Gain on Free-Bound Transitions by Stimulated Radiative Recombination

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This paper investigates the possibility of observing gain on a free-bound transition, i.e., by stimulated radiative recombination. The basic equations are derived from the Einstein-Milne relations for the continuum. Gain formulas for various plasma conditions are given. A continuously tunable short-wavelength laser seems to be possible.

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The intensive search for new lasers and the challenge to extend the region of known laser lines into the uv and soft x-ray regime has led to the investigation of high-density plasmas as possible laser media. Plasma lasers emitting in the infrared, visible, and uv have been reported,[1–4] and recently, substantial gain in the soft x-ray region has been demonstrated in plasmas produced by high-power laser radiation.[5,6] All of these experiments involve generation of inversion between bound states of an ionized species in the plasma. The purpose of this Letter is to show that conditions can be found under which there is gain at a free-bound transition, i.e., a transition the upper level of which lies in the ionization continuum of the lasing species. The recombination continuum originating from transitions of this kind produces a large part of the continuum emission of high-density plasmas.[7] It is clear that to achieve gain at such a transition, the plasma must be far from Saha-Boltzmann equilibrium. As shown in the main part of the paper, several possibilities of such nonequilibrium exist, e.g., if the ions are highly overstripped or if the electron velocity distribution is non-Maxwellian.

Aside from its scientific interest, a free-bound laser could provide a number of advantagous features: The energy stored in the free-electron gas of a plasma would be directly extracted by the laser pulse, laser operation at rather short wavelengths would be possible, and, last but not least, such a laser would be tunable since the upper level is a continuum of states.

The principle of the laser involves stimulated transitions between two energy levels of a system. Except in the free-electron laser, such energy states are represented by discrete levels of an atom, ion, or molecule. The basic equations governing radiative transitions between such states are the Einstein relations which relate the coefficients of spontaneous emission, absorption, and stimulated emission to each other. To extend the laser principle to free-bound transitions, the basic relations between the rates of spontaneous radiative recombination, photoionization, and stimulated radiative recombination must be applied. They can be derived by analogy with the Einstein radiation theory and are known as the Einstein-Milne relations for the continuum.[8]

Let the number of photoionizations per unit volume in time dt and in the frequency range $(\nu, \nu+d\nu)$ be

$$N_0 p_\nu I_\nu \, d\nu \, dt, \tag{1}$$

where N_0 is the number density of the atoms, p_ν is the probability of photoionization of an atom per unit intensity, and $I_\nu \, d\nu$ is the intensity for the radiation in this frequency range. Furthermore, let the number of radiative recombination by electrons in the velocity range $(v, v+dv)$ be

$$N_1 n_e(v)[F(v)+G(v)I_\nu] v \, dv \, dt, \tag{2}$$

where N_1 is the number density of the ions, $n_e(v)$ is the distribution function of the electrons, and $F(v)$ and $G(v)$ are the coefficients for spontaneous and stimulated recombinations, respectively.

By postulating that the radiation of a system in Saha-Boltzmann equilibrium obeys the Planck radiation law, we obtain the Einstein-Milne relations, which relate $F(v)$, P_ν, and $G(v)$ to each other:

$$F(v) = (2h\nu^3/c^2)G(v), \tag{3}$$

$$p_\nu = (8\pi m^2 v^2 g_1/h^2 g_0)G(v), \tag{4}$$

where m is the electron mass and g_1 and g_0 are the degeneracies of the ion ground level and the final atom level. Equations (3) and (4) are the continuum analog of the Einstein relations for emission and absorption of radiation. They are valid even under nonequilibrium conditions and can be used to derive the absorption coefficient of a plasma by photoionization, corrected for stimulated emission. Using

$$h\nu = mv^2/2 + E_b, \tag{5}$$

where E_b is the binding energy of the lower level, one obtains

$$v \, dv = (h/m) d\nu \tag{6}$$

and, finally, for the absorption coefficient,

$$\kappa_\nu = -\frac{1}{I_\nu}\frac{dI_\nu}{dx} = p_\nu h_\nu \left[N_0 - \frac{N_1 n_e(v) h G(v)}{m p_\nu} \right]. \tag{7}$$

