Novel Short-Pulse Photoionization Electron Source: Li ($1s2s2p$) $^4P^\circ$ Deexcitation Measurements in a Plasma

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As a result of photoionization of rare-gas atoms by x rays emitted from a laser-produced plasma, high densities ($>10^{16}$ cm^{-3}) of hot ($\sim$45-eV) electrons are produced in a pulse of $\sim$600 ps duration. Following excitation by means of this electron source, the deexcitation rate constant from the Li ($1s2s2p$) $^4P^\circ$ level is measured to be 3×10^{-7} cm^3 s^{-1} at an electron density of $\sim10^{15}$ cm^{-3}.

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For many applications, such as the study of fast chemical reactions or the investigation of kinetic processes in plasmas, it is important to produce efficient excitation of atomic and molecular species on a time scale short compared with the characteristic times of the processes of interest. One technique for achieving this end which has proved widely successful has involved the use of an intense short pulse of laser light as the source of excitation. This method, however, is unsatisfactory for investigating highly excited states with energies of 10–100 eV or for exciting optically forbidden transitions.

In this Letter we report a novel technique for generating a subnanosecond pulse of hot electrons suitable for the production of highly excited atoms and molecules. 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 this "photoionization electron source" allow efficient excitation of both dipole-allowed and dipole-forbidden transitions to states with energies as high as 100 eV. The short duration of the pumping pulse —determined by the pulse length of the plasma-generating laser[1] —makes this source well suited to the study of dynamic interactions involving such states.

In the work described here electron densities in excess of 10^{16} cm^{-3} with mean temperatures of $\sim$45 eV have been created in a pulse of 600 ps duration. For comparison, transversely excited atmospheric discharges[2] and e-beam–excited plasmas[3] operate with typical electron densities of $\leq10^{15}$ cm^{-3}, temperatures of $\sim$5 eV, and pulses of, at shortest, several nanoseconds. When electron cooling by inelastic collisions with absorber atoms (72 eV/ns at a Ne density of 10^{17} cm^{-3}) during the x-ray pulse is taken into account, the photoionization electron source (PES) described here can be considered to produce an effective current density of 3×10^5 A cm^{-2}. The utility of the PES is a result of the modest energy (60 mJ) of the laser which was required to produce the x-ray–emitting plasma and of its repetition rate of 2 pulses/s.

To make use of the hot electrons produced by the PES, "target" atoms were mixed with the rare-gas absorber. The electrons then excited the target atoms by inelastic collisions. In this investigation the novel characteristics of the PES were used to examine the production of Li atoms in the metastable ($1s2s2p$) $^4P^\circ$ level at 57.4 eV and to measure the decay rate from this level under conditions of relatively high electron density. The production of large densities of such metastable atoms in the alkali metals is of interest because of their suitability as storage states for recent extreme-ultraviolet (XUV) laser proposals.[4,5] Although the lifetime of Li ($1s2s2p$) $^4P^\circ$ has been measured[6] to be 5 μs in an atomic beam, it was unclear before this investigation whether high electron and ion densities would result in anomalously rapid decay of this level because of coupling to states which autoionize to the Li$^+$ ($1s^2$) continuum. The results described here show conclusively that no such anomalously rapid decay occurs.

The configuration of the experiment is illustrated in Fig. 1. A massive plane tantalum target was placed inside a cell containing a mixture of a rare-gas absorber, Ne, and the target species, Li. A 60-mJ, 600-ps pulse from a Nd-doped yttrium aluminum garnet laser (1.06 μm) was focused onto the target and a plasma was formed.[7] For the focal spot intensity (1.0×10^{13} W/cm^2) and pulse length used, the resulting x-ray

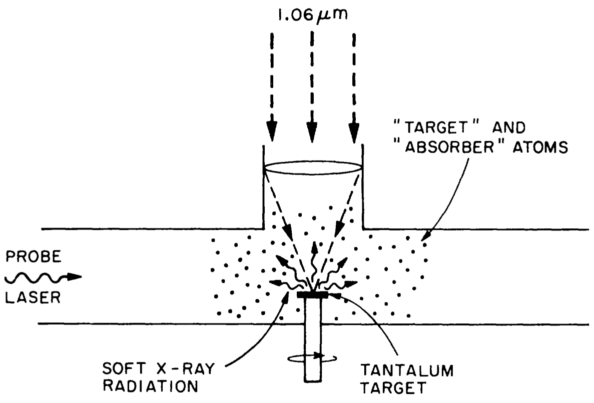

FIG. 1. Schematic of experimental configuration.

