Proposal for a 19.9-nm laser in lithium pumped by a noncoherent x-ray pulse

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We demonstrate theoretically the feasibility of 19.9-nm lasing on the Li^+ $1s2p\ ^1P \rightarrow 1s^2$ transition in Li vapor pumped by a short, noncoherent soft-x-ray pulse with the narrow-band spectrum centered at the $[1s(3s3p)\ ^3P]\ ^2P^\circ$ resonance of neutral Li (≃71 eV). Broadband pumping of $1s^22p$-excited Li atoms is also briefly discussed.

An x-ray laser pumped through inner-shell photoionization has not been realized, although a number of proposals[1-4] as well as successful experiments with longer-wavelength radiation[5] have been reported. One of the main obstacles to the proposed lasing to the ionic ground level[1] has been that such pumping would create free electrons that are energetic and numerous enough to destroy population inversion by electron-impact ionization of neutral atoms.

Here we demonstrate theoretically how the problem of 19.9-nm lasing in Li^+ pumped by inner-shell photoionization and photoexcitation on neutral Li atoms can be solved. To limit the detrimental effect that the photoelectrons created by the pumping radiation have on the population inversion, we suggest using a short soft-x-ray pulse with a narrow-band spectrum centered at the Li $[1s(3s3p)\ ^3P]\ ^2P^\circ$ resonance to excite ground-state Li atoms. Subsequent fast autoionization of part of the inner-shell excited Li atoms to the upper laser level (Li^+ $1s^2p\ ^1P$) is complemented by the direct 1s-photoionization of Li atoms to the upper laser level. (Direct inner-shell photoionization pumping by the Se^{24+} x-ray laser has been proposed for a Na vacuum laser[6] at a much longer wavelength of 37.2 nm.) The suggested pumping scheme relies on both the relatively large photoabsorption cross section and the probability of the inner-shell (auto)ionization to the Li^+ $1s2p\ ^1P$ state in the vicinity of $[1s(3s3p)\ ^3P]\ ^2P^\circ$ resonance of neutral Li[7-9] (≃71 eV about the ground level). At the same time, our calculations show that the energy distribution of emerging photoelectrons does not permit them to destroy population inversion significantly. The spectral intensity of the short pumping pulse that is necessary to attain significant gain seems feasible at the state of the art of the laser-plasma technology. It is also possible that a much lower x-ray pulse spectral intensity may be needed in order to pump the excited Li atoms by a broadband short x-ray pulse.

It is well known that the absorption of soft-x-ray radiation (the photon energy in the region 57–100 eV) by Li atoms may produce double-excited states with energies much higher than the 2s-ionization potential of 5.39 eV of neutral Li.[6,10-13] A Li atom in many of these states is unstable against autoionization: in a very short time (10–100 ps) it emits a free electron, thus becoming a Li^+ ion in one of the excited states. (The probability that this ion will be left in the ground state is negligible, since the probability of autoionization to the energetically closest ionic state is usually the largest.[14])

Double-excited autoionizing Li atomic states correspond to broad resonances in the photoabsorption spectra of Li vapor for the energy of the incident photons above 58 eV. For an incident photon energy large enough to ionize a ground-state Li atom to the Li^+ $1s2p\ ^1P$ state, the strongest such resonance corresponds to the $[1s(3s3p)\ ^3P]\ ^2P^\circ$ (71.14 eV above the Li ground level) state of Li atoms.[7,8,11] The energy interval between $E' = 71.05$ eV and $E'' = 71.25$ eV corresponds approximately to the width of this resonance. Absorption of a photon with such energy would immediately result[15] in either (i) excitation of a Li atom to the $[1s(3s3p)\ ^3P]\ ^2P^\circ$ state followed by autoionization to one of the four ionic states $1s2s\ ^1S$, $1s2s\ ^3S$, $1s2p\ ^1P$, or $1s2p\ ^3P$ and by freeing of an electron with an energy of approximately 5.0, 6.7, 3.5, or 4.5 eV, respectively; (ii) direct (nonresonant) inner-shell ionization to the same ionic and photoelectron states; or (iii) 2s ionization, i.e., creation of a Li^+ ion in the ground $1s^2$ state and a free 65.7-eV electron (see Fig. 1). The average (over the suggested pumping interval) total photoabsorption cross section can be estimated as $\bar{\sigma}_{tot} \simeq 5.8 \times 10^{-18}$ cm^2.[11] Since the $[1s(3s3p)\ ^3P]\ ^2P^\circ$ autoionization time $\simeq 1.5 \times 10^{-14}$ s (Ref. 11) is much shorter than any other characteristic time of a Li atom, we do not separate processes (i) and (ii), which both populate the upper laser level.

