Stimulated Free-Bound Emission from X-Ray Laser-Interaction Plasmas

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Nonlinear bound-free x-ray-absorption processes in an intense monochromatic x-ray radiation field such as an x-ray laser are investigated by using a collisional-radiative model. Above a threshold of the x-ray intensity, gain of stimulated free-bound emission is induced by strong photoionization. The wavelength of the stimulated free-bound emission is longer than that of the incident x rays, leading to a down-conversion of the frequency.

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The output power of soft-x-ray lasers pumped by optical lasers has been increasing over the last few years. A brightness above 10^{14} W/cm^{2}sr is now available in a saturated x-ray laser with a double-pass cavity [1]. Focusing of such an intense x-ray laser will allow the study of laser-matter interactions at specific x-ray wavelengths at x-ray intensities never achieved before in the laboratory. In particular, nonlinear phenomena at x-ray wavelengths should be of special interest in plasmas with a strong x-ray radiation field, similar to the nonlinear optical phenomena found with high-power optical lasers. Nonlinear absorption of x-ray radiation will be one of the first fields of study with intense x-ray sources.

In this Letter, x-ray-absorption processes in x-ray laser-interaction plasmas are investigated by using a collisional-radiative model. Above a threshold of the incident x-ray laser intensity, bound-free absorption over an ionization energy gap less than the incident x-ray photon energy inverts to gain. Gain on free-bound transitions by stimulated recombination has been discussed previously in Ref. [2], where analytical estimates indicated that a strong nonthermal equilibrium condition with a sufficient electron density was essential to create the gain. This Letter predicts numerically the existence of gain on free-bound transitions in x-ray laser interaction plasmas for the first time.

Photoabsorption by bound-free transitions depends on the populations of both lower (N_1) and upper (N_2) charge states through photoionization ($N_1 I_x \sigma$) and stimulated recombination ($aN_2 I_x \sigma$). The populations of each ionization stage in a radiation field I_x are related as

$$\frac{dN_1}{dt} = \Gamma_1(N_1^0 - N_1) - (N_1 - aN_2)I_x\sigma \tag{1}$$

and

$$\frac{dN_2}{dt} = \Gamma_2(N_2^0 - N_2) + (N_1 - aN_2)I_x\sigma \tag{2}$$

with a quasi-two-level absorption model [3], where Γ_i is a relaxation rate and N_i^0 an initial population for each ionization stage i. Γ_i includes all collisional and radiative transitions from the i stage except those with a radiation field. σ is a photoabsorption cross section and a a function of stimulated recombination. In a steady state with a pump radiation field of $h\nu_p$, a bound-free absorption coefficient $\sigma\Delta N_d$ of photons with an energy of $h\nu_a$ is obtained from Eqs. (1) and (2) as

$$\sigma\Delta N_d = \sigma[N_1 - a(h\nu_a)N_2] = \sigma\frac{\Gamma_1\Gamma_2\Delta N_d^0 + [a(h\nu_p) - a(h\nu_a)](\Gamma_1 N_1^0 + \Gamma_2 N_2^0)I_x\sigma}{\Gamma_1\Gamma_2 + [a(h\nu_p)\Gamma_1 + \Gamma_2]I_x\sigma} \tag{3}$$

where $\Delta N_d^0 = N_1^0 - a(h\nu_a)N_2^0$. $a(h\nu_p)$ is a stimulated recombination function for the incident photon $h\nu_p$ and $a(h\nu_a)$ a function due to stimulated recombination for photons of $h\nu_a$ to satisfy continuum spectra of bound-free absorption. $a(h\nu_a) > a(h\nu_p)$ at a condition of absorption with an ionization energy gap less than a photon energy of the incident radiation, $h\nu_a < h\nu_p$, and $a(h\nu_a) < a(h\nu_p)$ at $h\nu_a > h\nu_p$. Equation (3) for $a(h\nu_a) = a(h\nu_p)$ represents an absorption coefficient for the incident radiation. In the strong radiation field ($I_x \to \infty$), the absorption coefficient of the incident radiation is close to zero, i.e., $\sigma\Delta N_d^0 \to 0$, which is a well-known formula of saturated absorption. Equation (3) indicates that bound-free absorption for $h\nu_a < h\nu_p$ can invert to negative (gain) due to $a(h\nu_a) > a(h\nu_p)$ at incident x-ray intensities above the threshold $I_{\rm th}$ given by

$$I_{\rm th} = \frac{\Gamma_1\Gamma_2\Delta N_d^0}{[a(h\nu_a) - a(h\nu_p)](\Gamma_1 N_1^0 + \Gamma_2 N_2^0)\sigma}. \tag{4}$$

The gain on the free-bound transitions [SFBE gain (stimulated free-bound emission gain)] will be induced by photoionization in a strong radiation field. The photon energy of the SFBE is lower than the incident x-ray photon energy, i.e., there is a down-conversion of the frequency.

