Proposal for a 207-Å laser in lithium

S. E. Harris

Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305

Received October 1, 1979

A technique is proposed for using the properties of the autoionizing levels of neutral Li to construct a 207-Å laser. Energy is first stored in the lowest quartet level at 57.4 eV and then, by use of an intense laser beam, rapidly transferred to a radiating level in the doublet series.

At constant oscillator strength, the radiative lifetime varies as the square of the transition wavelength and for a strong transition near 200 Å is about 10^{-11} sec. To overcome the difficulty of obtaining sufficient gain in this short a time, it was suggested several years ago[1,2] that atoms first be stored in a metastable level and then, by use of an intense tunable laser, rapidly transferred to the upper level of the lasing transition. These proposals were based on single-electron-excited states and thereby included the difficult requirement that the population of the metastable level be inverted with respect to the ground state of the species.

This Letter describes how, by permitting the participation of both inner- and outer-shell electrons, the properties of the *autoionizing levels* of Li I may be utilized to construct a two-step laser that has the following properties: (1) the storage level is metastable against both autoionization and radiation, (2) the upper level of the lasing transition (the target level) is metastable against autoionization but is strongly radiatively allowed, and (3) the terminal level of the lasing transition is other than ground and is emptied both radiatively and by photoionization by the incident pump laser.

An energy-level diagram of the proposed system is shown in Fig. 1. Figure 2 shows the changes in electron configuration and spin that are involved.

Before proceeding, I note that other authors[3–5] have noted how, in different ways, the properties of long-lived autoionizing states may bear on the soft x-ray laser problem.

By use of electron excitation, energy is first stored in the $1s2s2p\ ^4P^0$ level. This level is the lowest of the quartet series and, as described in classic papers by Feldman and Novick,[6] is coulombically stable (spin forbidden) against both autoionization and radiation. Its fine-structure components, $J = 1/2, 3/2, 5/2$, are separated by about 1 cm^{-1} and have experimentally measured lifetimes of 0.14, 0.46, and 5.8 μsec, respectively.[7] The level may be populated not only by direct electron excitation but also by radiative cascade from all higher levels in the quartet series[8,9] and by recombination. The cross section or direct excitation by 60-eV electrons is about 10^{19} cm^2.[6,7]

In the second step of excitation, we use an intense pump laser at $\lambda_p = 2949$ Å to transfer the (previously unmoved) $2s$ electron to a $2p$ orbit and also to flip its spin, thereby rapidly populating the radiatively allowed $1s2p^2\ ^2P$ level (Figs. 1 and 2). The oscillator strength of this intercombination line ($1s2s2p\ ^4P^0$–$1s2p^2\ ^2P$) is not known and is key to this proposal. As a guide, I take it equal to that of the comparable 2^3S–2^1P of Li^+, which involves the same orbit and spin change, and assume that $f = 2.83 \times 10^{-7}$.[10]

It is particularly important that the target level

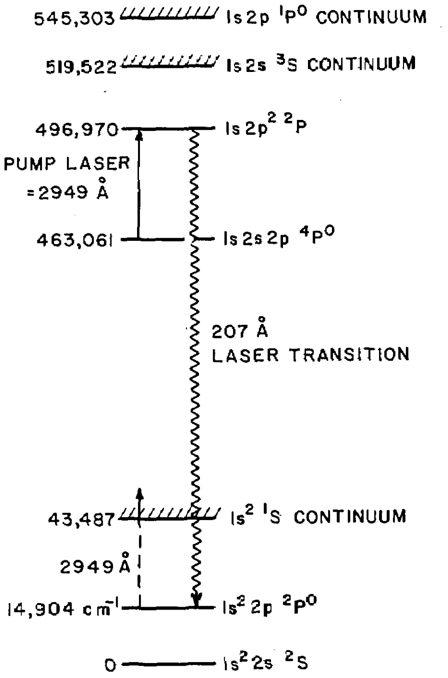

Fig. 1. Energy-level diagram for the proposed 207-Å laser in neutral Li.

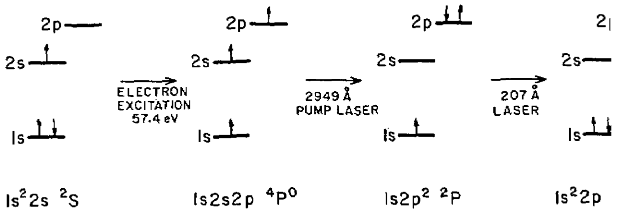

Fig. 2. Configuration and spin of the pertinent levels.