Process (iii) creates ground-state Li ions, thus decreasing the population inversion. However, the 2s-photoionization cross section is quite low for a photon energy so high above the 2s-ionization potential of 5.39 eV: $\sigma_{2s} \simeq 7 \times 10^{-20}$ cm^2.[16] As a result, the Li^+ ground-level population resulting from direct 1s-photoionization does not exceed $\sigma_{2s}/\bar{\sigma}_{tot} \simeq 0.013$ of the population of the ionic excited levels and may be neglected.

Processes (i)–(iii) are directly initiated by the pumping radiation. In turn they bring about a variety of secondary processes. We briefly discuss the most important of these processes: (a) the 2s ionization of Li atoms by electron impact (which has been considered the main danger

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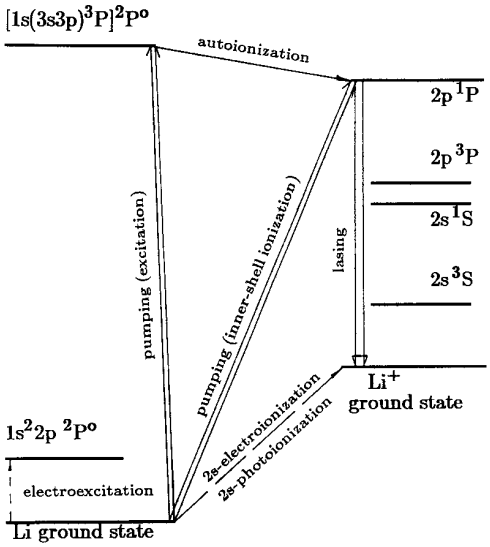

Fig. 1. Partial energy-level diagram of Li^+ 19.9-nm lasing.

to the population inversion); (b) collisional and radiative transitions from the upper laser level to levels other than the lower laser level; (c) the electron-impact excitation of the ground-state Li atoms. (From now on, we assume a Li atomic density of $N_0 = 5 \times 10^{17}$ cm^{-3}.[7]) The rates of most of these processes are substantially different for the following three groups of free electrons created by the pumping radiation: the fast electrons (with an energy of $\simeq$65.7 eV), which result from 2s photoionization; the slow electrons, which originate from the autoionization or direct 1s photoionization of Li atoms to the Li^+ 1$s2s$ ^3S level (with an energy of $\simeq$6.5 eV, higher than the first ionization potential of Li atoms); and for the ultraslow electrons, which are created by ionization to the Li^+ 1$s2s$ ^1S, 1$s2p$ ^1P, and 1$s2p$ ^3P levels (with energies lower than the first ionization potential of Li atoms). Immediately after the pumping pulse the densities of the fast, slow, and ultraslow electrons (N_f, N_s, and N_u, respectively) are related according to the following equations:

$$N_f \simeq (\sigma_{2s}/\bar{\sigma}_{\text{tot}})(N_s + N_u), \tag{1}$$

$$N_s + N_u \simeq N_2 r^{-1}, \qquad N_s = k(N_s + N_u), \tag{2}$$

where N_2 is the upper laser level population, r is the ratio of the number of desirable transitions (that is, the transitions from the Li ground level to the upper laser level) to the total number of absorbed photons. It can be estimated from Ref. 7 that $k \simeq 0.6$ and $r \simeq 0.2$. It is taken into account in relations (1) and (2) that the proportion of fast electrons is relatively low, since the 2s-photoionization cross section is $\simeq$1/80 of the 1s-photoionization cross section for a photon energy near 70 eV.

