5500 Å. As can be seen by the vertical lines in this figure a two step atomic uranium vapor LIS process that utilizes an efficient laser for the last photoionization step, will require a rare gas halide laser which lies in the wavelength range from 2470 to 4000 Å. Because of this fact work at LLL has centered around the use of XeF or XeCl as one photon of a two photon LIS process.

The mechanisms by which excited states of the rare-gas halides and dihalogens are efficiently formed by electron beam and discharge excitation has been a matter of intense speculation. It is known[22] that rare-gas metastable attack on some halogen-containing molecules, such as the reactions

$$Xe^* + F_2 \rightarrow XeF^* + F \tag{3}$$

$$Xe^* + NF_3 \rightarrow XeF^* + NF_2 \tag{4}$$

proceeds rapidly with high yield of excited rare-gas halides. The production mechanism in the discharge lasers presumably involves production of these metastables by discharge electrons and their subsequent collision with F_2 or NF_3. In electron beam excited systems, which typically contain about 5 atm Ar, 150 Torr Xe, and 5 Torr F_2 or NF_3, the production mechanism may involve rare-gas excimer chain to produce Ar_2^*, followed by Ar_2^* collisions with Xe to produce Xe^* and subsequent attack on NF_3. Some ArF^* emission is seen from such systems, however, so the kinetic chain might involve the exchange

$$ArF^* + Xe \rightarrow Ar + XeF^* \tag{5}$$

with ArF^* produced by argon atomic metastable or excimer collisions. It has also been speculated that ionic reactions such as

$$Xe^+ + F^- + M \rightarrow XeF^* + M \tag{6}$$

where M represents a heavy body (xenon or argon) may be important.

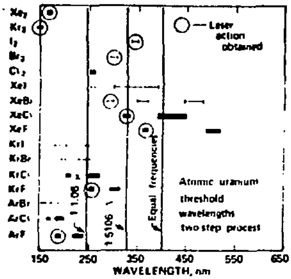

FIG. 4. Fluorscence spectra of the rare gas excimers and rare gas halides. The circles denote those systems and transitions for which stimulated emission has been observed. Shown also are the requisite wavelengths for a hypothetical two step LIS process.

Several important collisional loss processes for Xe^* are worth nothing. These are given below:

$$Xe^* + Xe^* \rightarrow Xe^+ + Xe + e \qquad (7)$$

and

$$e + Xe^* \rightarrow Xe^+ + 2e \qquad (8)$$

The first of these (namely Eq. 7) is a Penning ionization process and cross sections $\sim 10^{-14}$ cm^2 are probable. The second loss process (Eq. 8) involves the ionization (or excitation) of the excited state due to interactions with the cold background plasma. This is a well known phenomenon in gaseous discharges and cross-sections as large as $\sim 10^{-15}$ cm^2 are possible. This latter reaction is of utmost importance in determining the performance characteristics of electron beam sustainers or simple electrical discharge excitation schemes. It has been shown both experimentally and theoretically to substantially limit the effective use of a sustainer discharge to augment the e-beam pumped xenon excimer system. Roughly speaking, these two reactions indicate that to produce Xe^* at high efficiencies, $Xe^* \lesssim 10^{15}$ cm^{-3}. These processes will be important for XeF^* as well and with probably similarly large cross section values.

There is an additional collisional loss process which appears to be energetically possible; namely the collisional deactivation of XeF^* by fluorine. In their work on XeF^* Ewing and Brau[23] postulated the following deactivation scheme, namely

$$XeF^* + F_2 \rightarrow XeF + F_2^+. \qquad (9)$$

From their data they infer a rate coefficient of approximately 8×10^{-10} cm^3 s^{-1}. Since this process effectively competes with the radiative channel one must work at a sufficiently low halogen concentration that this reaction does not dominate the stimulated emission rate.

Obviously these systems are quite complicated and certainly warrant further study. Let us estimate however, the maximum intrinsic efficiency for the production of XeF^* under the assumption that the experimental conditions are chosen so as to minimize the collisional deactivation processes. From Eq. 3 we have that,

$$\eta_{XeF^*} = \eta_{Xe^*} \frac{E_{XeF^*}}{E_{Xe^*}} \qquad (10)$$

where η and E are the appropriate production efficiencies and photon (or state) energies. Theoretically η_{Xe^*} can be shown to have a value $\sim 50\%$ if the excited state densities are held below approximately 10^{15} cm^3. This implies a maximum intrinsic efficiency of $\sim 20\%$.

At the present time it appears that because of the repetition requirements ($> 10^4$ Hz), electron beam or e-beam/sustainer excitation of the rare gas halides is not a viable option and for this reason work on discharge excitation for these systems has been encouraged. Quite recently R. Airey[24] has shown that the strong band laser emission in XeF is tunable over approximately 20 Å in an unoptimized cavity configuration. Table V summarizes our best understanding at this time, of the published operating characteristics for discharge excited XeF and KrF.

Table V. Comparison of Demonstrated Discharge Excited Rare Gas Halogen Characteristics.

Molecule	Tube Length (cm)	Pulse Energy (mJ)	Pulse Frequency (Hz)	Efficiency
XeF[25]	50	10	5[27]	0.5%
XeF[26,28]	90	100	-	1%
KrF[25]	50	1.6	5[27]	0.06%
KrF[26,28]	90	30	-	0.3%

Conclusion

A brief summary of the laser requirements for an atomic uranium vapor LIS process have been presented. The near term use of copper vapor pumped dye lasers has been described and both the present and future status of these devices given. At this time it appears that they can meet the needs of an LIS process however, it remains to be seen if both the scaling and long lifetime (> 4000 hrs) requirements can be fulfilled.

The use of high repetition rate electrical discharge excitation of the rare gas halogens appears to be quite attractive in an LIS context. Much more work is required to bring these to the point where a critical assessment of their capabilities can be made.

Acknowledgements

Much of this material has been obtained from the 1976 LLL annual report for laser isotope separation;[29] we greatfully acknowledge the contributions of T. Kan, S. Hargrove, J. Holtz, M. Spaeth, P. Hammond and R. Davis.

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