X-ray Excitation of Energetic Metastable Levels in Atoms and Ions

By R.G. Caro, J.C. Wang, J.F. Young, and S.E. Harris

A fundamental problem associated with the development of XUV lasers is that, at constant oscillator strength, the radiative lifetime of an excited level in an atomic system varies as the square of the transition wavelength. For a strong transition near 200 Å, this lifetime is of the order of 10^{-11} seconds.

To overcome the difficulty of obtaining sufficient gain in this short time, it has been proposed that population first be stored in a long-lived energetic metastable level and then, by use of an intense short-pulse tunable laser, be rapidly transferred to the upper level of the lasing transition.[1,2]

This article describes an investigation of the excitation of energetic metastable levels suitable for use as storage levels in XUV laser systems. The production of large populations in excited levels with energies in the 20-100 eV region necessitates the development of excitation techniques characterized by high peak powers. The work reported here involves the use of an x-ray "flashlamp" as an excitation source.[3]

In addition, a secondary source of excitation has been developed in which the x-rays, emitted from the "flashlamp," are converted to a burst of high density energetic electrons. This device has been called a "photoionization electron source." Both of these excitation techniques have been shown to produce populations, in energetic (20-60 eV) metastable levels of atoms and ions, which are 2 to 3 orders of magnitude larger than have been achieved by alternative excitation methods.

The x-ray 'flashlamp'

When a laser beam is focused onto a solid target with an intensity of 10^{12}-10^{14} W cm^{-2}, a dense plasma is formed at the surface of the target which absorbs, and is heated by, the laser radiation. Typical electron temperatures in such a plasma range from 10-100 eV with electron densities as high as 10^{21} cm^{-3}. This hot plasma acts like a blackbody and emits radiation in the XUV and soft x-ray region of the spectrum.

For laser pulse durations in excess of 10^{-10} sec, the duration of the emitted radiation corresponds approximately to that of the laser pulse.[4] This laser-produced plasma can thus be considered as a pulsed source of soft x-rays, or an x-ray "flashlamp."

Figure 1 shows the experimental configuration that was used in this work.[5,6] A massive plane tantalum target was placed inside a heat pipe containing atomic vapor at a density of 10^{16} to 10^{18} atoms cm^{-3}. A 1.06 μm laser beam was focused onto the target by a lens with an effective f number of f/8, and a plasma was produced at the surface of the target. The emitted soft x-ray radiation propagated into the surrounding atomic vapor, causing inner-shell photoionization of the atoms and the consequent production of ions in excited states.

A key element to the success of this experiment was the simple geometry involved, in which the 1.06 μm laser beam propagated through the atomic vapor before reaching the massive target and producing a plasma. The elimination of the requirement for a target foil[1] that was to be illuminated from behind, and destroyed after a single shot, made multiple-shot data collection feasible and thus enabled the investigation described here to be carried out.

The laser used in this work produced output pulses at 1.06 μm with an energy of 100 mJ and a duration of 600 psec. The temperature of the x-ray-emitting laser-produced plasma was determined to be between 10 and 100 eV, and the conversion efficiency from laser photons to soft x-rays was in excess of 10%. This relatively high conversion efficiency is believed to be due to the large atomic weight of the target (Z=73) and the relatively low laser intensities (10^{13} W cm^{-2}) involved.[5,6]

When the inner-shell photoionization excitation technique was applied to Li vapor, Li^+ ions were produced in the excited Li^+ (1s2s) levels at energies of approximately 60 eV (Fig. 2). The metastable $Li^+(1s2s)^1S$ population density was determined by measuring the absorption of laser probe beam as a function of wavelength at the $Li^+[(1s2s)^1S - (1s2s)^1P]$ transition at 958.1 nm. The

