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Laser Techniques for Extreme-Ultraviolet Spectroscopy

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1. Introduction

In this paper we describe several techniques for using lasers to study core-excited energy levels in the spectral region between 10 eV and 100 eV. We are particularly interested in levels that are metastable against autoionization and, in some cases, against both autoionization and radiation.

A primary motivation for our study of these levels is their potential for XUV laser systems [1,2], where energy is first stored in a metastable level and then, by using a picosecond laser pulse, is transferred to a level which radiates strongly in the XUV. The lower laser level is a valence level of the atom which is either empty or may be emptied by an incident laser beam. An energy level diagram for such a laser system is shown in Fig. 1.

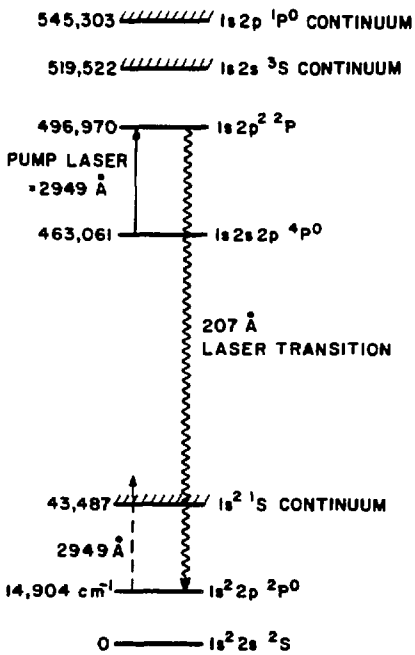

Fig. 1. Energy level diagram for the proposed 207 Å laser in neutral Li.

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2. Metastability in the Extreme Ultraviolet

A "typical" core-excited level of an alkali atom (for example, the $3p^5 4s^2\ ^2P_{1/2}$ level of K) autoionizes in about a tenth of a picosecond and has a linewidth, as observed in absorption [3], of about 63 cm^{-1}. Autoionization occurs by a 4s electron making a dipole transition to the 3p shell with the ejection of the other 4s electron into the $3p^6\varepsilon(p)$ continuum.

The classic [4] example of a very much longer-lived level is the $(1s2s2p)^4P_{5/2}$ level of neutral Li. Since the spins of the three electrons are aligned, autoionization into the $Li(1s^2)$ continuum requires a spin-spin interaction, with the result that, as an isolated atom, the lifetime of this level is 5.1 μs. Even as LS coupling breaks down, the quartet level in each configuration of highest J is necessarily a pure quartet and retains its metastability against autoionization. Also, at least in the lighter alkalis, the quartet level of highest J in each L multiplet retains its metastability. For example, the $2p^6 3s3p\ ^4S_{3/2}$ level of Na has a calculated autoionizing time of 2.4 μs.

As noted above, we are also interested in core-excited levels which have strong radiative transition strengths and which retain sufficient metastability to allow their access from energy stored in the longer-lived quartet levels. If L and S are good quantum numbers, then doublet levels in the alkalis or alkali-like ions which are of even parity and odd angular momentum, or, instead, of odd parity and even angular momentum, may not autoionize. Here, relative metastability is dependent on the extent to which LS coupling holds and to some extent on the accidental position of nearby energy levels. In Li, levels like $(1s2p^2)^2P$ are readily observed in emission [5].

It is also of importance to determine the metastability of core-excited levels when in the presence of electrons or ions. In the heavier alkalis, both fine structure changing collisions and Stark mixing with nearby levels may play a role in the quenching of these levels.

In the following sections of the paper we will discuss several laser-based techniques which allow the study of metastable XUV levels.

3. Anti-Stokes Radiation Source

The anti-Stokes radiation source [6-8] is based upon spontaneous Raman scattering of incident laser photons from excited metastable atoms. Metastable atoms may be produced in a discharge (in recent work both cw hollow cathode and high power pulsed microwave discharges have been used) and also by photoionization of ground level atoms by soft x-rays from a laser produced plasma [9].

