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STUDIES ON PRODUCTION OF METASTABLE CORE-EXCITED ATOMS

BY LASER-PRODUCED X-RAYS

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The overall objective of the work on this program was to study methods for production of core-excited metastable atoms by laser-generated x-rays. We are interested in the spectroscopy of these levels, their autoionizing and radiative rates, and their metastability in the presence of hot electrons and ions. The concept of using x-rays emitted from a laser-produced plasma to excite large densities of energetic excited levels in atoms and ions has been thoroughly experimentally investigated using modest, 100 mJ, plasma-producing lasers. One of the objectives of this work was to verify that these techniques could be scaled up to higher energies, such as 20 J. Thus a major effort this year has been devoted to the design and construction of the high energy (20 J) 1064 nm plasma-forming laser system and the tunable probe/transfer laser.

The laser construction has been jointly supported by a grant from the DoD-University Research Instrumentation Program and the Office of Naval Research; thus this contract has essentially supplemented those programs in this regard. Although many of the components are commercial, this was an extensive facilities construction project with unique problems. In addition to the general design and staging of the system, we have been particularly concerned with: establishing precise synchronization of the two laser systems (dye and 1064 nm); providing accurate timing for the various components; obtaining the required 1 ns length pulse for the 1064 nm system; reliably selecting a single pulse; the development of a mode-locked and Q-switched dye laser oscillator; and creating the required environment for the reliable operation of such a system. A paper describing the laser system is currently in preparation.

A second objective of the program was to use the laser system to investigate the the excitation of Li ion states, measuring populations both in

the metastable $1s2s$ ion level previously studied at low energies, and in the $1s3s$ radiating level that we have proposed as the upper level of an VUV laser system at 165.3 nm. Our initial results indicate that the metastable population is scaling with energy as expected, but that the laser level population is significantly below predictions.

Our work in both these areas was presented at the OSA Third Topical Meeting on Short Wavelength Coherent Radiation: Generation and Applications, Monterey, CA (March 24-26, 1986). This paper is attached as Appendix A.

PUBLICATIONS

1. S. E. Harris, J. F. Young, R. G. Caro, R. W. Falcone, D. E. Holmgren, D. J. Walker, J. C. Wang, Joshua E. Rothenberg, and J. R. Willison, "Laser Techniques for Extreme Ultraviolet Spectroscopy," in Laser Spectroscopy IV, H. P. Weber and W. Luthy, eds. (New York: Springer-Verlag, 1983), pp. 376-381.
2. R. G. Caro, J. C. Wang, J. F. Young, and S. E. Harris, "The Excitation of Metastable Extreme Ultraviolet Levels," in Laser Techniques in the Extreme Ultraviolet, S. E. Harris and T. B. Lucatorto, eds. (New York: AIP, 1984), pp. 417-426.
3. P. J. K. Wisoff and R. G. Caro, "A Superheated Na Cell for X-Ray Photoionization Experiments," Appl. Phys. B **35**, 65-69 (1984).
4. R. G. Caro, J. C. Wang, J. F. Young, and S. E. Harris, "X-Ray Excitation of Energetic Metastable Levels in Atoms and Ions," Opt. News **11**, 9-11 (March 1985).
5. S. E. Harris, R. G. Caro, R. W. Falcone, D. E. Holmgren, J. E. Rothenberg, D. J. Walker, J. C. Wang, J. R. Willison, and J. F. Young, "Metastability in the XUV: Lasers and Spectroscopy," in Atomic Physics 9, R. S. Van Dyck, Jr. and E. N. Fortson, eds. (Singapore: World Scientific Publishing Co., 1985), pp. 462-479.
6. R. G. Caro and J. C. Wang, "Population Distributions in Li Vapor by a Photoionization Electron Source," Phys. Rev. A (to be published).