A steady-state collisional-radiative model is used [4] to investigate the atomic process in x-ray laser-interaction plasmas. The radiation field effect is modeled by considering photoionization and stimulated recombination. All the ionization stages of fully stripped to neutral ions are treated and the excited states of H-like to Li-like are included. The model considers important atomic processes: spontaneous radiative transition, collisional excitation, and the collisional deexcitation for bound-bound transition including autoionization levels, autoionizing and electron capture into doubly excited state, collisional ionization and three-body recombination, and photoionization, spontaneous recombination, and stimulated recombination. In this model, each excited level is coupled to the ground state of the next ionization stage (atomic processes of the ionization from excited states and recombination from ground states). However, recombination from excited states and ionization to excited states are neglected.

Free-bound radiative transitions in x-ray laser-interaction plasmas are determined by photoionization and radiative recombination taking account of stimulated recombination [5]. The number of photoionizations in time dt on the frequency range $(\nu,\nu+d\nu)$ is given by

$$N_1 P_\nu d\nu dt = N_1 (U_\nu / h\nu) c \sigma_{pn}(\nu) d\nu dt\,, \tag{5}$$

where $\sigma_{pn}(\nu)$ is the cross section for photoionization from nth excited atomic level. U_ν is a radiant energy per unit volume of the incident x-ray laser and c is the speed of light. Photons with an energy larger than ionization potential $(h\nu > \chi)$ participate in the absorption. The number of radiative recombinations is described by

$$N_2 N_e (R_{\rm spo} + R_{\rm stim}) d\nu dt = N_2 N_e V f(V) \sigma_{cn}(V) (1 + c^3 U_\nu / 8\pi h \nu^3) dV dt\,, \tag{6}$$

where $R_{\rm spo}$ corresponds to spontaneous radiative recombination and $R_{\rm stim}$ stimulated recombination. σ_{cn} is the cross section of electron capture and $f(v)$ an electron distribution function. A photon energy of emission and absorption is related with a free-electron energy $\frac{1}{2} m V^2$ and the ionization potential χ as $h\nu = \frac{1}{2} m V^2 + \chi$. From Eqs. (5) and (6) using the principle of detailed balance, we obtain a relationship (Einstein-Milne relation) between the cross sections for photoionization and radiative recombination as

$$\sigma_{\nu n}(\nu) = \frac{g_+}{g_n} \frac{m_e^2 V^2 c^2}{(h\nu)^2} \sigma_{cn}(V)\,, \tag{7}$$

for the nth excited atomic level, where g is a statistical weight [5]. The radiative recombination and photoionization rate coefficients are calculated from Eqs. (5)-(7) for a Maxwellian electron distribution satisfying the principle of detail balance. The Kramers cross section $\sigma_{cn}(v)$ is used for radiative electron capture, the cross section of which is corrected with a radiative bound-free Gaunt factor [6]. The effective charge state in the Kramers formula is given by a hydrogenic screening model [7]. The photoionization cross section $\sigma_{pn}(\nu)$ is calculated using Eq. (7) and the cross section $\sigma_{cn}(v)$.