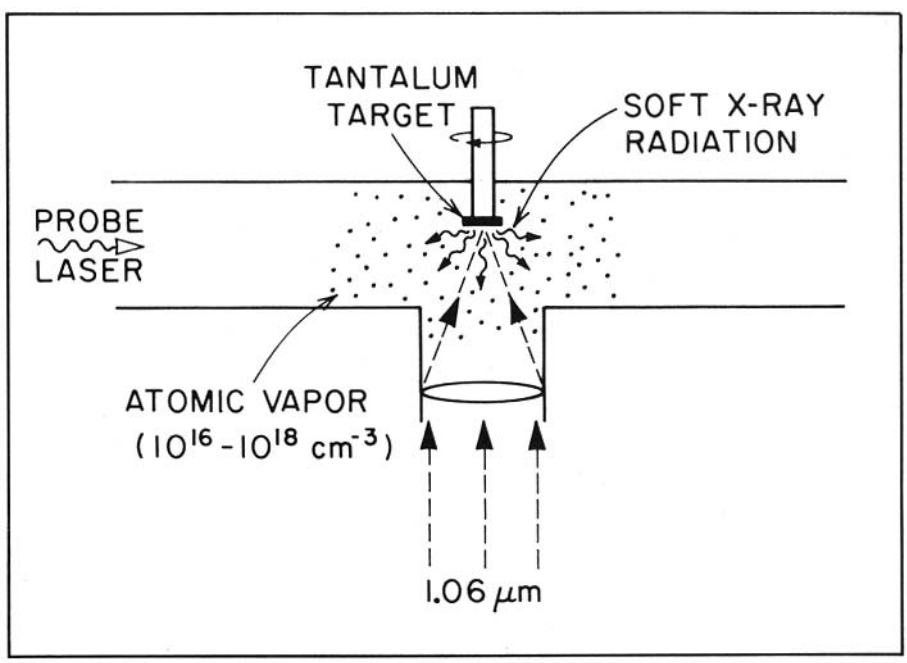

Fig. 1. Schematic of experimental configuration.

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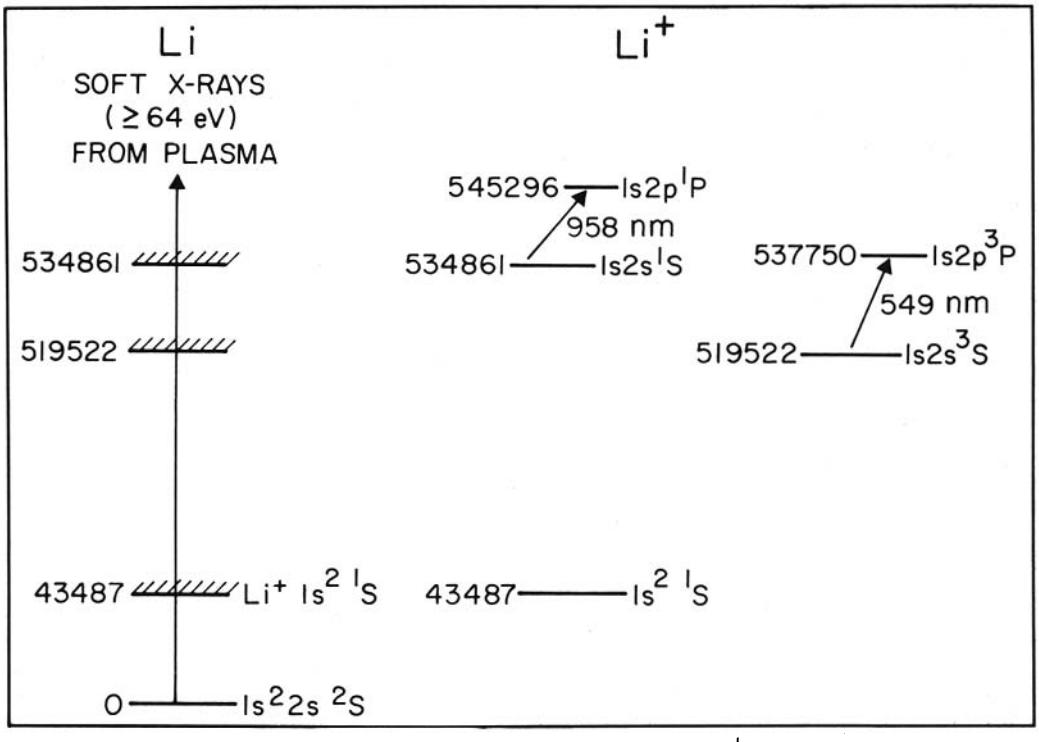

Fig. 2. Energy level diagram of Li and Li^+

absorption traces were fitted to numerically generated Voight profiles. In this manner, populations of $Li^+(1s2s)^1S$ ions as high as 3×10^{14} cm^{-3} were measured.

