

# Some nonlinear optical phenomena

Boualem Hammouda

*Université d'Alger, Algiers, Algeria*

Richard K. Osborn

*Department of Nuclear Engineering, The University of Michigan, Ann Arbor, Michigan 48109*

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Some nonlinear optical phenomena are investigated, especially stimulated scattering, from the point of view of the kinetic theory of radiation (i.e., photon transport theory). Kinetic theory provides a perspective, different from Maxwell's wave theory, from which an examination of these complex matters may proceed with some advantages: (i) considerable mathematical simplification in some instances, (ii) clear and natural separation of microscopic versus macroscopic nonlinearities, (iii) kinetic theory couples the radiation field nonlinearly to a formally exact description of the matter field, and (iv) it is believed that the mathematical model provided by kinetic theory is perhaps better suited for numerical studies of the effect of diverse nonlinear optical phenomena upon laser-fusion implosion dynamics than Maxwell's wave theory. Although the main emphasis is upon stimulated scattering, the incorporation of other nonlinearities into the kinetic model is discussed briefly.

## I. INTRODUCTION

In this investigation we examine some nonlinear optical phenomena from the point of view of a kinetic model for the radiation field. Although few new results will be presented, we feel that such an examination is useful for several reasons. First of all, it is often clarifying to view a complex subject from a variety of perspectives. The kinetic model of the radiation field (descriptive of photons or radiation intensity) provides an alternative to Maxwell's wave theory which is probably valid for the study of those linear and nonlinear interactions of electromagnetic radiation with matter which are not acutely phase sensitive (i.e., as presently developed the kinetic theory is quite inadequate for the description of reflection and refraction). Second, the investigation of some nonlinear interactions of electromagnetic radiation with matter is greatly simplified mathematically when viewed from the kinetic perspective. We find this to be especially the case in the study of stimulated scattering, which will receive the main emphasis herein. In fact, it is a serendipitous gift of the kinetic model that it largely describes the stimulated scattering of light by matter at the lowest level of formulation. Third, the interaction of radiation with matter can become nonlinear with respect to the radiation variables at two distinctly different levels, i.e., the microscopic level and the macroscopic level. Examples of the former are multiphoton absorption and ionization, multiphoton bremsstrahlung, the nonlinear index of refraction, a special mechanism for laser gain saturation to be discussed later, and to a large extent stimulated scattering. Examples of the latter are found in situations where intense laser light impinges on matter inducing significant changes in material density, velocity, and temperature which in turn modify the radiation field. These two kinds of nonlinearities may be manifest separately under certain circumstances. It is a virtue of the kinetic model in that it provides, clearly and naturally, a separation of these kinds of nonlinear effects. Fourth, the kinetic model (at least at the level to be discussed here) relates the dynamics

of the radiation field to a material density-density correlation function,  $S(\Delta k, \Delta \omega)$ , without the necessity of specifying the state of the matter field (e.g., solid, liquid, gas, or plasma) and without regard for the dynamical state of the matter field (e.g., thermodynamic or completely otherwise). Of course, if the radiation field is macroscopically modifying the matter field, the correlation function  $S$  will be an explicit function of the radiation variables. Fifth, we believe that the mathematical model provided by kinetic theory may be better suited than Maxwell's wave theory to a study of the effect of stimulated scattering upon laser-fusion implosion dynamics.

In Sec. II we introduce the mathematical model. With an eye to the study of stimulated scattering; we then specialize to a description of forward and backward streaming photons with specified energies. In Sec. III, stimulated Brillouin scattering of light by plasmas and neutral systems is studied. Formulas for thresholds and growth rates are derived and compared with results obtained elsewhere by other arguments. Section IV contains an enumeration and discussion of some other, familiar and not so familiar, nonlinear interactions of radiation with matter.

## II. MATHEMATICAL MODEL

The material system may be described either at the kinetic level<sup>1-3</sup> or the hydrodynamic level<sup>4-7</sup> depending upon the desired sophistication. Actually, the coupling of photons with matter is via density-density correlation functions. However, the emphasis here is upon nonlinearities in the radiation field so we will depend on others for the calculation of these correlation functions. We merely note in passing that the one-fluid, two-temperature hydrodynamic model commonly employed in laser fusion numerical studies is naturally coupled to the radiation variable studied here. In the momentum equation, electromagnetic stress is to be added to material stress, and if only diagonal elements are retained, the kinetic pressure  $P$  goes to  $P + I_1/C$ , where  $I_1$  is the light intensity. In the electron energy

equation, intensity enters again because of heating by net inverse bremsstrahlung. In what follows, our first task is to introduce the kinetic equation describing the radiation field.