The cross section of the 2s-ionization of a neutral Li atom by electron impact is $\sigma_{\text{ion}} \simeq 10^{-16}$ cm^2 for the slow electrons[17] and is negligible for the ultraslow electrons, since the energies of the latter are lower than the Li 2s-ionization threshold. Immediately after a pumping pulse, that is, before other processes change the electron energy and density, the electron-impact ionization rate coefficient for the slow electrons can be defined as

$$X_{\text{ion}} \simeq \sigma_s v_s, \tag{3}$$

v_s being the velocity of the slow electrons, which yields $X_{\text{ion}} \simeq 1.5 \times 10^{-8}$ cm^3 s^{-1}. Hence the rate of the Li 2s ionization caused by the slow electrons, or, in other words, the rate of the population inversion decrease resulting from this process, could be estimated as

$$R_s \equiv -\mathrm{d}N_0/\mathrm{d}t = \mathrm{d}N_s/\mathrm{d}t = X_{\text{ion}} N_s N_0. \tag{4}$$

It follows from relations (2) and Eq. (4) that $R_s \simeq 1.8 \times 10^{10} N_2$ immediately after the pumping pulse. This rate is comparable with the rate of $\simeq 2.5 \times 10^{10} N_2$ (Ref. 18) of the spontaneous radiative decay of the upper laser level. Nevertheless, the ionization caused by the slow electrons would hardly diminish the inversion significantly. Before a substantial number of Li atoms could be ionized by the slow electrons, almost all of these electrons would lose their energy through the collisional excitation of the ground-state Li atoms to the 1$s^2 2p$ level [process(c)]. Indeed, as a result of one such collision the energy of a slow electron decreases by $\simeq$1.8 eV to a level lower than the 5.4 eV necessary for Li 2s ionization. The cross section of this process $\sigma_{\text{exc}} \simeq 8.8 \times 10^{-15}$ cm^2 (Ref. 15) leads to the rate coefficient $X_{\text{exc}} \simeq 1.5 \times 10^{-6}$ cm^{-3} s^{-1} $\simeq 100 X_{\text{ion}}$. Therefore less than 1% of the slow electrons would have time to ionize Li atoms before losing their energy. We refer to the time $\tau_s \equiv (X_{\text{exc}} N_0)^{-1} \simeq 1.5$ ps as the lifetime of the slow electrons. Because of the relatively small difference in the energy of the slow and the ultraslow electrons, the lifetime of the ultraslow electrons would be of the same order of magnitude.

X_{ion} is an order of magnitude larger for the fast electrons than for the slow electrons because of both the larger cross section[17] and higher velocity of the fast electrons. But, owing to the relatively low density of the latter, their contribution to populating the lower laser level would be even smaller than that of the slow electrons. Hence we can essentially neglect the effect that the Li 2s ionization by electron impact has on the population inversion.

The population inversion for Li 19.9-nm lasing could also be destroyed by transitions from the upper laser level to levels other than the ionic ground level. The strongest transition of this kind is the transition down to the Li^+ 1$s2s$ $^1S_0$ level [with a transition energy of $\simeq$1.3 eV and an oscillator strength of $\simeq$0.2 (Ref. 18)]. The electron-impact excitation cross sections are[19] 1.5×10^{-16}, 2.4×10^{-16}, and 2×10^{-17} for the fast, slow, and ultraslow electrons, respectively, that is, an order of magnitude lower than the cross sections for the Li 2s–2p electron-impact excitation. Therefore this process is also of little significance for the population inversion. Then the radiative rate of the 1$s2p$ ^1P–1$s2s$ ^1S transition is two orders of magnitude smaller than that of the lasing transition. Finally, the cross section of the ionization process Li^+ 1$s2p$ $^1P \rightarrow Li^{2+}$ 1s by 19.9-nm radiation is as low as 2.5×10^{-21} cm^2,[20] so that reabsorption of the 19.9-nm radiation by the Li ions at the upper laser level would not decrease this level's population substantially.

