Shake-up as a mechanism for vacuum-ultraviolet lasers

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We show how electron shake-up, as it occurs during core photoionization, may produce population inversion in the vacuum ultraviolet. Calculations for Li show the possibility of lasers at 165.3 and 113.2 nm.

During the past several years it has been shown that laser-produced soft x rays may be used to produce large densities of core-excited atoms[1,2] and, in some cases, inversion and lasing on visible and ultraviolet transitions.[3] It is of interest to search for mechanisms to allow the use of this efficient method of population production to produce inversion in the VUV and XUV regions of the spectrum. For example, one such method using super-Coster–Kronig decay was recently suggested.[4]

In this Letter we suggest the use of shake-up, as it occurs during core electron photoionization, to produce such inversions. We illustrate this with calculations for Li, which show the possibility of efficient lasers at 165.3 and 113.2 nm.

Figure 1 is an energy-level diagram for the Li system. Soft x rays from a laser-produced plasma are used to photoionize the 1*s* electron of neutral Li, thereby (as shown earlier)[1,2] producing population densities as large as 10^{15} ions cm^{-3} in the Li^+ $(1s2s)^3S$ level. According to the calculations of Larkins *et al.*[5] and the recent measurements of Ferrett *et al.*,[6] Krummacher *et al.*,[7] and Geard,[8] the shake-up of the valence electron from the 2*s* to the 3*s* orbit will produce populations of approximately 20% of this magnitude in the Li^+ $(1s3s)^3S$ level. This level will invert with regard to the Li^+ $(1s2p)^3P$ level, producing gain at 165.3 nm.

Although, in principle, the process of conjugate shake-up may populate the Li^+ $(1s2p)^3P$ level, it is not likely to do so at a magnitude that will destroy the inversion. For an incident photon energy of 151 eV, Larkins *et al.*[5] calculate the ratios of population produced by photoionization in the Li^+ $(1s2s)^3S$, $(1s3s)^3S$, and $(1s2p)^3P$ levels to be 100, 24.4, and 0.7, respectively. Recent experimental work of Geard[8] in the near-threshold region of 75–90 eV shows these populations to have an average ratio of about 100, 20, and 10, respectively. Although it is much less favorable than those of Larkins, allowing for the favorable degeneracy factor of 3, the effective inversion produced by photoionization by photons in this energy range is about a factor of 6.

The Einstein *A* coefficient for the Li^+ transition at 165.3 nm is 3×10^8 sec^{-1}. For a Doppler width of 0.6 cm^{-1}, this implies a gain cross section of 1×10^{-13} cm^2. [In Li (Ref. 6) the hyperfine splitting is approximately 0.2 cm^{-1} and can be neglected.] The Li^+ $[(1s4d)^3D–(1s2p)^3P]$ transition at 113.2 nm also has a strong radiative transition probability. Here $A = 4 \times 10^8$ sec^{-1}, and the gain cross section is 9×10^{-14} cm^2. The upper level of this transition can be reached by using the quadrupole transition at 359 nm ($f = 4 \times 10^{-6}$) or by two-photon pumping.

Although the primary process of inner-shell photoionization leads to an inversion on the Li^+ $[(1s3s)^3S–(1s2p^3P]$ transition, secondary processes that occur in the photoionized medium may destroy this inversion. To analyze these processes, and to determine the optimum conditions for an experiment to demonstrate laser action at 165 and 113 nm, the photoionized plasma has been studied using a rate-equation model that will be described in detail elsewhere.[9] This model includes the processes of inner- and outer-shell photoionization of Li by the laser-plasma-produced soft x rays, ionization of Li atoms by collisions with electrons; three-body collisional recombination, excitation and de-excitation of excited states of Li and Li^+ by inelastic collisions with electrons and by spontaneous decay, and electron–electron colli-

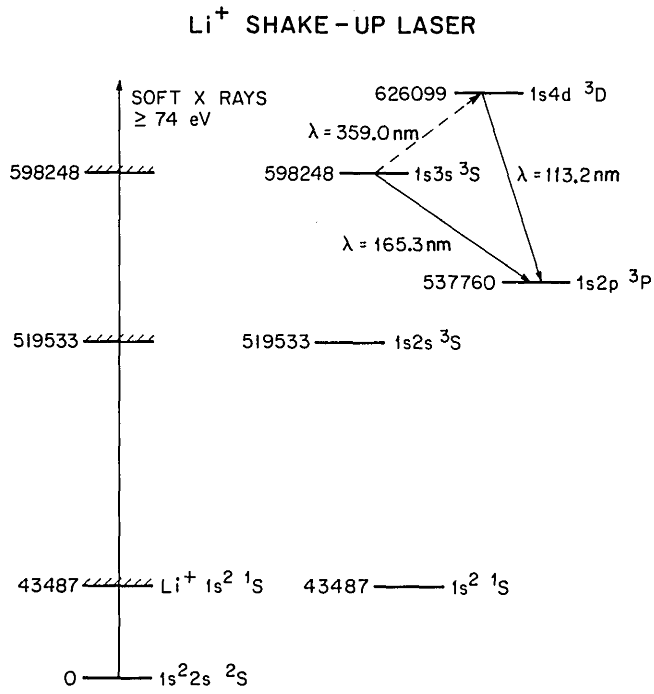

Fig. 1. Li shake-up laser energy levels.