Figure 2 shows a schematic of the pulsed high power microwave discharge version of this source. About 500 kW of peak power in a pulse 2 μs long was applied to a 90 cm long quartz tube placed in an x-band waveguide. Metastable $He(1s2s)^1S$ densities of between 10^{13} and 10^{14} atoms per cm^3 were obtained, and resulted in an anti-Stokes signal about 100 times larger than that obtained in earlier hollow cathode work. However, the increased background plasma noise now necessitated the use of a spectrometer as a blocking filter. With this arrangement the observed signal corresponded to a count rate of 20 counts per pulse or 200 counts per second.

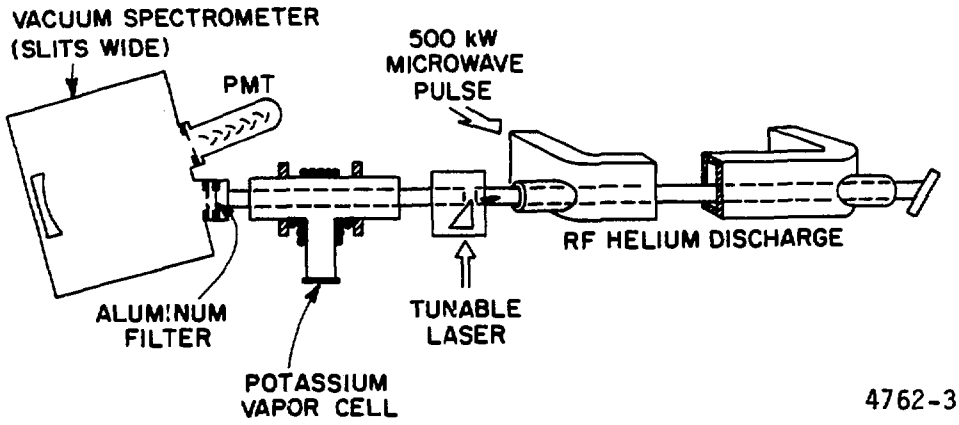

Fig. 2. Schematic of microwave pumped apparatus used for absorption spectroscopy of potassium.

The results of the study of transitions originating from the $3p^6$ shell of potassium are summarized in Tables 2 and 5 of Ref. [8]. The narrower, and therefore potentially the most interesting of the absorption lines have not been previously observed.

Figure 3 shows a schematic of a new method [9] for producing very large densities of $Li^+(1s2s)^1S$ metastables. Soft x-rays from a laser generated plasma produce metastable densities of 6×10^{14} ions/cm^3 at an incident 1.06 μm energy of 50 mJ. A tunable Li^+ anti-Stokes source based on this technique for metastable production should have a tuning range of at least $\pm$ 5000 cm^{-1} centered at 199 Å.

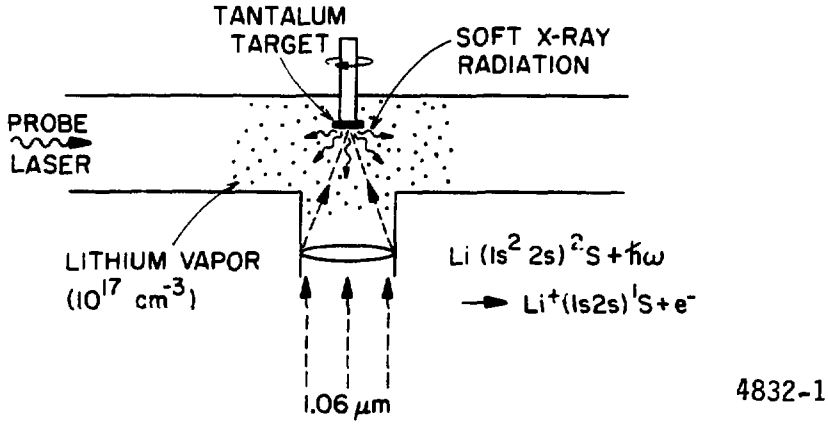

Fig. 3. Production of metastable Li atoms by photoionization.