7. D. J. Walker, R. G. Caro, and S. E. Harris, "Proposal for an Extreme Ultraviolet Auger Laser at 63.8 nm in Cs III" J. Opt. Soc. Am. B (submitted for publication).
8. R. G. Caro, P. J. K. Wisoff, G. Y. Yin, D. J. Walker, M. H. Sher, C. P. J. Barty, J. F. Young, and S. E. Harris, "Soft X-Ray Pumping of Inner-Shell Excited Levels for Extreme Ultraviolet Lasers," in Short Wavelength Coherent Radiation: Generation and Applications, D. T. Attwood and J. Bokor, eds. (New York: AIP) (to be published).
9. J. F. Young, J. J. Macklin, and S. E. Harris, "Ellipsoidal Focusing of Soft X-Rays for Longitudinally Pumping Short Wavelength Lasers," in Short Wavelength Coherent Radiation: Generation and Applications, D. T. Attwood and J. Bokor, eds. (New York: AIP) (to be published).

APPENDIX A

SOFT X-RAY PUMPING OF INNER-SHELL EXCITED LEVELS FOR EXTREME ULTRAVIOLET LASERS

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ABSTRACT

We report the construction of a facility for investigating x-ray pumping of extreme ultraviolet lasers. Measurements of large excited populations in Li^+ and an investigation of a Li^+ "shake-up" laser are described.

* * *

When a $1.06\text{ }\mu\text{m}$ laser beam is focused onto a high-Z target at intensities of $10^{12} - 10^{14}\text{ W cm}^{-2}$, a plasma is formed from which a burst of soft x-rays is emitted with a conversion efficiency in excess of 10%. The burst of x-rays can be used as an x-ray flashlamp to pump potential extreme ultraviolet (XUV) laser systems.¹⁻⁴

In this work we report the construction of a facility at Stanford University consisting of a Nd:Glass laser system capable of generating a 20 J pulse of 1 ns duration at $1.06\text{ }\mu\text{m}$. This laser is synchronized with a tunable dye laser which can produce pulses with energies as high as 1 mJ, a duration of approximately 20 ps, and which are tunable from 400 nm to 900 nm. This facility is well suited to the experimental investigation of a variety of x-ray flashlamp-excited XUV laser systems. In particular, it is ideal for the study of XUV lasers produced by the initial excitation of a highly energetic metastable level (by the x-ray flashlamp), followed by rapid transfer of population to a nearby radiating level by means of a tunable dye laser.

A schematic of this system is illustrated in Fig. 1. The heart of the system consists of two mode-locked and Q-switched Quantronix 416 Nd:YAG lasers for which the acousto-optic mode-locker units are driven by a common rf signal. In this way synchronization of the output pulse trains from the two lasers is obtained such that the relative timing of the two pulse trains is constant within $\pm 50\text{ ps}$ over a period of 4-8 hours. By inserting an etalon in the cavity of one of these lasers, a train of 1 ns pulses is produced. A single one of these pulses is selected by a Pockels cell pulse selector placed external to the cavity, and that pulse is amplified through a series of Nd:YAG and Nd:Glass amplifiers until a pulse of 20 J energy is obtained from the final 33 mm diameter Nd:Glass rod. The configuration of this part of the system is shown in Fig. 2.

Fig. 1. Schematic of Stanford laser facility.

The output from the second mode-locked and Q-switched Nd:YAG laser is converted to the second, or third, harmonic at 532 nm or 355 nm, and used to synchronously pump a dye laser oscillator,⁵ as shown in Fig. 3. This oscillator can produce easily tunable output pulses with 20 ps duration and nearly transform limited linewidth in the spectral region from 400 nm - 900 nm. Output energies range from 20 μ J at 560 nm when pumping with 532 nm light, to 2 μ J at 400 nm when pumping with the lower energy third harmonic of the Nd:YAG laser. The output from this dye oscillator is then amplified in a chain of three dye amplifiers, each pumped longitudinally by a pulse of 1 ns duration at 532 nm or 355 nm. This pump pulse is derived from the 1 ns, 1.06 μ m pulse at an early stage before amplification in the Nd:Glass amplifiers. It is then passed through a further three Nd:YAG amplifiers to produce a 200 mJ beam at 1.06 μ m that can be converted to the second or third harmonic for pumping the dye amplifier system. A schematic illustration of this dye amplifier system is shown in Fig. 4.