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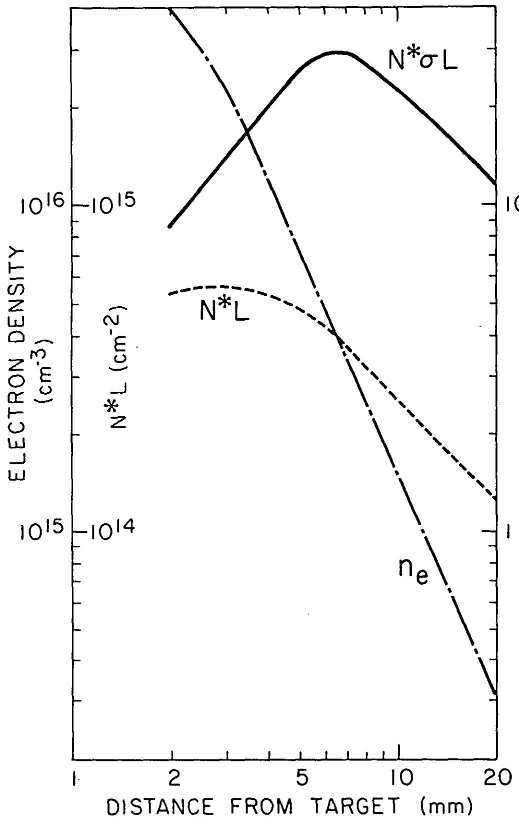

Fig. 2. Model predictions of electron density n_e, effective population times length N^*L, and gain $N^*\sigma L$ at 165.3 nm as a function of distance from target. The model assumes three target spots, each irradiated by a 1-nsec, 2-J, 1.06-μm laser pulse.

sions leading to thermalization of the initially non-Boltzmann electron distribution.

The modeling is for three target plasmas, each produced by 2 J of 1.06-μm laser energy in a 1-nsec pulse and focused to an intensity on target of 10^{13} W cm^{-2}. The target plasmas are assumed to be in a line, separated from one other by several centimeters. The reason for using several target plasmas is that they permit, at fixed maximum electron density, a larger gain–distance product than does a single plasma. The soft x rays are assumed to be emitted with a blackbody spectrum with a temperature of 30 eV, and the conversion efficiency from 1.06-μm energy to soft-x-ray energy[1,2] is taken as 10%. The density of the Li vapor that surrounds the target is taken as 10^{17} atoms cm^{-3}. This gives a stopping distance for the laser-produced x rays of approximately 3 cm.

Figure 2 shows the predictions of this model as a function of the perpendicular distance from any of the three target plasmas. The quantities shown are evaluated at 1.5 nsec after the beginning of the laser pulse, which is the time of peak 165.3-nm gain. The electron density n_e falls roughly as the square of the distance from target. The integral $N^*L \equiv \int N^* dl$ of the degeneracy weighted population, $N^* = [\mathrm{Li}^+ (1s3s)^3S\text{–}\mathrm{Li}^+ (1s2p)^3P/3]$, times incremental distance dl parallel to the target plasmas peaks in the vicinity of 2–5 mm from target. This quantity flattens as the target is approached, primarily as a result of inelastic electron collisions, which populate the lower laser level $\mathrm{Li}^+ (1s2p)^3P$, from the $\mathrm{Li}^+ (1s2s)^3S$ level. The gain $N^*\sigma L$ at 165.3 nm maximizes somewhat sharply at about 8 mm. This maximization is caused by increased Stark broadening and therefore decreasing gain cross section σ at small distances from target and by the decreasing N^*L product at large distances from target.

From Fig. 2 it is seen that a gain of exp(30) is predicted at 165.3 nm with 6 J of laser energy. Allowing for the use of mirrors, it seems likely that the laser will operate at energies of less than 1 J. At the assumed density of 10^{17} Li atoms cm^{-3}, the fraction of the soft-x-ray energy populating the $\mathrm{Li}^+ (1s3s)^3S$ level in a cylindrical region of 2-mm diameter, centered at the optimum distance of 8 mm, is about 1%. Assuming saturation of the gain medium, and 10% laser to soft-x-ray conversion, this leads to an overall 1.06- to 165-nm efficiency of 0.1%. Since the cross section for photoionization of the valence electron at 165.3 nn is about 1.8×10^{-18} cm^2, the $1/e$ absorption depth at a Li ground-level density of 10^{17} atoms cm^{-3} is 5.5 cm, and attention will have to be given to designing a sufficiently short Li zone.

Although we will not go into detail here, we note that attractive shake-up and shake-up transfer lasers are also possible in the singlet series of Li^+. The wavelengths of the $(1s3s)^1S \rightarrow (1s2p)^1P$ and $(1s4d)^1S \rightarrow (1s2p)^1P$ transitions are 175.5 and 123.7 nm, respectively. Geard[8] indicates a $\mathrm{Li}^+ (1s3s)^1S$ to $\mathrm{Li}^+ (1s3s)^3S$ population ratio that is possibly as large as 50%. An advantage of the singlet systems is that the lower laser level $\mathrm{Li}^+ (1s2p)^1P$ has a radiative decay time of 39 psec, and therefore the question of the magnitude of the conjugate shake-up process need not be considered. Also, because this system does not self-terminate, long pulse excitation and therefore resonators could be employed.

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