A total photoabsorption coefficient κ is calculated for bound-free $\kappa_{\rm bf}$ and free-free $\kappa_{\rm ff}$ transitions, i.e., $\kappa = \sum (\kappa_{\rm bf} + \kappa_{\rm ff})$. Photon scattering and bound-bound absorption are ignored because of its small cross section and the coincidence problem of photon energies between the incident and the absorption. The absorption coefficient for bound-free transitions is given by

$$\kappa_{\rm bf}(\nu) = -\frac{1}{I_\nu}\frac{dI_\nu}{dx} = \sum_n \sigma_{\nu n} \left(N_1 - N_2 N_e(V) \frac{h^3}{8\pi m_e^3 V^2} \frac{g_n}{g_+} \right) = \sum_n \sigma_{\nu n} \left[N_1 - N_2 N_e \frac{h^3}{2(2\pi m_e kT)^{3/2}} \exp\left(-\frac{\varepsilon}{kT} \right) \frac{g_n}{g_+} \right], \tag{8}$$

for a Maxwellian electron distribution. kT is an electron temperature and ε is a free electron energy ($\varepsilon = \frac{1}{2} m V^2 = h\nu - \chi$). No strong effect of photoionization and stimulated recombination on the electron distribution is assumed in the steady-state model. The free-free electron collisional rates are 10^{14}–10^{15} sec^{-1} at electron densities of 10^{21}–10^{22} cm^{-3} and a temperature of 5 eV, which are larger than the photoionization rates in the carbon plasmas with a 23-nm laser intensity less than 10^{14}–10^{15} W/cm^2. At a lower electron density and/or higher laser intensity, however, the non-Maxwellian effect on the electron distribution must be taken into account. The free-free absorption coefficient is given by

$$\kappa_{\rm ff}(\nu) = \frac{4}{3} \left(\frac{2\pi}{3 m_e kT} \right)^{1/2} \frac{Z^{*3} e^6 N_i^2}{c m_e h \nu^3} \left[1 - \exp\left(\frac{h\nu}{kT} \right) \right] g_{\rm ff}\,, \tag{9}$$

where $g_{\rm ff}$ is a free-free transition Gaunt factor [6] and Z^* the average ionization stage. The LTE (local thermal equilibrium) condition is assumed for free electrons because of the short time relaxation of free-free electrons, i.e., the assumption of a Maxwellian electron distribution.

Figure 1 shows the spectra of x-ray-absorption coefficient (45–60 eV) in carbon plasmas with a fixed electron temperature of 5 eV and density of 2×10^{21} cm^{-3} as a function of incident x-ray laser intensities. The assumed wavelength of the incident x-ray laser is 23 nm (54 eV, e.g., Ne-like Ge x-ray laser) corresponding to a photon energy higher than the photoabsorption edges of Be-like, B-like, and neutral carbon ions. The absorption spectra change and the coefficient decreases with the x-ray laser intensity. The absorption coefficient for photons of 48 to 53 eV inverts to negative (gain) above a threshold of the incident x-ray laser intensity ($>10^{13}$ W/cm^2). The gain on free-bound transitions (SFBE gain) is induced by photoionization in the strong radiation field. The absorption coefficient for the incident x ray can never change to gain because of the principle of detailed balance. The simple model given by Eqs. (1) and (2) describes the dependence of the SFBE gain coefficient at a condition of nearly saturated absorption $[N_1 \cong a(h\nu_p)N_2]$ as

$$\lim_{I_x \to \infty} \sigma[a(h\nu_a)N_2 - N_1] = \sigma[a(h\nu_a) - a(h\nu_p)]N_2$$

$$\sim \sigma N_2 N_e kT^{-3/2}\left[\exp\left(-\frac{h\nu_a - \chi}{kT}\right) - \exp\left(-\frac{h\nu_p - \chi}{kT}\right)\right], \tag{10}$$

where $\chi < h\nu_a < h\nu_p$. A larger gain coefficient is created for lower photon energy from Eq. (10), the dependence of which is consistent with the numerical calculations from the collisional-radiative model as shown in Fig. 1.

The electron density dependence of the SFBE gain coefficient is shown in Fig. 2 as a function of the incident x-ray laser intensity. The gain coefficient increases nonlinearly with plasma density. Such a nonlinear increase is attributed to the dependence of stimulated recombination on both free-electron (N_e) and ion (N_2) densities as represented by Eq. (8) or (10). The intensity threshold also increases with the electron density. Large relaxation rates (Γ_i) due to three-body recombination in high-density plasmas augment the intensity threshold (I_{th}) as explained by Eq. (4). The initial population ratio ΔN_d^0, which is also one of the factors to decide the threshold as described by Eq. (4), increases with the electron density due to three-body recombination, resulting in increase of the threshold. The dashed lines at high laser intensities are calculations under a condition that the free-electron collisional rate is larger than the photoionization, where a non-Maxwellian effect must be taken into account on the electron distribution.

