4-1-1995

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PHYSICAL REVIEW A VOLUME 51, NUMBER 4 APRIL 1995

Dynamics of optical-field-ionized plasmas for x-ray lasers

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(Received 17 October 1994)

The success of recombination-pumped x-ray laser schemes is determined by the kinetics of ions in plasmas with relatively dense, cold-electron distributions. We examine how laser gain in such systems is affected by a multipeaked electron distribution generated by sequential ionization of atoms using high-intensity, ultrashort-pulse lasers. We also investigate the role of heating processes that modify electron energy distributions during the recombination and the accompanying collisional cascade. We find that conditions for the success of these schemes are critically modified by the inclusion of these effects.

PACS number(s): 32.80.Rm, 42.55.Vc, 52.40.Nk

Optical-field ionization (OFI) has recently been considered as a mechanism for the production of cold electrons that could drive a recombination-pumped x-ray laser [1]. For example, laser systems have been proposed in plasmas of aluminum [2], carbon [2], and neon [3]. Experimentally, gain has been reported in a lithium system [4]. A common feature of these systems is that they require electron-ion recombination to occur on a time scale that is short compared to the natural lifetime of the laser transition, thus requiring a plasma with high electron density n_e and cold-electron temperature T_e.

High-intensity, ultrashort-pulse lasers ionize atoms in a regime where the resulting free-electron energies can be predicted by a quasi-static-field, tunneling-ionization model discussed by Corkum and others [1,5,6]. Under such conditions, sequential OFI results in multiply charged ions and a multiply peaked electron energy distribution. The strong dependence on electron energy of the three-body, electron-ion recombination cross section makes the successful, high-gain operation of a recombination-pumped laser dependent on the presence of low-energy electrons [7–9]. OFI of weakly bound electrons yields such electrons; however, higher-energy electrons, generated by ionization of more tightly bound electrons, limit laser gain in two ways. First, high-energy electrons inhibit the cascade of the excited-state population to the upper laser level and, second, these electrons collisionally heat colder electrons.

Nagata *et al.* [4] recently reported gain at 13.5 nm on the Lyman-α transition of Li^{2+}. In this experiment, Li metal was ablated from a solid target using a relatively low-intensity laser pulse. The resulting plasma plume was then irradiated by a high-intensity, short-pulse laser, which generated Li^{3+} (see Fig. 1 of Ref. [4] for the experimental setup). Subsequent recombination of Li^{3+} with cold electrons was proposed as the pumping mechanism of the laser transition. In this Rapid Communication we examine dynamics in the Li system.

The fully ionized Li plasma will initially contain three distinguishable electron distributions: one "cold" and two "hot." Using a measurement of the time-integrated slope of the radiation continuum, the cold-electron distribution was estimated to have a temperature on the order of 1 eV [4]. This temperature is either the residual electron energy following hydrodynamic expansion of the ablated Li plasma plume or the average electron energy following multiphoton ionization of the outer-shell electron in Li during the relatively low-intensity leading edge of the short-pulse laser. This cold distribution is primarily responsible for the recombination that forms excited Li^{2+}. The hot-electron distributions result from sequential OFI of Li^{1+} and Li^{2+} by the high-intensity short-pulse laser. For example, using the quasi-static-field tunneling model [1,6] we calculate that the electron distributions will have temperatures T_e (OFI) of 25 and 140 eV, when produced by a 400-nm, linearly polarized, 125-fs pump pulse with a peak intensity of 2×10^{17} W/cm^2.