The experiments that have been performed to date with this laser facility have had two objectives. The concept of using x-rays emitted from a laser-produced plasma to excite large densities of energetic excited levels in atoms and ions has been thoroughly experimentally investigated¹⁻³ using modest, 100 mJ, plasma-producing laser energies. It was necessary, however, to verify that these techniques could be scaled up to the 20 J energies available from the current facility. In order to compare with the original work of Caro, et al.,¹⁻² experiments have been performed to investigate the excitation of Li^+ (1s2s) ions at 60 eV as a result of photoionization by laser-produced x-rays.

SCHEMATIC OF LASER SYSTEM

Fig. 2. Schematic of 1.06 μm plasma-producing laser system. HWP, QWP = half wave plate and quarter wave plate.

In addition, the Li^+ vacuum ultraviolet laser system at 165.3 nm, recently proposed by Harris and Caro,⁶ has been experimentally investigated. The energy level diagram of this system is shown in Fig. 5. When a (1s) electron is removed from a Li atom by x-ray photoionization, the principal product is the Li^+ (1s2s) ion. However, due to the process of "shake-up," there is a significant probability that the Li^+ (1s3s) ion will be produced.⁷⁻⁹ Indeed, experiments indicate⁹ that 15% of the ions created by (1s) removal should be in the level Li^+ (1s3s)³S₁. In contrast, only 7% should be found in the Li^+ (1s2p)³P levels and so an inversion should be produced on the [(1s3s)³S - (1s2p)³P] transition at 165.3 nm.

Fig. 3. Schematic of dye laser oscillator showing second and third harmonic generation (SHG, THG); pump beam focusing lens (L1); Littrow grating (G); prism beam expander (PBE); Glan-Taylor prism polarizer (GT); Pockels cell (PC); 15 cm radius mirror (M); intra-cavity 7.5 cm focal length lens (L2); dye cell (DC); high reflector (HR).

Fig. 4. Schematic of dye amplifier system.

As shown in Fig. 6 and discussed in Ref. 6, modeling of the photoionization process, as well as of a number of competing processes that occur in the photoionized plasma, indicates that gains in excess of $\exp(10)$ should be observable at 165.3 nm when an experiment with the geometry shown in Fig. 7 is performed and a 10 J plasma-producing laser is used. As can be seen in Fig. 7, the

Li⁺ SHAKE-UP LASER

Fig. 5. Li shake-up laser energy levels.

plasma-producing laser beam is split into three beams, each of which is focused onto a separate tantalum target at an intensity of the order of $10^{12} \text{ W cm}^{-2}$. The plasmas that are produced emit soft x-rays that propagate out into the surrounding Li vapor (10^{17} cm^{-3}) and photolionize the atoms to produce Li⁺ (1s2s) and Li⁺ (1s3s) ions in large quantities.

Although relative magnitudes of the populations in the various Li⁺ levels can be deduced by observations of emission, quantitative measurements can more easily be made by means of absorption spectroscopy. In this technique, the wavelength of the beam from the picosecond dye laser is scanned through a transition which has as its lower level, that in which population is to be measured. By analyzing the absorption profile obtained, a measure of the excited level population-length product, N^*L , can be deduced.¹⁻² By altering the delay between the dye laser probe beam and the plasma-producing 1.06 μm beam, the temporal behavior of the population can be examined.

In Figs. 8 and 9 are shown the dependence of the N^*L product of the Li⁺ (1s2s)3S ion as a function of time (Fig. 8) and distance from the Ta targets (Fig. 9). For comparison, the predictions of a theoretical model¹⁰ are shown as line A in Figs. 8 and 9. It can be seen that the functional dependence of the data is in good agreement with the theoretical predictions, while the absolute magnitude is low by a factor of approximately 2-3.

Fig. 6. Model predictions of electron density n_e , effective number density-length product, N^*L , and gain, $N^*\sigma_L^e$ at 165.3 nm as a function of distance from the target. The model assumes three target spots, each irradiated by a 1 ns, 2 J, 1.06 μm laser pulse.