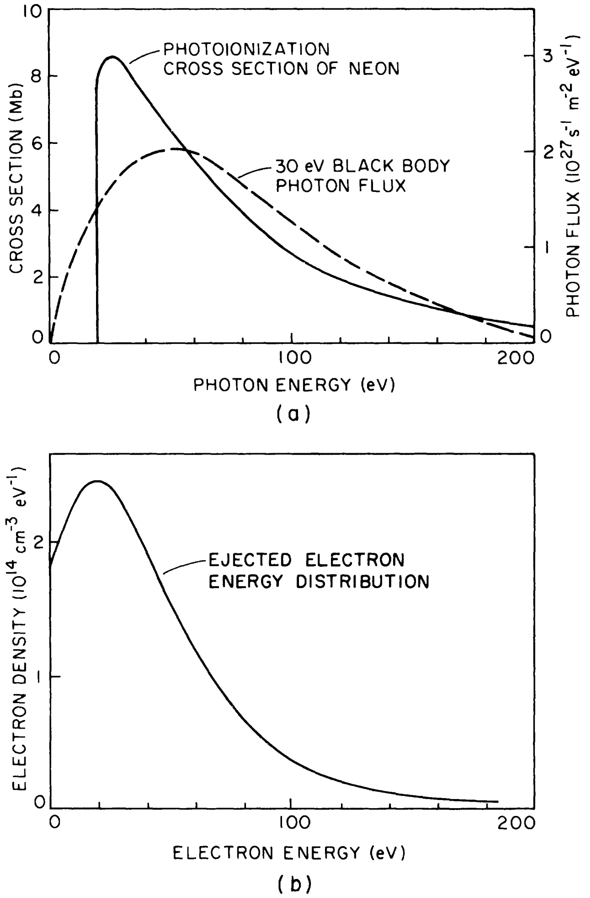

FIG. 2. (a) Photoionization cross section of Ne and the x-ray spectrum 1 mm from the target. (b) Predicted ejected electron distribution for a Ne density of 3×10^{17} cm^{-3}.

emission can be considered as that of a 30-eV blackbody.[7] The conversion efficiency of 1.06-μm radiation to x rays under the conditions of this experiment is believed to be approximately 10%.[7] The x rays emitted from the plasma photoionize the surrounding absorber atoms to produce high densities of electrons.

For a spectral intensity distribution, $I(\omega)$, at a fixed distance from the tantalum target, the ejected-electron energy distribution is given by $n(E) = N\sigma(\omega)I(\omega)\Delta t/\hbar\omega$. N and $\sigma(\omega)$ are the number density and photoionization cross section of the absorber atom, Δt is the x-ray pulse length, and $E = \hbar\omega - E_I$. E_I is the ionization threshold of the absorber atom. Figure 2(a) shows the good spectral overlap of the emission of a 30-eV blackbody and the photoionization cross section of Ne. The predicted electron distribution for a Ne absorber gas density of 3×10^{17} cm^{-3} is plotted in Fig. 2(b). This electron distribution has a maximum at 20 eV and a mean electron energy of 45 eV. The total ejected electron density is 2×10^{16} cm^{-3}.

In order to investigate the excitation of the Li$(1s2s2p)\,{}^4P^\circ$ level by the PES, the plasma cell was heated to provide a target Li atom density of 10^{17} cm^{-3}. Varying amounts of Ne absorber gas were then added. The appropriate energy level diagrams are shown in Fig. 3. The number-density–length product of Li$({}^4P^\circ)$ atoms, N^*L, was determined by measuring the absorption of a probe dye-laser beam, as a function of wavelength at the Li $(1s2s2p)\,{}^4P^\circ$–$(1s2p^2)\,{}^4P$ transition at 371 nm, and matching the resulting absorption traces with numerically generated Voigt profiles. All measurements were taken at a distance of 1 mm from the tantalum target where, as a result of geometrical considerations,[7] the number density, N^*, is given by $N^* = 6.4\,N^*L$. The dye laser was pumped by a harmonic of the same Nd-doped yttrium aluminum garnet laser beam which produced the plasma and thus had a pulse length of 600 ps.

Figure 4 shows the measured dependence of N^*L on the Ne density. The prediction of a rate-equation model is plotted as the solid curve. This model predicts both the magnitude and the shape of the experimental curve very well. The saturation of N^*L at high Ne densities can be explained by noting that the density of Li$({}^4P^\circ)$ atoms depends not only on the number of hot electrons, but also on their cooling time. At low Ne densities this cooling time is determined by inelastic collisions with Li atoms. At high Ne densities, however, electron cooling is dominate by collisions with

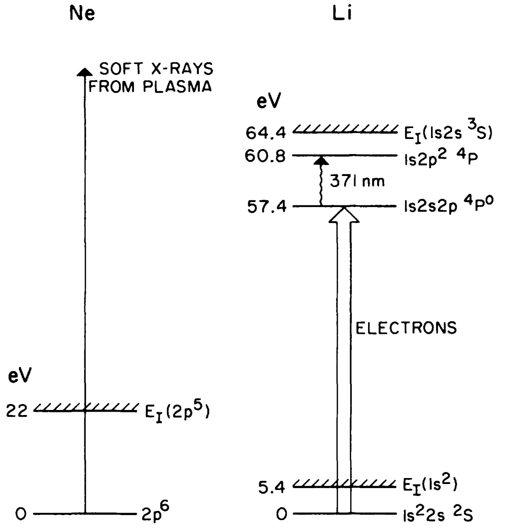

FIG. 3. Energy-level diagram showing photoionization of the absorber atom and the subsequent electron excitation of the target atom. The laser-probe transition is also indicated.