Thus we may conclude that all the competing processes discussed would not strongly interfere with lasing, and we neglect the lower laser level population in the following estimates of gain. From the standard known expression for the small-signal gain coefficient on the central frequency of the lasing transition γ_0 in the presence of

Doppler broadening (see, e.g., Ref. 1), one can obtain the following equation:

$$\gamma_0 \simeq 0.025 g_1 g_2^{-1} f_{12} (\Delta\nu^D)^{-1} (N_2 - g_2 g_1^{-1} N_1), \quad (5)$$

where N_1 (g_1) and N_2 (g_2) are the population density (degeneracy) of the lower and the upper laser levels, respectively, and $\Delta\nu^D$ and f_{12} are the Doppler width and the oscillator strength of the lasing transition, respectively. We assume that $f_{12} \simeq 0.45$,[18] and $\Delta\nu^D \simeq 10^{11}$ s^{-1} for the Li vapor temperature $T \simeq 1000$ K. For N_2 we can write[21]

$$N_2 \simeq h^{-1} \tau_p \bar{J}\, \bar{\alpha}\, \bar{\eta} (\Delta\nu_p / \bar{\nu}). \quad (6)$$

Here the overbar indicates averaging over the pumping-frequency interval $\Delta\nu_p = (E' - E'')/h$, J is the intensity of the pumping radiation per unit frequency (spectral intensity), $\bar{\nu}$ is the average pumping frequency, and τ_p is the pumping-pulse duration. We assume the absorption quantum efficiency $\bar{\eta}$ to be equal to the ratio r of the number of the desirable inner-shell transitions to the number of all the absorbed photons. The average absorption coefficient $\bar{\alpha}$ can be estimated as[21]

$$\bar{\alpha} \simeq \bar{\sigma}_{\text{tot}} [(g'/g_0) N_0 - N']. \quad (7)$$

The density N' of the $2s$-excited or $2s$-ionized Li atoms may be neglected here, since we expect it to be much smaller than N_0. After photoionization the system Li^+ + photoelectron would be in the $^2P^\circ$ state[9] with degeneracy $g' = 6$. Then $\alpha \simeq 9$ cm^{-1} for the assumed Li vapor density of $N_0 = 5 \times 10^{17}$ cm^{-3}. The pumping pulse duration could be chosen as 0.1 of the upper laser level lifetime, $\tau_p \simeq 4$ ps, so that most of the excited ions would not decay during pumping. Relations (5) and (6) now yield:

$$\gamma_0 (\text{cm}^{-1}) \simeq 2.7 \times 10^{-8} I (\text{W/cm}^2) \bar{\alpha} (\text{cm}^{-1}). \quad (8)$$

Here $I \equiv J\Delta\nu_p$ denotes the intensity of the pumping pulse in the frequency interval $\Delta\nu_p \simeq 4.8 \times 10^{13}$ s^{-1} with the photon energy $h\bar{\nu} \simeq 71.1$ eV ($\Delta\nu_p/\bar{\nu} \simeq 2.8 \times 10^{-3}$). For $I = 4.3 \times 10^7$ W/cm^2 (such intensity is radiated in the frequency interval $\Delta\nu_p$ into the solid angle of 1 sr normally to the surface of a blackbody at the temperature of $\simeq$32 eV) one can obtain $\gamma_0 \simeq 10$ cm^{-1}.

The attainable gain–length product is estimated by us for the traveling-wave design.[1] Let the pumping pulse move along the lasing medium. The pulse duration is much shorter than the lifetime of the upper laser level, and we neglect the spontaneous decay when we estimate the space distribution of the population inversion density N_2. Also, since we neglect the influence of competing processes on the laser level population, the density N_2 [and therefore the small-signal gain coefficient $\gamma(z)$] decreases with the distance z from the starting point only because of the pumping-pulse absorption in Li vapor: $\gamma(z) = \gamma_0 \exp(-\bar{\alpha} z)$. The gain–length product can be defined as

$$G = \int_0^\infty \gamma(z) dz = \gamma_0 / \bar{\alpha} \simeq (2.7 \times 10^{-8}) I, \quad (9)$$

which turns out to be independent of the Li vapor density. Now, in order to attain $G = 10$, the pumping intensity of $\simeq 3.7 \times 10^8$ W/cm^2 is necessary. (Such intensity is radiated in the frequency interval $\Delta\nu_p$ into the solid angle of 1 sr normally to the surface of a blackbody at a temperature of $\simeq$105 eV.)