The analysis in this Rapid Communication is carried out using the atomic and plasma physics kinetics code FLY [10]. FLY solves rate equations to determine the time-dependent population of energy levels in the different ion species found in a plasma. In our case, the energy levels of the states with principle quantum numbers n (up to $n=10$) for Li, Li^{1+}, and $Li^{2}+$ are calculated. The results were found to be unaffacted

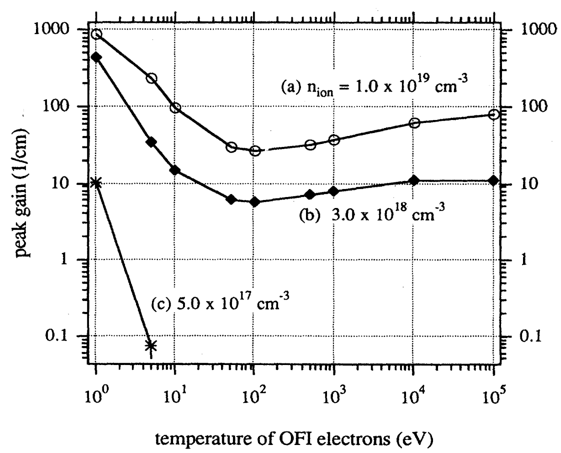

FIG. 1. Peak gain on the Ly-α transition in Li^{2+} as a function of the OFI electron temperature. The cold-electron temperature [T_e(cold)] is held constant at 1 eV and $\frac{1}{2}n_e(\text{OFI})=n_e(\text{cold})=n_{\text{ion}}$. (a) $n_{\text{ion}}=1.0\times10^{19}$ cm^{-3}; (b) $n_{\text{ion}}=3.0\times10^{18}$cm^{-3}; (c) $n_{\text{ion}}=5.0\times10^{17}$ cm^{-3}.

by increasing the number of energy levels in each ion species so that levels up to $n=20$ were included. Gain on the Ly-α transition is computed assuming linear Stark broadening of the gain cross section [2].

Experiments in Li [4,11] have been carried out in a regime of n_e and T_e, where cooling of the plasma filament will be determined by free-streaming electrons [2]; the actual cooling in this regime is, however, unimportant on the time scales relevant to achieving peak gain in the Li system. This can be understood by considering the time required for a Maxwellian distribution of electrons to cool to one-half of its initial temperature. Assuming free-streaming conditions and an electron energy flux out of the filament given by $(n_e k_B T_e v_e)/4$ [12], this time is approximately R/v_e, where R is the plasma filament radius, k_B is Boltzmann's constant, and v_e is the electron thermal velocity. The half-value cooling time is then defined by $(T_e/2)(dT_e/dt)^{-1}$. Plasma filament lengths of greater than 1 cm will be desirable in this system in order to observe gain (see below), which implies a focused beam waist of at least 25 μm for a 400-nm Gaussian pump pulse. These considerations yield half-value cooling times of 8 ps (for a temperature of 25 eV), 4 ps (140 eV), and 50 ps (1 eV). The resulting cooling of the electron distribution on this time scale will not significantly affect the peak gain generated in the Li plasma and is neglected in the subsequent analysis.

We first consider the effect of the OFI electrons on gain that can be achieved on the Ly-α transition. Figure 1 shows the calculated peak gain for $T_e(\text{cold})=1$ eV as a function of $T_e(\text{OFI})$ for a variety of ion densities, n_{ion}. We assume for simplicity that both OFI electrons have the same temperature and vary this $T_e(\text{OFI})$ from 1 to 100 keV. The initial total electron density n_e is determined by (i) charge neutrality (thus $n_e=\Sigma Z_i n^i_{\text{ion}}$ where Z_i is the ion charge and n^i_{ion} is the ion density of charge state i), (ii) the assumption that $\frac{1}{3}$ of the total n_e is represented by the $T_e(\text{cold})$, and (iii) the requirement that the gain of the system be observable (>10 cm^{-1}). This required gain value is set by the length of the pumped medium as determined by the maximum confocal parameter of currently available short-pulse lasers focused to the power density required to fully ionize Li (typically ≤ 1 cm) [4,11]. These conditions strongly suggest that the ion densities used in this analysis be higher than those reported in Ref. [4]. In Fig. 1 we have bracketed the parameter space of interest in the Li system.