Figure 3 shows the SFBE gain coefficient of a 25.8-nm x ray as a function of incident x-ray wavelength in the carbon plasma. The solid line is an effective gain coefficient due to both free-bound and free-free transitions. The dashed line shows a gain coefficient for free-bound transitions from Li-like to Be-like ions where the population inversion occurs mainly. The gain coefficient increases with a photon energy of the incident x rays at energies less than 56.5 eV. The dependence given by Eq. (10) explains the increase of $\sigma[a(h\nu_a)N_2 - N_1]$ with photon energy $h\nu_p$ of the incident x rays. Increasing the photon energy more than 56.5 eV causes edges to appear in the gain spectra. The higher energy photons can also ionize the Li-like ions (upper state), causing the population of the upper state to be reduced either directly from the ground level or indirectly through the excited level of Li-like ions. Edges for incident photon energies of 56.5 and 64.5 eV correspond to the ionization potentials of $1s^22p$ and $1s^22s$ in Li-like ions, respectively.

X-ray laser-interaction plasmas may be useful for pro-

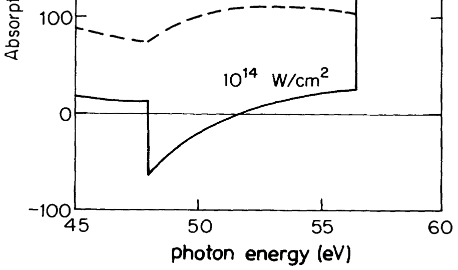

FIG. 1. Absorption spectra (45–60 eV) in carbon plasmas at $T_e = 5$ eV and $N_e = 2\times10^{21}$ cm^{-3} as a function of the 23 nm (54 eV) x-ray laser intensity.

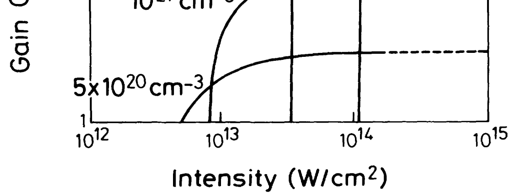

FIG. 2. Electron density dependence of the SFBE gain coefficient for 25.8-nm x rays as a function of the incident x-ray (23 nm) laser intensity in the carbon plasmas. Dashed lines are gain coefficients where the assumption of a Maxwellian free-electron distribution will fail.

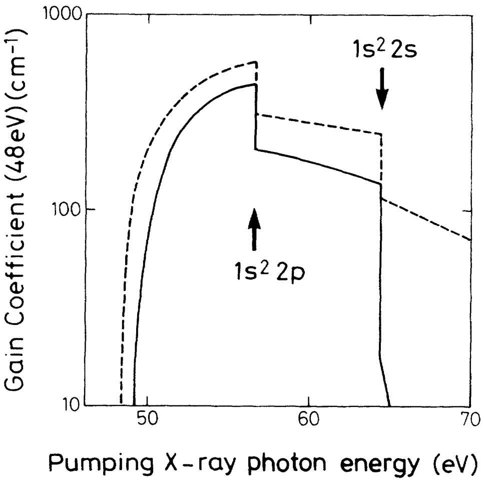

FIG. 3. Spectra of the SFBE gain coefficient for 25.8-nm x rays as a function of the incident x-ray laser wavelength (photon energy) at a fixed x-ray laser intensity of 3×10^{14} W/cm^2 in carbon plasmas ($N_e=5\times10^{21}$ cm^{-3} and $T_e=5$ eV). The solid line is the total gain coefficient and the dashed line the gain coefficient of Li-like ions.

ducing free-bound lasers (FBL) which have the potential of tunability [2]. Detailed analysis using a time-dependent atomic model coupled with a hydrodynamic code will be necessary to evaluate the practicability of FBL using x-ray laser-interaction plasmas. In this Letter, a novel nonlinear process in an intense monochromatic x-ray radiation field, which occurs on bound-free transitions, is examined by using a steady-state collisional-radiative model for the first time. Above a threshold of the incident x-ray intensity, bound-free absorption can change to gain with photon energies between the incident x rays and ionization limit. The frequency of the incident x rays can be down-converted in plasmas with the strong radiative field through the SFBE process.

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