So far, we have been considered pumping by the $1s$ photoionization of Li atoms from their ground state $1s^2 2s$ only. For this process the probability that the ion will be left in the $1s2p$ configuration is significantly smaller than for the $1s2s$ configuration. Only in the vicinity of the $[1s(3s3p)\,^3P]\ ^2P^\circ$ resonance does this proportion change somewhat to favor 19.9-nm lasing. As a result such lasing would require narrow-band pumping. However, some recent experimental results for the soft-x-ray inner-shell ionization of another alkali, Na, allow us to suggest that the proposed 19.9-nm x-ray laser in Li may be efficiently pumped by photoionization of excited $1s^2 2p$ Li atoms with a short x-ray pulse of a much broader spectrum.

It was shown[22] that the distribution of Na^+ states after inner-shell $2p$ photoionization of the $2p^6 3p$-excited Na atoms is drastically different from that after $2p$ photoionization of the ground-state $2p^2 3s$ Na atoms, with basically the same total $2p$-photoionization cross section. While $2p$ photoionization of the ground-state $2p^6 3s$ Na atoms results in Na^+ ions predominantly in the $2p^5 3s$ configuration, $2p$ photoionization of the $2p^6 3p$-excited Na atoms populates primarily the $2p^5 3p$ configuration of the Na^+ ions, if the photon energy is large enough.

Unfortunately, to the best of our knowledge, no experimental data have been published for the distribution of the ionic states resulting from inner-shell photoionization of excited Li atoms. However, recently published calculations[23] show that, as for Na, the inner-shell photoionization of $1s^2 2p$-excited Li atoms by energetic enough photons results predominantly in the Li^+ $1s2p$ configuration and not in the $1s2s$ configuration. In particular, for the energy of the incident photons between the $1s2p$ and $1s3p$ thresholds (that is, between $\simeq$66 and $\simeq$75 eV), the pumping quantum efficiency $\eta \simeq r$ appears to be as large as 0.5. At the same time, the intensity of the collisional processes initiated by this broad-band pumping pulse is not expected to be substantially different from the intensity of the collisional processes following the narrow-band pumping considered previously. The energy spectrum of the emerging photoelectrons, being broader in the case of broadband pumping, would not be broad enough to change the rate coefficients substantially. As a result, the spectral intensity of the x rays required to pump the $1s^2 2p$-excited Li atoms would be lower by 2 orders of magnitude than that for pumping of the ground-level atoms.

Li atoms can be excited to the $1s^2 2p$ state by a resonantly tuned 670.8-nm dye laser.[12,24] Unfortunately, a side effect of such excitation is strong $2s$ ionization, so that only $\simeq$15% of all the Li atoms in a volume can be $2p$ excited before significant ionization occurs. In order to realize the advantages of pumping of excited atoms more fully, it would be desirable to excite a much larger fraction of the Li atoms, at least in the traveling-wave design.

It is worth noting that using excited Na atoms instead of the ground-state atoms in the Na laser proposed in Ref. 6 may result in much higher gain.

In conclusion, we have theoretically demonstrated the feasibility of 19.9-nm lasing on the Li^+ $1s2p\ ^1P \rightarrow 1s^2$ transition in Li vapor pumped by a short, noncoherent soft-x-ray pulse with a narrow-band spectrum centered near the $[1s(3s3p)\,^3P]\,^2P^\circ$ resonance of neutral Li ($\simeq$71 eV).

Such pumping allows one to overcome the detrimental effect of the emerging photoelectrons on the population inversion. Significant gain is expected at the state of the art of laser-plasma technology. The possibility exists of even more efficient broadband pumping of $1s^2 2p$-excited Li atoms.

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