Three regions of interest can be discerned in Fig. 1: the relatively high gain regions at the lowest and highest OFI electron temperatures and a region of lower gain near temperatures of 100 eV. The three-body electron-ion recombination rate (for a single electron distribution) summed over all levels is given by [9]

$$R_{\text{three-body}}\ (\text{sec}^{-1})=\frac{1.8\times 10^{-8} Z^3 n_e^2 \ln(\Lambda)}{T_e^{9/2}}, \qquad (1)$$

where T_e (K) is the electron temperature, Z is the ion charge state, n_e (cm^{-3}) is the electron density, and $\ln(\Lambda)$ is the Coulomb logarithm. At low OFI electron temperatures, recombination will be enhanced and rapid collisional cascade deposits population in the lowest excited state (and upper laser state), $n=2$ in Li^{2+} [8]. Cold OFI electron temperatures therefore generate a high peak value of gain. High values of $T_e(\text{OFI})$ also result in relatively large gain, since the OFI electrons essentially decouple from the system; at high temperatures the cross sections for electron-ion collisions that deplete upper laser state populations decrease (such depletion mechanisms are discussed below). For fixed ion density the reduction in gain from the low $T_e(\text{OFI})$ case to the high $T_e(\text{OFI})$ case results solely from the reduction in the effective cold-electron density. It should be noted that peak gain does not scale linearly with $n_e(\text{cold})$ because the recombination rate depends nonlinearly on this density.

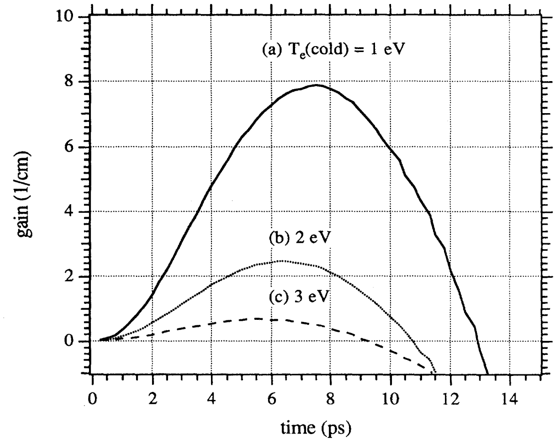

FIG. 2. Gain as a function of time for various fixed $T_e(\text{cold})$, $n_{\text{ion}}=3.0\times 10^{18}$ cm^{-3}, $n_e(\text{OFI})=6.0\times 10^{18}$ cm^{-3}, $T_e(\text{OFI})=1$ keV, and $n_e(\text{cold})=3.0\times 10^{18}$ cm^{-3}. The curves represent (a) $T_e(\text{cold})=1$ eV; (b) $T_e(\text{cold})=2$ eV; (c) $T_e(\text{cold})=3$ eV.

In the region of $T_e(\text{OFI})$ of approximately 100 eV the OFI electrons will reduce three-body recombination (compared to colder temperatures). Additionally, they retard the collisional cascade of population to $n=2$, since in this range of $T_e(\text{OFI})$, intralevel collisions tend to establish a distribution within the entire manifold of excited Li^{2+} states; in the steady state, this distribution would become a Boltzmann distribution characterized by $T_e(\text{OFI})$. Thus gain is reduced as compared with the case of colder or hotter OFI electrons.

Figure 1 also illustrates the strong dependence of gain on ion density. As the ion density drops from (a) 1.0×10^{19} cm^{-3} to (b) 3.0×10^{18} cm^{-3} to (c) 5.0×10^{17} cm^{-3}, reduced gain results from the decreased three-body and collisional deexcitation rates at lower electron densities, and from the decreased number of ions radiating. Assuming realistic $T_e(\text{OFI})$ and neglecting additional heating effects discussed below, we conclude that ion densities of greater than 10^{18} cm^{-3} will be required for gain in this system. Other considerations will become important at these electron densities; for example, ionization defocusing of the pump-laser pulse limits the length of the generated plasma [13].