### A. Photon transport equation

To describe the transport of radiation in a medium we define the photon number density  $F_\lambda$ , such that  $F_\lambda(\mathbf{r}, \omega, \Omega, t) d^3r d\omega d\Omega$  is the number of photons in  $d^3r$  about  $\mathbf{r}$  with frequencies in  $d\omega$  about  $\omega$  traveling in directions in  $d\Omega$  about  $\Omega$  with polarization (spin)  $\lambda$  at time  $t$ . We can derive an equation for this phase space density function characterizing the transport process by simply balancing the various mechanisms by which photons can be gained or lost from a specified volume. However, this particle description of the radiation field requires that the photon mean-free-paths be sufficiently large compared with the interaction distances. Also, it is only valid in plasmas in underdense regions.

The mechanisms which can change the photon number density are; streaming, scattering, absorption, and photon sources, if any. For the time being we consider only passive media, and delay the introduction of a gain term accounting for stimulated emission in active media until Sec. IV. Such a balanced equation that describes the transport of bosons was introduced as far back as 1933 by Uehling and Uhlenbeck.<sup>8</sup> Their interest was in a kinetic theory for liquid helium, but the ideas carry over to the photon gas by close analogy. We write this equation as

$$\frac{\partial F_\lambda}{\partial t} + \nabla \cdot \eta c \Omega F_\lambda = \int d\omega' d\Omega' N c \sigma_{\lambda\lambda'}(\omega'\Omega', \omega\Omega) F_\lambda' - \int d\omega' d\Omega' N c \sigma_{\lambda\lambda'}(\omega\Omega, \omega'\Omega') F_\lambda - \nu F_\lambda, \quad (1)$$

where  $\eta$  is the index of refraction for the medium,  $N$  is the density of the scattering particles (electrons),  $\nu$  is the net rate of absorption of photons, and  $\sigma_{\lambda\lambda'}(\omega\Omega; \omega'\Omega') d\omega' d\Omega'$  is the scattering cross section. The scattering cross section may be displayed as

$$\sigma_{\lambda\lambda'}(\omega\Omega; \omega'\Omega') = \frac{\omega'}{\omega} \sigma_t \left( 1 + \frac{(2\pi c)^3}{\omega'^2} F_\lambda' \right) S(\Delta\mathbf{k}, \Delta\omega) f_{\lambda\lambda'}(\omega, \omega'), \quad (2)$$

where  $\sigma_t$  is the Thomson cross section,

$$S(\Delta\mathbf{k}, \Delta\omega) \equiv \frac{1}{2\pi N} \int dt \exp(i\Delta\omega t) \langle \rho^*(\Delta\mathbf{k}, 0) \rho(\Delta\mathbf{k}, t) \rangle$$

is the Fourier transformed density-density correlation function ( $\Delta\mathbf{k} = \mathbf{k} - \mathbf{k}'$  and  $\Delta\omega = \omega - \omega'$ ) describing the dynamics of the external degrees of freedom of the scattering system (free electron or atom or molecule), and  $f_{\lambda\lambda'}(\omega, \omega')$  is a function describing the dynamics of the internal degrees of freedom of the scattering system. This nonlinear cross section is derived by using the usual golden rule. The term independent of the photon density is extensively used in light scattering studies,<sup>9</sup> while the term proportional to the photon density in the final state is discussed by Dirac<sup>10</sup> and is needed to recover the Planck distribution for the radiation gas in

thermal equilibrium (as also is spontaneous emission which we are herein ignoring). We emphasize the fact that the interaction of individual photons with matter is treated as a perturbation, while material dynamics, buried in the scattering function  $S(\Delta\mathbf{k}, \Delta\omega)$  and  $f_{\lambda\lambda'}(\omega, \omega')$ , can be described formally exactly. The function  $S$  may include macroscopic radiation effects; i.e., in cases in which the scattering medium is not in thermal equilibrium it may depend upon the variables characterizing the radiation field. Since we are here focusing on microscopic nonlinearities in the description of the radiation field, the assumption of thermal equilibrium for the matter field will be invoked throughout the subsequent discussion.