Fig. 7. Schematic of experimental geometry.

Fig. 8. $N^*L [Li^+ (1s2s)^3S]$ as a function of time with respect to the peak of the $1.06 \mu m$ laser pulse. Distance from targets = 7 mm; Li density = $10^{17} cm^{-3}$; energy incident on each of three targets = 2.3 J.

Fig. 9. $N^*L [Li^+(1s2s)^3S]$ as a function of distance from the target at a time 1.9 ns after the peak of the $1.06 \mu m$ pulse. Li density = $10^{17} cm^{-3}$; energy incident = 2.3 J on each of three targets.

To obtain these data points the absorption on the $\text{Li}^+ [(1s2s)^3S - (1s2p)^3P]$ transition at 548.5 nm was monitored. In a similar fashion, by measuring the absorption on the $\text{Li}^+ [(1s3s)^3S - (1s4p)^3P]$ transition at 368.4 nm, the $\text{Li}^+ (1s3s)^3S$ population can be monitored. In this case, the absorption is in a regime that allows the simultaneous deduction of both N^*L and the Lorentz width, $\delta\omega_L$, of the transition. The predominant mechanism contributing to this Lorentz width is Stark broadening and, from Griem,¹¹ the Stark broadening coefficient is approximately $\delta\omega_L = 2.6 \text{ cm}^{-1}$ at an electron density of 10^{16} cm^{-3} and a temperature of 0.5 eV. Using this data, the electron density dependence on time at a distance of 7 mm from the target has been determined (Fig. 10). Again, the theoretical predictions, curve A, are in good agreement with the measured values. The dependence of electron density on distance from the target is shown in Fig. 11. Here the agreement with theory is only fair, and the model predictions are poor in close to the target where pumping rates are high. It has been reported elsewhere¹⁰ that this simple model is no longer correct under conditions where predicted electron densities exceed 10^{16} cm^{-3} .

Fig. 10. Electron density as a function of time after the peak of the plasma-producing pulse. Measurements were at 7 mm distance from the targets with 2.7 J incident on each of three targets; Li density = 10^{17} cm^{-3} .

In contrast, in Figs. 12 and 13 are shown the dependence of the $\text{Li}^+ (1s3s)^3S$ number density-length product, N^*L , on time, and distance from the target. In these cases, the measured values are lower than those predicted from Fig. 6 by a factor of approximately 30. The gains depicted in Fig. 6 are small signal gains in the absence of amplified stimulated emission (ASE) at 165.3 nm. If the populations of Fig. 6 were being realized, ASE would act to

Fig. 11. Electron density as a function of distance from the three targets at a time 1.4 ns after the peak of the 1.06 μm pulse. Li density = 10^{17} cm^{-3} ; energy per target = 2.3 J.

Fig. 12. $N^*L [\text{Li}^+ (1s3s)^3S]$ as a function of time after the peak of the 1.06 μm pulse. Li density = 10^{17} cm^{-3} ; distance from targets = 7 mm; energy incident on each of three targets = 2.5 J.

Fig. 13. $N^*L [Li^+ (1s3s)3S]$ as a function of distance from the three targets at 1.4 ns after the peak of the 1.06 μm pulse. Li density = 10^{17} cm^{-3} ; energy per target = 2.3 J.

equilibrate the populations in the $Li^+ (1s3s)$ and $Li^+ (1s2p)$ levels.¹² To estimate the effect of this, curves A in Figs. 12 and 13 represent approximate theoretical predictions of the N^*L values for the $Li^+ (1s3s)3S$ level under circumstances in which it is in equilibrium with the $Li^+ (1s2p)3P$ level (as a result of ASE). Even under these conditions, there remains a discrepancy of approximately an order of magnitude between the measured values and the predicted values represented by curve A. Furthermore, in these particular experiments there was no sign of especially intense emission from the $Li^+ (1s3s)3S$ level - suggesting that ASE is probably not an important process here. This leads us to the conclusion that the $Li^+ (1s3s)3S$ population is anomalously low.