Gain in recombination-pumped systems decreases sharply with increasing T_e. This gain reduction can only be compensated by increasing the ion density in order to maintain fast recombination and collisional deexcitation. The dependence of gain on T_e in the Li system is illustrated in Fig. 2, where $T_e(\text{cold})$ is varied from 1 to 3 eV. Electron heating in such an energy range may result from a variety of processes. First electron-electron collisions will occur and tend to equilibrate the cold and hot (OFI) parts of the total electron distribution. Spitzer [14] gives the electron-electron equilibration time, t_{eq}, between two distributions described by temperatures and densities, T_1, n_{e1} and T_2, n_{e2},

$$t_{eq}\ (\text{sec}) = 0.137 \frac{(T_1+T_2)^{3/2}}{(n_{e1}+n_{e2})\ln(\Lambda)} . \tag{2}$$

For a Li plasma where $n_e(\text{cold}) = 1\times10^{19}$ cm^{-3}, $T_e(\text{cold}) = 1$ eV, $n_e(\text{OFI}) = 2\times10^{19}$ cm^{-3}, and $T_e(\text{OFI}) = 25$ eV, Eq. (2) implies a cold-electron heating rate of approximately 60 eV/ps at early times. The time scale for electron heating is therefore rapid relative to the time scale for the buildup of gain (about 10 ps for these conditions), and thus electron temperature equilibration results in a severe reduction of gain under these conditions. This problem cannot be easily avoided by generating hotter OFI electrons because the cold-electron heating rate depends weakly on the hot-electron temperature $\sim T_e^{1/2}(\text{OFI})$. For example, by using circularly polarized light to ionize the plasma we can generate $T_e(\text{OFI}) = 1$ keV; under these conditions the cold-electron heating rate is still high, approximately 20 eV/ps.

It is possible, however, that the simple thermalization model above [Eq. (2)] may not be appropriate for this system. The electron distribution generated by a high-intensity, ultrashort-pulse pump laser is not Maxwellian, nor is the subdistribution that results from any particular ionization stage [1,15,16]. In fact, experiments suggest that the time required for an OFI generated distribution to relax into a Maxwellian distribution is longer than would be predicted by Eq. (2) [15]. To demonstrate convincingly that even under this scenario high gain will not persist, we do not limit ourselves to thermalization arguments. We will now show that even if no intradistribution cross relaxation exists (therefore assuming that the cold-electron distribution does not heat), gain in the Li system is much smaller than previously predicted.

Electron heating in recombination-pumped laser systems also results from electron-ion collisional processes. Energy conservation requires that three-body recombination and the subsequent collisional cascade of electrons through bound states result in heating of the residual free electrons. We calculate the effects of such heating in the Li system using a time-dependent kinetics calculation to track the evolution of the population of the Li^{2+} energy levels. We assume that Li^{2+} is generated solely through three-body recombination and that population transfer between levels is purely collisional (except for decay on the Ly-α transition, which we assume to be radiative) [8,9]. The time history of the cold, free-electron bath temperature $T_e(t)$ is given by

$$T_e(t) = \frac{T_e(0) + \frac{2}{3}\sum_i \alpha_i(t) E_i}{1 - \sum_i \alpha_i(t)} , \tag{3}$$

where $\alpha_i(t)$ is the number of ions in the ith energy level at time t, normalized to the cold-electron density, and E_i is binding energy of the ith energy level. Since the dominant process for recombination involves three bodies and occurs primarily to the upper ionic states, the energy absorbed by the electron bath per electron capture is small. Energy will therefore be preferentially incorporated into the low-energy part of the electron distribution [see Eq. (2)] and will rapidly thermalize. Similarly, energy gained through collisional de-excitation of excited-state ions will be rapidly thermalized among the low-energy electrons.