Suitable for storage

It was observed that these ions had an effective lifetime (probably due to electron de-excitation) of 3.6 nsec. This measured lifetime is approximately 100 times longer than the radiative lifetime (35 psec) of the $Li^+(1s2p)^1P$ level. The $Li^+(1s2s)$ level thus is sufficiently metastable, under the conditions in which it is produced in these experiments, to be suitable for use as a storage level for potential XUV laser systems.

In a similar experiment involving Na, populations of 10^{16} cm^{-3} were measured in the $Na^+(2p^53s)^3P_2$ level at 33 eV. This corresponded to a fractional excitation to that metastable level of 5%, and illustrates the effectiveness of x-ray inner-shell photoionization as a means of producing large excited level populations at high energies in ionic systems.

In an important step toward the goal of the construction of an XUV laser, Silfvast et al.[7] have recently used the x-ray photoionization technique described here to produce visible and ultraviolet laser action in Cd^+ and Zn^+. In that work, laser emission was observed from the excited level of the ion which was produced by inner-shell photoionization. Laser action was also observed as a result of the transfer of the stored population to still higher ionic levels by means of a tunable laser.

Since typical XUV gain cross sections are of the order of 10^{-14} cm^2, excited metastable populations such as those that have been produced in the Li^+ and Na^+ metastable ions, if inverted with respect to the ground level of the ion, would be sufficient to produce lasing in the XUV.

But, as noted by Mani et al.[1] and earlier by Duguay,[3] three level systems of this type are difficult to invert. The reason for this is that when an excited metastable ion is produced, a hot electron is also produced. This electron can then collide with a neutral atom to create a secondary electron and an ion in the ground state.

To avoid this effect the ambient pressure of the neutral species must be reduced until secondary electrons are not produced on the time scale of the incident laser pulse.

To circumvent this problem, four-level systems such as that shown in Fig. 3 have been proposed by Harris et al.[2,8] In these systems XUV lasing would occur from an inner-shell excited level of an atom to a lower level which was a valence level of the atom. The lower level would either be empty of could be emptied by an incident laser beam.

Although photoionization is eminently suitable for the excitation of energetic levels in **ionic** systems, the production of analogous metastable levels in **atomic** systems, such as those of Fig. 3, require an alternative excitation technique. For this purpose we have developed a photoionization electron source (PES),[9] which produces an intense subnanosecond burst of hot electrons suitable for the excitation of energetic atomic levels.

In order to produce this burst of electrons, the soft x-rays emitted from a laser-produced plasma have been used to photoionize an absorber rare gas with the consequent production of large densities of hot electrons. The high density and temperature of the electron distribution produced by the PES allow efficient excitation of both dipole-allowed and dipole-forbidden transitions to states with energies as high as 100 eV.

In addition, the short duration of the pumping pulse—determined by the pulse length of the plasma-generating laser—makes this source well suited to the study of dynamic interactions involving such states.

'Absorber' gases

Again, the experimental geometry is that of Fig. 1, but with the cell containing an absorber rare gas for the production of electrons as well as a target species to be excited by the hot photoelectron. When Ne was used as the absorber gas at a density of 3×10^{17} cm^{-3}, and a 50 mJ laser beam generated the laser-plasma x-ray flashlamp, a photoelectron energy distribution was produced[9] which had a maximum at 20 eV and a mean electron energy of 45 eV.

The total ejected electron density was 2×10^{16} cm^{-3}. The photoionization electron source described here can be considered to produce an effective current density of 3×10^5 A cm^{-2} for 600 psec.

The four-level XUV laser proposals of Harris et al.[2,8] involve the use of metastable quartet levels in the alkali atoms as long-lived storage levels. These levels are embedded in the continuum of the alkali atom but are metastable against both radiative decay and autoionization by virtue of spin conservation.

An example of such a level is the Li(1s2s2p)^{4}P^0 level at 57.4 eV (Fig. 3). Levels of this type can be effectively populated by means of the PES.

When Li was used as the target atom and mixed with the absorber gas, Ne, in the cell of Fig. 1, populations as large as 2×10^{13} cm^{-3} were excited to the Li(1s2s2p)^{4}P^0 level by the electrons of the PES. Under these excitation conditions (electron densities $> 10^{15}$ cm^{-3}) the lifetime of the Li(^{4}P^0) level is believed to be determined by the electron de-excitation rate and was measured to be approximately 3 nsec.