Derivations of formulas to describe the scattering function, using differing approaches and differing approximation schemes, have been presented.<sup>1-3</sup> A general feature is that it exhibits peaks symmetric with respect to zero-frequency shift,  $\Delta\omega = 0$ . In the plasma case,<sup>11,12</sup> there are two acoustic modes at  $\Delta\omega = \pm\omega_a$ , and two plasma modes at  $\Delta\omega = \pm\omega_p$ , and a Doppler peak centered at  $\Delta\omega = 0$ . In the case of neutral fluids<sup>9,13</sup> there are two acoustic (Brillouin) lines at  $\Delta\omega = \pm\omega_a$  besides the central Rayleigh line at  $\Delta\omega = 0$ . Thus, we display

$$S(\Delta\mathbf{k}, \Delta\omega) = \sum_m S_m(\Delta\mathbf{k}, \Delta\omega), \quad (3)$$

and approximate the contribution from each mode by a Lorentzian peaked at  $\omega_m$ , i.e.,

$$S_m(\Delta\mathbf{k}, \Delta\omega) = H_m \{ \gamma_m / [(\Delta\omega - \omega_m)^2 + \gamma_m^2] \}, \quad (4)$$

where  $\gamma_m$  is the width of the  $m$ th mode and  $H_m/\gamma_m$  is its maximum height. For example, Mountain<sup>9</sup> finds, for the case of a simple fluid, a structure like that in Eqs. (3) and (4) with,

$$\gamma_a = \frac{1}{2} \left( \frac{4}{3} \frac{\mu_s + \mu_b}{N} + \frac{1}{N} \left[ \frac{\Gamma}{C_v} - \frac{\Gamma}{C_p} \right] \right) \Delta k^2, \quad (5)$$

$$\gamma_c = \Gamma \Delta k^2 / N C_p,$$

for the widths of the acoustic and central peaks, where  $\mu_s$  and  $\mu_b$  are the shear and bulk viscosities,  $\Gamma$  is the thermal conductivity,  $C_v$  and  $C_p$  are the specific heats at constant volume and constant pressure, and  $N$  is the average particle number density.

One advantage of approaching the problem of stimulated scattering from the point of view of kinetic theory is that it provides, quite generally, a nonlinear relation between radiation intensity [Eq. (1)] and well defined, formally exact, functions of the dynamical variables of the scattering medium [Eq. (2)]. The scatterer may be a neutral fluid or solid, in which case the microscopic function,  $f_{\lambda\lambda'}(\omega, \omega')$ , is determined by conventional arguments and may be approximated in terms of atomic or molecular polarizabilities. If it is a fully ionized plasma,  $f_{\lambda\lambda'} = |\epsilon_{\lambda\lambda'}|$ , which describes the polarization (of the radiation field) dependence of the scattering cross section.

### B. Intensity equations

We assume that the incident laser and scattered beams have well-defined directions,  $\Omega_i$  and  $\Omega_s$ , and

frequencies,  $\omega_l$  and  $\omega_s$ . If we neglect multiple scattering, we may display

$$F_\lambda = F_l \delta(\Omega - \Omega_l) + F_s \delta(\Omega - \Omega_s), \quad (6)$$

where  $F_l$  and  $F_s$  are functions of frequency peaked around  $\omega_l$  and  $\omega_s$ , respectively. We integrate Eq. (1) over small ranges in frequency and solid angle ( $\Delta\omega_l$  and  $\Delta\Omega_l$ ) centered at  $\omega_l$  and  $\Omega_l$  and define the intensities for the incident and scattered beams as

$$I_l = \hbar \omega_l C \int_{\Delta\omega_l} d\omega F_l, \quad I_s = \hbar \omega_s C \int_{\Delta\omega_s} d\omega F_s. \quad (7)$$

We find that

$$\frac{\partial I_l}{\partial t} + \nabla \cdot \eta C \Omega_l I_l = -\nu I_l - r_l \frac{\omega_s}{\omega_l} I_l + r_s \frac{\omega_l^2}{\omega_s^2} I_s - G_l I_s I_l, \quad (8)$$

where

$$r_l = NC \sigma_t \Delta\omega_s \Delta\Omega_s S(\Delta\mathbf{k}, \Delta\omega) f_{\lambda\lambda'}(\omega_l, \omega_s), \quad (9)$$

$$r_s = NC \sigma_t \Delta\omega_l \Delta\Omega_l S(-\Delta\mathbf{k}, -\Delta\omega) f_{\lambda\lambda'}(\omega_s, \omega_l),$$

and

$$G_l = [(2\pi C)^3 N \sigma_t / \hbar \omega_s^2 \omega_l] [S(\Delta\mathbf{k}, \Delta\omega) f_{\lambda\lambda'}(\omega_l, \omega_s) - S(-\Delta\mathbf{k}, -\Delta\omega) f_{\lambda\lambda'}(\omega_s, \omega_l)]. \quad (10)$$