0146-9592/80/010001-03$0.50/0

($1s2p^2\,{}^2P$) be metastable against autoionization. This results from a combination of (coulomb) parity and angular-momentum selection rules. By parity, the ejected electron must be s or d, and thus the end product must have a total angular momentum (ion plus electron) that is S or D. The initial momentum is P, and therefore the autoionization is not allowed. This level has been observed to decay radiatively at 207 Å with a measured lifetime of 15×10^{-12} sec.[11] In recent experiments, McIlrath and Lucatorto[12] measured the energy of this level by photoabsorption from the laser-excited $1s^22p\,{}^2P^0$ level. The fact that $1s2p^2\,{}^2P$ is not seen in the ejected-electron spectrum[13,14] (whereas $1s2p^2\,{}^2D$ and $1s2p^2\,{}^2S$ are seen) confirms that its lifetime is dominantly radiative.

The other important property of this system is that the pump laser has a wavelength that is sufficiently short to photoionize and thereby to empty the terminal $1s^22p\,{}^2P^0$ laser level (Fig. 1) but still not short enough to photoionize either the $1s2s2p\,{}^4P^0$ storage level or the $1s2p^2\,{}^2P$ target level. (Note that the latter may only photoionize into the $1s2p\,{}^1P^0$ or $1s2p\,{}^3P^0$ continua.)

The laser transition at 207 Å will be Doppler broadened and at a Li atom temperature of 800 K will have a linewidth of 3.7 cm^{-1}. For the radiative lifetime of $\tau_{sp} = 15 \times 10^{-12}$ sec, the laser-gain cross section is $\sigma_G = 9.7 \times 10^{-14}$ cm^2; i.e., an inversion density of 10^{12} atoms/cm^3 will produce a gain of 9.7% /cm.

For the assumed intercombination oscillator strength of $f = 2.83 \times 10^{-7}$, the absorption cross section for the 2949-Å pump laser is $\sigma_{\text{abs}} = 4.5 \times 10^{-19}$ cm^2, where the dominant broadening of this transition is the natural linewidth of the upper level. This in turn implies a laser-induced transition rate (on the intercombination transition) of $W = 0.67\,P/A$ sec^{-1}, where P/A (W/cm^2) is the 2949-Å laser-power density.

If the pumping laser is on for a time long compared with τ_{sp}, and if the density N (atoms/cm^3) of the metastable storage level is not depleted, then the target level density is $W\tau_{sp}N$, and the gain on the 207-Å laser transition is:

$$\text{Gain} = (9.7 \times 10^{-25})N(\text{atoms/cm}^3)\,\frac{P}{A}\,(\text{W/cm}^2)\ \text{cm}^{-1}. \qquad (1)$$

For example, for $N = 10^{12}$ atoms/cm^3 and a pump-laser power density of $P/A = 5 \times 10^{10}$ W/cm^2, the 207-Å gain would be 4.8%/cm.

In Eq. (1), we assume that the lower level, $1s^22p\,{}^2P^0$, of the laser transition is empty. Although this level may decay radiatively to ground, the decay rate (27 nsec if optically thin) is not sufficient to prevent self-termination on the 207-Å transition. The cross section for photoionization from this level by the 2949-Å pump laser is about 1.5×10^{-17} cm^2,[15] and thus, at a pump-power density of 5×10^{10} W/cm^2, the level will be emptied in about 10^{-12} sec. The cross section for absorption of 207-Å radiation by ground-state Li is about 10^{-19} cm^2 (Ref. 16) and should not be a problem.

At least in early experiments, it will be desirable to use a method of electron heating that lends itself to long dimensions and also that does not tend to saturate at the first ionization potential. An attractive possibility on which we are now working in our laboratory is heating with high-peak-power microwave radiation in the presence of a longitudinal magnetic field. The magnetic field is adjusted so the electron-cyclotron frequency is larger than the microwave frequency, thereby ensuring that the plasma dielectric constant remains real. We note, though, that visible lines within the Li quartet series are observed in hollow-cathode discharges.[17]

Table 1. Summary of Parameters

Transition wavelength	207 Å
Metastable storage density	10^{12} atoms/cm^3
Li discharge length	3 m
2949-Å pump-pulse length	30 psec
2949-Å pump energy	3.3 mJ
Single-pass gain	$e^{14.5}$
Per-pulse energy at 207 Å	6.6 μJ
207-Å angular collimation	0.16 mrad

A summary of parameters, as they might be in an early experiment, is given in Table 1. We assume a storage density of 10^{12} atoms/cm^3. (This is about the same as that of the He 2^1S or 2^3S levels for He–Ne laser operating conditions.) A 2949-Å laser pump-power density of 5×10^{10} W/cm^2, confocally focused to a cell length of 3 m (beam area $\cong 2.2 \times 10^{-3}$ cm^2), and on for a time of 30 psec, requires a 2949-Å pump energy of 3.3 mJ and yields a single-pass gain of $e^{14.5}$. Available reflectors at 207 Å have at least a few percent reflectivity and should permit multipass operation with a reduction in the necessary storage density or pump power. Assuming such operation, which allows most of the stored energy to be extracted on the final pass, the per-pulse output energy will be about 6 μJ. The 207-Å collimation will be determined by the 2949-Å pump laser acting as an optical stop and, for the assumed 3-m length, will be 0.16 mrad.