There is gain instead of absorption if the second term in the bracket dominates. Reversing the order of the term and inserting for $G(v)$ from Eq. (4), one obtains for the gain

$$\alpha_\nu = \sigma_\nu \left[N_1 n_e(v) \frac{h^3}{8\pi m^3 v^2} \frac{g_0}{g_1} - N_0 \right], \quad (8)$$

where $p_\nu h_\nu$ has been replaced by σ_ν the photoionization cross section.

Equation (8) is the free-bound analog of the usual gain equation for a laser with the inversion replaced by the term in the bracket. Note that the lower level of the transition is not necessarily the ground state of the atom and therefore N_0 may be the number density of any level of the atom. Equation (8) is also valid for ionization stages higher than one. In this case N_1 is the number density of ions in, say, the ionization stage k, and N_0 the respective density in the ionization stage k-1. To simplify the terminology in the following, the two species will still be called ions and atoms.

For the gain to be positive, a situation very far from thermal equilibrium is required. The electron and ion densities must be high but the population of the lower level must be much smaller than in Saha-Boltzmann equilibrium. Depending on the distribution of the electron gas in phase space, three cases can be defined:

(a) *The electrons have a Maxwellian velocity distribution.*—In this case $n_e(v)/v^2$ is a maximum for $v=0$. The gain is maximized if the upper level is just the ionization limit of the atom and is given by

$$\alpha_\nu = \sigma_\nu \left[N_1 N_e \frac{h^3}{8\pi m^3 \pi^{1/2} (2kT_e/m)^{3/2}} \frac{g_0}{g_1} - N_0 \right], \quad (9)$$

where N_e is the number density of the electrons and T_e is the electron temperature. One has numerically in mixed units

$$\alpha_\nu = \sigma_\nu [1.66 \times 10^{-22} N_1 N_e (kT_e)^{-3/2} g_0/g_1 - N_0], \quad (10)$$

where α_ν is in inverse centimeters, σ_ν is in centimeters squared, N_1, N_e, and N_0 are in inverse centimeters cubed, and kT_e is the electron temperature in electronvolts.

(b) *The electrons have a narrow velocity distribution around v_0.*—With the velocity distribution approximated by a "rectangular" one so that $n_e(v) = n_e$ for $v_0 - \Delta v/2 < v < v_0 + \Delta v/2$ and 0 elsewhere, the number density of free electrons is

$$N_e = n_e \Delta v. \quad (11)$$

Using the relations

$$E_{\rm el} = m v_0^2/2 \text{ and } \Delta E_{\rm el} = m v_0 \Delta v \quad (12)$$

for the energy and the energy spread of the electrons, respectively, one obtains for the gain

$$\alpha_\nu = \sigma_\nu \left[N_1 N_e \frac{h^3}{8(2\pi)^{1/2} m^{3/2} \Delta E_{\rm el}^{1/2}} \frac{g_0}{g_1} - N_0 \right], \quad (13)$$

or with the same units as in Eq. (10),

$$\alpha_\nu = \sigma_\nu (1.26 \times 10^{-22} N_1 N_e \Delta E_{\rm el}^{-1} E_{\rm el}^{-1/2} g_0/g_1 - N_0), \quad (14)$$

where $E_{\rm el}$ and $\Delta E_{\rm el}$ are in electronvolts.

Equation (13) can also be derived directly by writing for the gain

$$\alpha_\nu = \sigma_\nu [N_e g_0/g_e - N_0], \quad (15)$$

where g_0 and g_e are the degeneracies of the lower (bound) level and of the electron gas, respectively. The degeneracy of an electron gas in the presence of ions with number density N_1 is given by[9]

$$g_e = \frac{4g_1}{N_1} \left(\frac{E_{\rm el}}{\pi} \right)^{1/2} \left(\frac{m}{2\pi\hbar} \right)^{3/2} \Delta E_{\rm el}. \quad (16)$$

In this expression a factor of 2 is included, which takes care of the two opposite directions of propagation of an electron with respect to the photon propagation vector. Inserting Eq. (16) into Eq. (15) leads to the gain formula of Eq. (13), as required.