For scattering systems in thermal equilibrium ( $\Delta\omega = \omega_l - \omega_s$  and  $\Delta\mathbf{k} = \mathbf{k}_l - \mathbf{k}_s$ ),

$$S(-\Delta\mathbf{k}, -\Delta\omega) f_{\lambda\lambda'}(\omega_s, \omega_l) = \exp(-\hbar\Delta\omega/\theta) S(\Delta\mathbf{k}, \Delta\omega) f_{\lambda\lambda'}(\omega_l, \omega_s), \quad (11)$$

so that Eq. (10) becomes

$$G_l = \frac{(2\pi C)^3 N \sigma_t}{\hbar \omega_l \omega_s^2} \left[ 1 - \exp\left(-\frac{\hbar\Delta\omega}{\theta}\right) \right] S(\Delta\mathbf{k}, \Delta\omega) f_{\lambda\lambda'}(\omega_l, \omega_s). \quad (12)$$

The transport equation for the scattered intensity  $I_s$  is obtained from Eq. (8) by the index interchange,  $l \rightarrow s$  and  $s \rightarrow l$ , leading to

$$\frac{\partial I_s}{\partial t} + \nabla \cdot \eta C \Omega_s I_s = -\nu I_s - r_s \frac{\omega_l}{\omega_s} I_s + r_l \frac{\omega_s^2}{\omega_l^2} I_l - G_s I_l I_s, \quad (13)$$

where now

$$G_s = \frac{(2\pi C)^3 N \sigma_t}{\hbar \omega_s \omega_l^2} \left[ \exp\left(-\frac{\hbar\Delta\omega}{\theta}\right) - 1 \right] \times S(\Delta\mathbf{k}, \Delta\omega) f_{\lambda\lambda'}(\omega_l, \omega_s) \approx -G_l, \quad (14)$$

for  $\omega_s \approx \omega_l$ . To estimate the effect of a finite laser bandwidth  $\gamma_l$  upon the transport coefficients in Eqs. (8) and (13), we model the photon density in the laser beam as

$$F_l = \frac{I_l}{\hbar C \omega_l} \frac{\exp[-(\omega - \omega_l)^2 / \gamma_l^2]}{\sqrt{\pi} \gamma_l}, \quad (15)$$

and, for convenience, a line in the fluctuation spectrum as

$$S_m(\Delta\mathbf{k}, \Delta\omega) = \frac{H_m}{\gamma_m} \exp\left(\frac{(\Delta\omega - \omega_m)^2}{\gamma_m^2}\right). \quad (16)$$

We find that the finite laser bandwidth effect is to multi-

ply the zero laser bandwidth transport coefficient by the factor,  $(\gamma_m/\gamma_l)$ , with  $\gamma_l^2 = \gamma_m^2 + \gamma_l^2$ .

### III. STIMULATED BRILLOUIN SCATTERING

Brillouin scattering is the inelastic scattering of photons by a medium with the exchange of a phonon between the medium and the light beam ( $\Delta\omega = \omega_l - \omega_s = \pm\omega_a$ ). We will model the acoustic peak in the scattering function as a normalized Lorentzian as in Eq. (4) with  $H_m = 1/\pi$ ,  $\gamma_m = \gamma_a$ , the sound wave damping frequency, and  $\omega_m = \pm\omega_a$  the acoustic frequency. We assume that the acoustic frequency is small compared with the laser frequency ( $\omega_s \approx \omega_l$ ) and that  $\hbar\Delta\omega/\theta \ll 1$ . With these assumptions we find that

$$r_l \approx r_s = NC \sigma_t f_{\lambda\lambda'} \frac{\gamma_a}{\gamma_l} = R, \quad (17)$$

and for backward, down-shifted Brillouin scattering

$$G_l \approx G_s = \frac{(2\pi C)^3 N \sigma_t \omega_a}{2\pi \omega_l^3 \theta \gamma_l} f_{\lambda\lambda'} \equiv G. \quad (18)$$

Thus Eqs. (8) and (13) become

$$\frac{\partial I_l}{\partial t} + \nabla \cdot \eta C \Omega_l I_l = -(\nu + R)I_l + R I_s - G I_s I_l, \quad (19)$$

$$\frac{\partial I_s}{\partial t} + \nabla \cdot \eta C \Omega_s I_s = -(\nu + R)I_s + R I_l + G I_l I_s. \quad (20)$$

These equations, with  $\nu = R = 0$ , have appeared elsewhere.<sup>4</sup> An inspection of Eqs. (10) and (14) reveals that  $G$  is positive for  $\Delta\omega = \omega_l - \omega_s = \omega_a$  (the Stokes line) and, since  $\omega_a = |\Delta\mathbf{k}|v_a$  where  $v_a$  is the acoustic speed in the medium, is a maximum in the backward direction. Thus, backward, down-shifted, stimulated Brillouin scattering is to be expected. We now turn to an investigation of some of the implications of Eqs. (19) and (20) for light scattering by various media.