4. Emission Spectroscopy

Figure 4 shows a schematic of the apparatus used for a different type of laser spectroscopy in the XUV. A microwave discharge is used to produce metastable $(1s2s2p)^4P$ atoms. A tunable laser causes their transfer to the $1s2p^2\ ^2P$ level, with subsequent radiation at 207 Å. Although the intercombination oscillator strength on this transition is only 2×10^{-8}, 36% of the $(1s2s2p)^4P$ population within the 0.2 mm^2 beam area is transferred with about 10 mJ of laser energy [10].

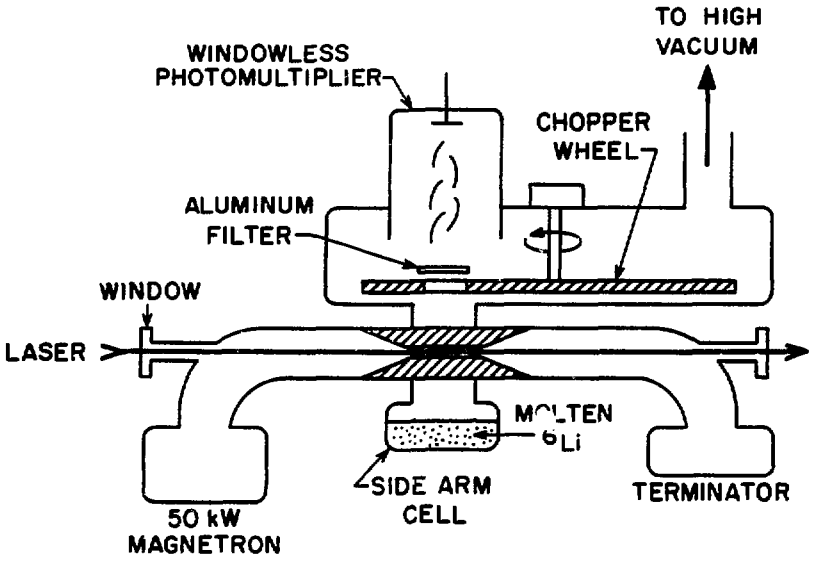

Fig. 4. Schematic of experimental apparatus for quartet-doublet transfer in neutral Li.

The advantage of this type of spectroscopy, as compared to anti-Stokes absorption spectroscopy, is that its resolution is determined by the Doppler linewidth in the visible, instead of in the XUV. In Na this should allow a resolution of about 0.1 cm^{-1}, which in turn should allow the determination of target lifetimes as long as about 50 ps.

Another technique which should also allow 0.1 cm^{-1} resolution in the XUV might be termed as depletion spectroscopy. Here one would observe a radiating line (for example, radiation from the $1s2s2p^2\ ^2P$ level in Li) and use a tunable laser to reduce this radiation as levels which autoionize are accessed.

5. Metastable Lifetime Measurement in a Plasma

In recent months, we have developed a new technique for producing large densities of $Li(1s2s2p)^4P^o$ atoms and for measuring their lifetime in the presence of large densities of electrons and ions [11].

To accomplish this an absorber gas (in this case Ne) is added to the cell shown in Fig. 3. The x-rays from the tantalum target photoionize the Ne and produce a burst of electrons with an average temperature of about 45 eV and a pulse duration equal to that of the incident laser (600 ps). By spin-exchange collisions, these electrons excite $Li(1s2s2p)^4P^o$ atoms. Under the conditions of these experiments, the cooling time of the generated electrons is $\sim$ 50 ps and therefore no quartet excitation occurs after the x-ray pulse is terminated.

The concentration of quartet atoms and their decay time in the presence of electrons and ions is measured by tuning a $\sim$ 600 ps long, variable delay, probe beam through the $Li(1s2s2p\ ^4P^o - 1s2p^2\ ^4P)$ transition at 371 nm and matching the resulting absorption traces with numerically generated Voigt profiles.