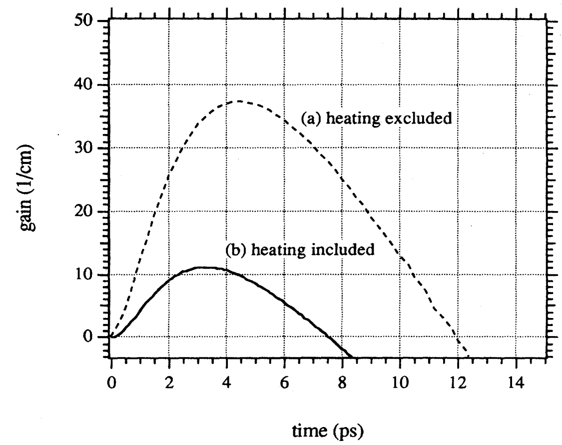

FIG. 3. Gain as a function of time with $n_{ion} = 1\times10^{19}$ cm^{-3}, $n_e(\text{cold}) = 1\times10^{19}$ cm^{-3}, $T_e(\text{cold}) = 1$ eV, $n_e(\text{OFI}) = 2\times10^{19}$ cm^{-3}, and $T_e(\text{OFI}) = 1$ keV for (a) no electron heating and (b) recombination and collisional electron heating included. Equilibration between the cold- and hot-electron distributions is ignored.

Figure 3 shows gain as a function of time for the initial conditions of $n_{ion} = 1\times10^{19}$ cm^{-3}, $T_e(\text{cold}) = 1$ eV, and $T_e(\text{OFI}) = 1$ keV. Although the collisional effects of hot OFI electrons on ion populations are included, we note again that here we have ignored cold-electron equilibration. Gain is indicated for simulations (a), without, and (b), with, collisional heating; by observation, collisional heating effects reduce the peak gain by a factor of 3.

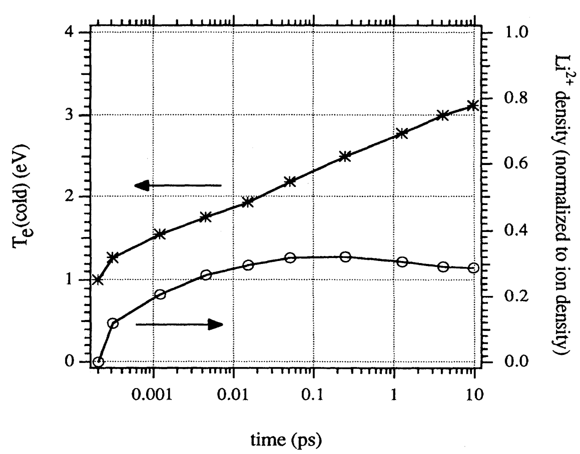

FIG. 4. $T_e(\text{cold})$ and fractional population of the recombined, H-like species, as functions of time. Conditions are the same as in Fig. 3(b).

In Fig. 4 we show the time history of the low-energy free-electron temperature, and the population of Li^{2+} ions normalized to the initial, total ion density. At early times, electron heating is dominated by the three-body recombination process, consistent with the rapid increase in Li^{2+} population. This recombination heating causes the temperature of the low-energy electrons to double in less than 100 fs and, additionally, recombination to decrease dramatically, due to the $T_e^{-9/2}$ dependence of the recombination rate. Following initial recombination heating, collisional deexcitation of ions begins to dominate electron heating. Because the electron temperature rises extremely rapidly, gain is insensitive to the actual initial cold-electron temperature; even with the coldest initial temperatures, heating processes rapidly bring the electron bath to several eV. This heating results in an inability to produce high gain in the Li system when it is operated at low ion density ($<10^{19}$ cm^{-3}).

In conclusion, we find that collisional processes due to OFI generated electrons, electron energy equilibration within a doubly peaked distribution, and collisional heating of low-energy electrons play important roles in limiting the gain of the Li recombination-pumped x-ray laser system. The results also imply that the Li system must be studied under higher density conditions than previously considered.

The authors wish to acknowledge helpful discussions with Ernie Glover, David Eder, Martin Hofer, and Luiz Da Silva. This work was supported by the U.S. Air Force Office of Scientific Research and through a collaboration with Lawrence Livermore National Laboratory under Contract No. W-7405-ENG-48.

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