The author acknowledges several helpful discussions with T. Lucatorto, D. Ederer, and A. Weiss of the National Bureau of Standards. Discussions with R. Anholt, R. Falcone, W. Meyerhoff, and J. Young are also gratefully acknowledged. This research was supported by the U.S. Office of Naval Research.

References

1. H. Mahr and U. Roeder, "Use of metastable ions for a soft x-ray laser," Opt. Commun. **10**, 227 (1974).
2. S. A. Mani, H. A. Hyman, and J. D. Daugherty, "Lithium-ion soft x-ray laser," J. Appl. Phys. **47**, 3099 (1976).
3. E. J. McGuire, "The *L-MM* auger spectra of Na and Mg," Phys. Rev. A 14, 1402 (1976).
4. E. J. McGuire and M. A. Duguay, "Soft x-ray gain in the alkali earths," Appl. Opt. 16, 83 (1977).
5. C. K. Rhodes and R. M. Hill, SRI International, Menlo Park, California 94025, "Laser excitation of innershell atomic states by multiquantum processes," unpublished (1977).
6. P. Feldman and R. Novick, "Autoionizing states in the

alkali atoms with microsecond lifetimes," Phys. Rev. Lett. **11**, 278 (1963); "Auto-ionizing states in the alkali atoms with microsecond lifetimes," Phys. Rev. **160**, 160 (1967).

7. M. Levitt, R. Novick, and P. D. Feldman, "Determination of energies and lifetimes of the metastable auto-ionizing $(1s2s2p)^4P$ states of Li^6 and Li^7 by a Zeeman-quenching technique," Phys. Rev. A **3**, 130 (1971).
8. H. G. Berry, "Multiply-excited states in beam-foil spectroscopy," Phys. Scr. **12**, 5 (1975); H. G. Berry, E. H. Pinnington, and J. L. Subtil, "Energies and mean lives of doubly excited terms in lithium," J. Opt. Soc. Am. **62**, 767 (1972).
9. C. F. Bunge and A. V. Bunge, "Absolute term values for the quartet states of neutral lithium," Phys. Rev. A **17**, 816 (1978); "Configuration-interaction studies of transition energies and oscillator strengths for the lowest quartet states of neutral lithium," Phys. Rev. A **17**, 822 (1978).
10. G. W. F. Drake and A. Dalgarno, "Intercombination oscillator strengths in the helium sequence," Astrophys. J. **157**, 459 (1969).
11. J. P. Buchet, M. C. Buchet-Poulizac, and H. G. Berry, "Classifications of some transitions in doubly excited Li I and Li II," Phys. Rev. A **7**, 922 (1973).
12. T. J. McIlrath and T. B. Lucatorto, "Laser excitation and ionization in a dense Li vapor: observation of the even-parity, core-excited autoionizing states," Phys. Rev. Lett. **38**, 1390 (1977).
13. D. Rassi, V. Pejcev, and K. J. Ross, "The ejected-electron spectrum of Li I autoionizing levels excited by low-energy electron impact on lithium vapour," J. Phys. B **10**, 3535 (1977).
14. P. Ziem, R. Bruch, and N. Stolterfoht, "Autoionization spectra of Li I and Li II excited by H^+ and He^+ impact," J. Phys. B **8**, L480 (1975).
15. M. Aymar, E. Luc-Koenig, and F. Combet Farnoux, "Theoretical investigation on photoionization from Rydberg states of lithium, sodium, and potassium," J. Phys. B **9**, 1279 (1976).
16. D. L. Ederer, T. Lucatorto, and R. P. Madden, "Autoionization spectra of lithium," Phys. Rev. Lett. **25**, 1537 (1970); "Resonances in the photoionization continuum of lithium I (55 to 70 eV)," J. Phys. Paris **C4**, 85 (1971).
17. G. Herzberg and H. R. Moore, "The spectrum of Li^+," Can. J. Phys. **37**, 1293 (1959).