Figure 5 shows the measured density times length (N^*L) product as a function of time for the $(1s2s2p)^4P$ atoms. The peak number density of quartet atoms of $\sim 3 \times 10^{13}$ cm^{-3}, though obtained only over lengths of several mm, is about

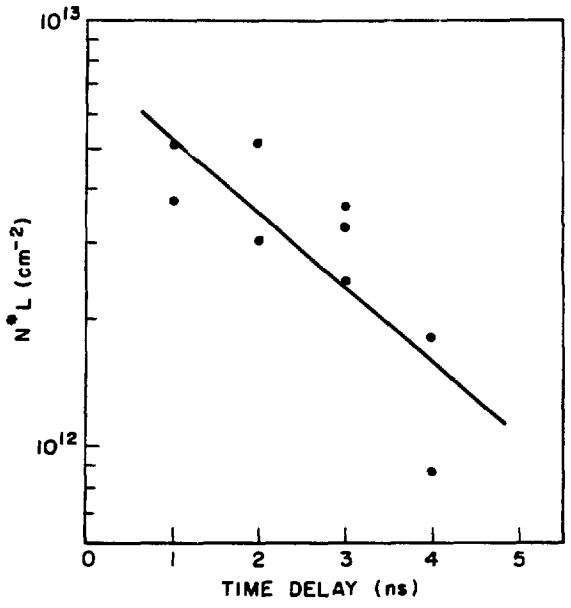

Fig. 5. Li quartet density vs. time.

three orders of magnitude larger than we have been able to obtain using pulsed hollow cathode techniques. The lifetime of 2.5 ns at an average electron density in the afterglow of $\sim 10^{15}$ electrons/cm^3 shows that the Li$(1s2s2p)^4P$ level is not anomalously susceptible to de-excitation by electrons or ions.

6. Possible Laser Systems

Although much of the above discussion has focused on Li, our primary candidate for a laser in the XUV has now shifted to Na. The properties of this system are summarized in Table 1. The reasons for the shift are several fold: (1) Since it is opaque in the XUV, Ne which was necessary to produce the quartet atoms in Li, cannot be used to construct an XUV laser. We expect that photoionization of the $2p^6$ shell of Na itself will produce the hot electrons necessary for the $2p^53s3p\ ^4S_{3/2}$ excitation. (2) This same level will be excited by an exothermic charge transfer reaction ($\Delta E = 0.14$ eV) with the Na^+ $2p^53s$ ($J = 2$) level. (3) The intercombination oscillator strength on the $2p^53s3p\ ^4S_{3/2} - 2p^53s3d\ ^2D^o_{5/2}$ level is about five orders of magnitude larger than it is in Li,

Table 1. Na Laser

Storage Level	$2p^53s3p\ ^4S_{3/2}$	$\tau_{AI} \sim 2.4$ μs	$\tau_{rad} = 0.1$ μs
Upper Laser Level	$2p^53s3d\ ^2D^o_{5/2}$	$\tau_{AI} = 45.1$ ps	
Intercombination Transition	$2p^53s3p\ ^4S_{3/2} \rightarrow 2p^53s3d\ ^2D^o_{5/2}$	$\lambda = 3326$ Å	$gf = 0.011$
Laser Transition	$2p^53s3d\ ^2D_{5/2} \rightarrow 2p^63d\ ^2D_{5/2,3/2}$	$\lambda = 375.3$ Å	$gf = 0.485$
Laser Gain Cross Section at 1.2 cm^{-1} Linewidth	$\sigma = 5.2 \times 10^{-14}$ cm^2		

thereby allowing convenient laser energies and larger volumes. (4) The lower laser level 3d is somewhat more isolated from ground than is the 2p level in Li. If necessary, other $2p^5 3snd\ ^2D^o_{5/2}$ target levels could be used.

We also note that quartet levels of the column II metals (for example, Ca and Mg) may be produced by photoionization or electron ionization of the excited lowest triplet levels of the neutral. The advantage of these systems is that the lower level of the laser transition is a level of the ion, instead of the neutral. This provides further isolation from excitation by electrons.

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