As well as the discrepancy between the predicted and measured values of the absolute magnitude of the $Li^+ (1s3s)3S$ population, the theoretical curve in Fig. 13 has a quite different functional dependence on distance than does the data. For comparison, in the analogous curve of Fig. 9, the $Li^+ (1s2s)3S$ population has a functional dependence on time that is well predicted by theory.

Clearly, the $Li^+ (1s3s)3S$ population is not being produced in the quantity predicted by theory. The actual reason for the low $Li^+ (1s3s)$ population is unclear at this time. Estimates of rates for the de-excitation of the level $Li^+ (1s3s)3S$ by electrons, or photons from the laser-produced plasma, yield values that are too small to explain this result. Although anomalously fast deexcitation cannot be ruled out, the decay curve of Fig. 12, showing 2-3 ns decay time for the $Li^+ (1s3s)3S$ level makes this unlikely.

Similarly, anomalously small excitation rates for the $\text{Li}^+ (1s3s)$ levels seem unlikely in the face of the observation that the $\text{Li}^+ (1s2s)$ level and electron density measurements agree reasonably well with theory, and the well documented predictions of the shake-up ratio for $\text{Li}^+ (1s3s)$ production. Further work is necessary in order to explain this disconcerting result.

Although laser action at 165.3 nm has not been demonstrated, the fundamental utility of this laser facility for creating large density-length products of excited levels has been verified in the form of the $\text{Li}^+ (1s2s)3S$ measurements reported here. In the future it is expected that this facility will be used to investigate a variety of photoionization-pumped lasers in the VUV and XUV regions of the spectrum with emphasis on the laser classes produced by shake-up⁶ and Auger¹³ processes, as well as those that make use of metastable¹⁴ and quasi-metastable¹⁵ levels in atomic and ionic systems. It seems that these techniques should enable laser action to be demonstrated at a variety of wavelengths between 10 nm and 100 nm in the future.

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REFERENCES

1. R. G. Caro, J. C. Wang, R. W. Falcone, J. F. Young, and S. E. Harris, Appl. Phys. Lett. 42, 9 (1983).
2. R. G. Caro, J. C. Wang, J. F. Young, and S. E. Harris, Phys. Rev. A 30, 1407 (1984).
3. J. C. Wang, R. G. Caro, and S. E. Harris, Phys. Rev. Lett. 51, 767 (1983).
4. W. T. Silfvast, O. R. Wood, II, and D. Y. Al-Salamah, "High-Gain VUV and Visible Inversions in He⁺ and Ar⁺ Produced by Two-Electron (Shakeup) Photoionization" (to be published).
5. P. J. K. Wisoff, R. G. Caro, and G. Mitchell, Opt. Comm. 54, 353 (1985) and "High Power Picosecond Blue Pulses from a Dye Laser Synchronously Pumped by a Q-Switched, Mode-Locked Nd:YAG Laser," Opt. Lett. (submitted for publication).
6. S. E. Harris and R. G. Caro, Opt. Lett. 11, 10 (1986).
7. F. P. Larkins, P. D. Adeney, and K. G. Dyall, J. Electron. Spectrosc. Relat. Phenom. 22, 141 (1981).
8. T. A. Ferrett, D. W. Lindle, P. A. Heimann, and D. A. Shirley, Lawrence Berkeley Laboratory, CA 94720 (personal communication).
9. P. Geard, Ph.D. Dissertation, University of Paris-Sud, Orsay, France (1984).
10. R. G. Caro and J. C. Wang, "Population Distribution in Li Vapor Excited by a Photoionization Electron Source, Phys. Rev. A, (to be published).
11. H. R. Griem, Spectral Line Broadening by Plasmas (Academic Press, New York, 1974).
12. R. W. Falcone, Physics Department, University of California, Berkeley, CA (private communication).
13. A. J. Mendelsohn and S. E. Harris, Opt. Lett. 10, 128 (1985).
14. S. E. Harris, Opt. Lett. 5, 1 (1980).
15. S. E. Harris, D. J. Walker, R. G. Caro, A. J. Mendelsohn, and R. D. Cowan, Opt. Lett. 9, 168 (1984).