Similar results were obtained for the analogous Na(2p^{5}3s3p)^{4}D$_{7/2}$ level at 33 eV. The applicability of the PES to the excitation of such levels is illustrated by the observation that the population densities measured in the quartet levels in this work exceed the best results obtained by more conventional excitation techniques[10] by 2 to 3 orders of magnitude.

The large populations excited in this work, together with the reasonable observed metastability, confirm experimentally for the first time the suitability of the quartet levels in the alkali metals for use as storage levels in XUV laser systems. Although less efficient than photoionization excitation, PES excitation should allow the testing of a number of proposed XUV laser systems in atomic species.

As a useful comparison with other excitation techniques, the PES was used to excite the He(1s2s)^{1}S level. for 2×10^{18} atoms cm^{-3} of He acting as both absorber and target species, a He(1s2s)^{1}S population of 3×10^{14} cm^{-3} was measured. This corresponds to a fractional excitation of the He(1s2s)^{1}S level of 2×10^{-4} and thus also compares very favorably with that produced using more conventional excitation techniques.

In summary, the development of the laser-produced x-ray flashlamp and the photoionization electron source has enabled us to investigate excitation of, and storage in, a variety of ionic and atomic metastable levels. The scaling up of these excitation techniques, together with the spectroscopic evaluation of new XUV laser systems should, in time, allow the development of a wide variety of extreme ultraviolet and soft x-ray lasers.

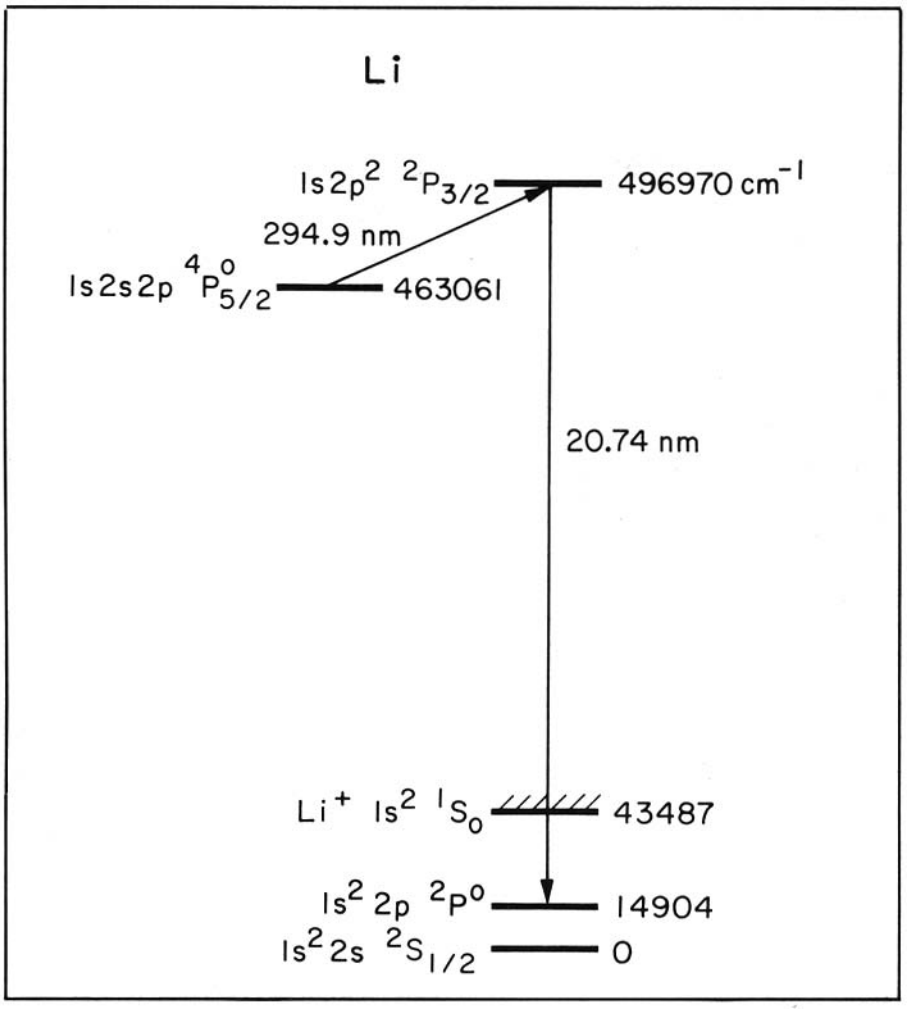

Fig. 3. Proposed XUV laser system in neutral Li.

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