(c) *The electrons not only have a narrow velocity distribution but also have high directionality.*—This occurs, for example, in an electron beam. In this case the electron distribution is highly peaked in phase space and their degeneracy g_e is very low. The smallest possible value of g_e is unity, and therefore the highest possible gain is given by

$$\alpha_\nu = \sigma_\nu (g_0 N_e - N_0). \quad (17)$$

The condition of unity degeneracy determines from Eq. (16) how narrow the distribution of electron velocities and directions for a given electron energy and ion density has to be.

Consider a plasma consisting of completely stripped ions of charge Z and density N_1 in an electron gas of density ZN_1. The electrons are assumed to have a Maxwellian velocity distribution and therefore maximum gain occurs from the ionization limit into a hydrogenlike state of quantum number n. The lowering of the ionization limit in a plasma is neglected for this estimate and the frequency of the emitted radiation is given by

$$h\nu = \mathscr{R} Z^2/n^2, \quad (18)$$

where $\mathscr{R}$ is the Rydberg constant. An approximate value of the photoionization cross section can be obtained from the Kramers formula with unity Gaunt factor[10]:

$$\sigma_\nu = 2.76 \times 10^{29} Z^4/\nu^3 n^5 = 7.76 \times 10^{-18} n/Z^2. \quad (19)$$

Inserting this into Eq. (10), with $g_0/g_1 = 2\ n^2$ for hydrogenlike ions, one obtains for the gain

$$\alpha_\nu = 1.29\times 10^{-39}(N_1^2/Z^4)(kT_e/Z^2)^{-3/2}2n^3 - 7.76\times 10^{-18}N_0 n/Z^2. \qquad (20)$$

Taking $Z=1$, $kT_e=5$ eV, $n=1$, and $N_1=10^{20}$ cm^{-3}, one has gain for $N_0 < 2.96\times 10^{17}$ cm^{-3}. If N_0 is negligible compared with that number, a gain of 2.3 cm^{-1} is achieved at a wavelength of 91.2 nm.

To evaluate the pumping requirements for the realization of such plasma conditions, the loss of "inversion" by recombination has to be taken into account. Any ionizing radiation can be used for pumping. The necessary pump power absorbed by the medium is given by the recombination rate R_{rec} multiplied by the average energy for reionization of an atom. One obtains

$$P = R_{\text{rec}}(\mathscr{R} + kT_e)1.6\times 10^{-19}\ \text{W/cm}^3. \qquad (21)$$

Taking the recombination rate from Bates, Kingston, and McWhirter,[11] one obtains in an optically thin plasma

$$P = 8.6\times 10^{12}\ \text{W/cm}^3. \qquad (22)$$

A free-bound laser would be an interesting way of generating coherent radiation in the uv and x-ray regions of the spectrum. Since the upper level is in the continuum, a given atom would lase at a much shorter wavelength than for a bound-bound transition. A major advantage would be the possibility of continuous tunability. It turns out, however, from the gain formulas of the preceding discussion that the requirements on a medium for it to exhibit reasonable free-bound gain are rather severe. Typically, electron and ion densities greater than 10^{20} cm^{-3} are required with a population of the lower level of less than 10^{18} cm^{-3} (see the previous example).

The situation easiest to verify experimentally is case (a), which only requires that the population of the bound level be much lower than given according to the Saha equilibrium. The Saha equation yields

$$N_0^{\text{Saha}} = 1.66\times 10^{-22}N_1 N_e (kT_e)^{-3/2}(g_0/g_1) \times \exp(E_b/kT_e), \qquad (23)$$

from which and Eq. (10) it follows that there is gain if

$$N_0 < N_0^{\text{Saha}} \exp(-E_b/kT_e). \qquad (24)$$

Gain to an excited level of the atom is therefore achieved if the temperature of the atom, as determined by the population of its bound levels, is much lower than the electron temperature determining the degree of ionization.