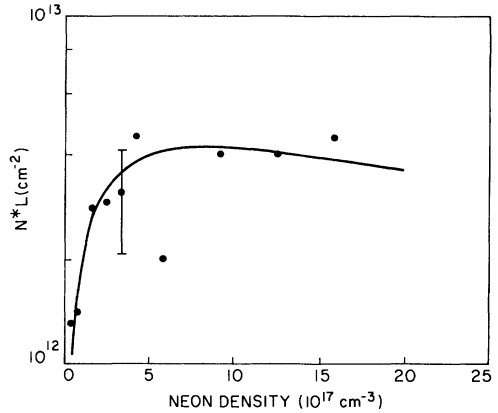

FIG. 4. N^*L for Li$(1s2s2p)\,^4P^\circ$ vs Ne density. The solid curve is the rate-equation model prediction multiplied by 1.2. Li density = 10^{17} cm^{-3}. The time delay between the peaks of the probe and 1.06-μm laser pulses ~ 1 ns.

Ne atoms. In this regime, as the Ne density increases the electron density increases but the cooling time decreases. Thus, the magnitude of the Li($^4P^\circ$) population approaches a constant value.

The large number densities of Li($^4P^\circ$) atoms measured in this work are not immediately applicable to the XUV laser proposal of Harris[4] because of the high absorption of Ne at the 207-Å XUV laser transition. However, the general suitability of the PES for use in similar XUV laser schemes, involving different target and adsorber atoms,[5] is well illustrated by the observation that the number densities of Li($^4P^\circ$) atoms measured here of ~ 3×10^{13} cm^{-3} exceed those obtained by conventional discharges[8] by approximately three orders of magitude.

Under the conditions of Li and Ne density used here, the cooling time is approximately 50 ps. Thus no excitation of Li($^4P^\circ$) metastable atoms occurs after the x-ray pulse is terminated, allowing the decay from this level to be observed. To measure the rate of this decay, a variable delay was inserted in the path of the probe beam. Figure 5 shows the dependence of the Li($^4P^\circ$) population on the delay between the probe laser and the plasma pulses. The observed decay time of 2.5 ± 0.5 ns is comparable to that measured under similar conditions[7] for decay of the Li$^+(1s2s)\,^1S$ ion.

The electron density, at a time 1 ns after the peak of the x-ray pulse, has been measured to be $(1.3\pm0.8)\times10^{15}$ cm^{-3} [from the Stark broadening of the Li$(1s^24d)$ level]. If one assumes that electrons are responsible for the deexcitation, these measurements imply a deexcitation rate constant for the Li $(1s2s2p)\,^4P^\circ$ level of $(3^{+7}_{-1})\times10^{-7}$ cm^3 s^{-1}. This is typical of a dipole-allowed electron excitation of several electronvolts. It can thus be concluded that no anomalously rapid deexcitation of the Li($^4P^\circ$) level occurs at electron densities of ~ 10^{15} cm^{-3}.

In the work reported here a novel photoioniza-

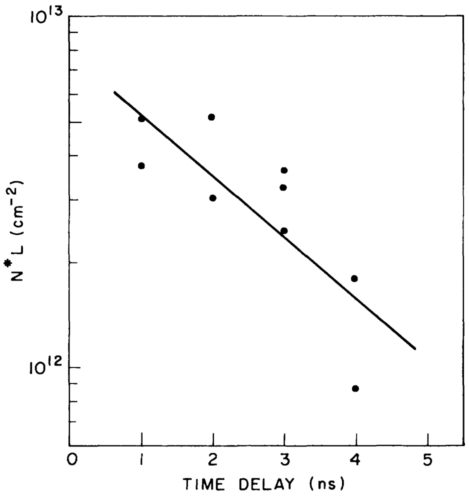

FIG. 5. N^*L for Li$(1s2s2p)\,^4P^\circ$ vs the time delay between the peaks of the probe and 1.06-μm laser pulses. Ne density = 3×10^{17} cm^{-3}; Li density = 10^{17} cm^{-3}. The solid line is a fit to the data yielding $\tau_{\mathrm{decay}} = 2.5$ ns.

tion source capable of producing a subnanosecond burst of hot electrons has been described. The high electron density and current produced by the PES have been used to excite large densities of Li($^4P^\circ$) atoms at 57 eV, and the short duration of the hot-electron pulse has enabled the deexcitation rate from the Li($^4P^\circ$) level to be measured. Although the duration of the plasma-producing laser pulse used here was 600 ps, the PES should work well for pulses as short as 50 ps. Still shorter pulses should be obtainable at the cost of poorer conversion efficiency from laser light to x rays.

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