A disadvantage of case (a) is that the gain is maximum at the ionization limit (unless σ_ν has a distinct maximum in the continuum), and therefore such a laser would only marginally be a free-bound laser.

In case (b) lasing from well within the continuum is achievable. Because of the high self-relaxation rate of the electrons, the required narrow velocity distribution can probably be maintained only for a very short period of time and traveling-wave excitation would be necessary. However, if the energy spread of the electrons can be made sufficiently small, considerably higher gain than in case (a) could be obtained.

Case (c) could be verified by crossing an electron beam and an ion beam. The population of the lower level would be zero in this case, making the gain in any case positive. For its magnitude to be experimentally measurable, the current densities of existing electron beam generators would have to be considerably increased.

Under high-density plasma conditions the gain will be reduced by scattering and absorption. The total cross section of an electron for scattering a photon is[12]

$$\sigma_e = 6.65\times 10^{-25}\ \text{cm}^2, \qquad (25)$$

and so scattering can safely be ignored. The main photon-loss mechanism is free-free absorption by the electron gas, given by[12]

$$\beta_{ff} = 3.4\times 10^6 Z^2 N_1^2 (kT_e/Z^2)^{-1/2}\nu^{-3} \times (1 - e^{-h\nu/kTe})\ \text{cm}^{-1}. \qquad (26)$$

Under the conditions of the example this results in

$$\beta_{ff} = 0.4\ \text{cm}^{-1} \qquad (27)$$

and the gain is reduced by 17%.

It is interesting to discuss the effects which determine the gain profile of a free-bound laser. The contribution of the lower (bound) level to the linewidth will be negligible in most cases and the gain profile will be controlled by the term $[n_e(v)/v^2]\sigma_\nu(\nu)$ in Eq. (8), i.e., by the electron velocity distribution and the variation of the photoionization cross section with frequency. If the electron velocity distribution is narrow, as in cases (b) and (c), $\sigma_\nu(\nu)$ can be considered as constant and the gain bandwidth is given by the electron energy spread. In the case of a Maxwellian electron velocity distribution [case (a)], however, the variation of $\sigma_\nu(\nu)$ with frequency has to be taken into account. For lasing into hydrogenic states one obtains, by use of Eq. (21), for the functional form of the gain profile ($\nu \geqslant \nu_c$)

$$\alpha_\nu(\nu) = \alpha_0 \exp[-(\nu-\nu_0)/kT_e](\nu_0^3/\nu^3), \qquad (28)$$

where α_0 is the gain at the maximum, given by Eq. (10), and ν_0 is the frequency at maximum gain, given by $h\nu_0 = \mathscr{R} Z^2/n^2$. For the previous example the gain is reduced to half of its maximum value within a bandwidth of $\Delta\nu/\nu_0 = 0.125$.

It is clear that the emission bandwidth of a laser operating on a free-bound transition will be much smaller than the gain bandwidth, since gain narrowing and possible cavity effects will reduce the spectral width of the emission. However, such radiation would still extract energy from all of the electrons since self-relaxation would quickly restore any holes burnt into the electron velocity distribution. In other words, the transition can be considered as homogeneously broadened down to a pulse duration given by the self-relaxation time of the electrons.

In summary, by analogy with the situation at a bound-bound transition, an "inversion" can be defined for a free-bound transition which leads to gain by stimulated radiative recombination. Gain formulas for three cases have been derived, the first being characterized by a Maxwellian electron velocity distribution, the second by a velocity distribution approaching a delta function, and the third by further restriction of the electrons in phase space by directionality.

Though difficult to achieve, gain on a free-bound transition would certainly be an effect of high interest, possibly leading to a tunable short-wavelength laser. A more detailed discussion concerning pumping requirements and competing processes will be left to a further publication.

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