#### A. Plasma case

In plasmas, stimulated Brillouin scattering occurs in underdense regions characterized by density gradient scale lengths large compared with the wavelength of the incident light. This phenomenon can prevent laser energy from reaching the region of plasma density near critical where absorption is efficient. For present purposes, we will assume a fully ionized plasma, i.e.,  $f_{\lambda\lambda'} = |\epsilon_{\lambda\lambda'} \epsilon_{\lambda\lambda'}|^2 = 1$  for linearly polarized incident light and scattering that does not change the state of polarization.

Above threshold, the Stokes wave grows in the backward direction, and its growth rate at the center line of the acoustic mode,  $\Delta\omega = \omega_a$ , is a maximum and is given by

$$G I_l = 2 \frac{(2\pi C)^3 N \sigma_t \omega_a}{2\pi \omega_l^3 \theta \gamma_l} I_l. \quad (21)$$

Introducing,  $\omega_a = |\Delta\mathbf{k}|v_i \approx 2k_i(\theta/m_i)^{1/2}$  and the electron quivering speed,  $v_i = eE_l/m_e\omega_l$ , with  $E_l^2 = 4\pi I_l/c$ , we may rewrite Eq. (20) in the form

$$G I_l = 2\omega_{pi}^2 v_i^2 / c v_i \gamma_l, \quad (22)$$

where  $\omega_{pi}$  is the ion plasma frequency. To within the

factor of two, this result is the same as the one given by Liu<sup>5</sup> for the case of weak coupling.

We may estimate thresholds for this stimulated back-scattering from Eq. (20), i.e., by requiring

$$GI_{\text{th}} > \nu + R + \eta C \Omega_s \cdot \nabla I_s / I_s \simeq \nu + R + \eta C / L, \quad (23)$$

where we have (arbitrarily) approximated the streaming loss rate as the reciprocal of the time required for photons to travel a distance  $L$ , a distance over which the plasma density does not vary very much. The quantities  $\nu$  and  $R$  are the rate coefficients for net inverse bremsstrahlung and Thomson scattering, respectively, and, in most situations of interest here,  $\nu \gg R$ . For fairly homogeneous plasmas,  $L$  is so large that  $\nu \gg \eta C / L$  and the threshold condition becomes

$$I_{\text{th}} > \nu / G, \quad (24)$$

which may be cast into the form

$$\frac{v_{\text{th}}^2}{v_e^2} > 4 \frac{\gamma_a}{\omega_a} \frac{\nu}{\omega_i} \frac{\omega_i^2}{\omega_{pe}^2} \frac{\gamma_t}{\gamma_a}. \quad (25)$$

where  $v_{\text{th}}$  is the electron quivering speed at the threshold intensity and  $v_e^2 = \theta / m_e$  is the square of the electron thermal speed. This differs from the formula presented by Forslund *et al.*<sup>4</sup> [their Eq. (23)] by the factor  $(\omega_i^2 \gamma_t / \omega_{pe}^2 \gamma_a) = (\omega_i^2 / \omega_{pe}^2)$  (they do not consider finite laser bandwidth effects). This discrepancy is near unity in regions close to critical density. For the case of inhomogeneous plasmas,  $L$  may be so small that  $\nu \ll \eta C / L$  and the threshold condition becomes

$$I_{\text{th}} > \eta C / LG, \quad (26)$$

$$\frac{v_{\text{th}}^2}{v_e^2} > \frac{8}{Lk_i} \frac{\omega_i^2}{\omega_{pe}^2} \frac{\eta \gamma_a}{2\omega_a} \frac{\gamma_t}{\gamma_a}.$$

This result differs from the one quoted by Chen<sup>14</sup> by the factor,  $(\eta \gamma_a \gamma_t / 2\omega_a \gamma_a) = \eta \gamma_a / 2\omega_a$ , zero laser bandwidth). Actually,  $\eta$  is close to unity as is also  $\gamma_a / \omega_a$  (Ref. 15), so that the discrepancy is of the order of a factor of one-half. Note that, for  $\gamma_t = (\gamma_a^2 + \gamma_i^2)^{1/2} \simeq \gamma_i$ , which is the case in many practical situations, the threshold for stimulated Brillouin back-scattering increases linearly with the laser bandwidth, as has previously been noted.<sup>16</sup>

We are unable to account for the discrepancies, noted above, between the results presented here and those quoted by other investigators. They probably arise, at least in part, because of the extreme differences in the approach to these complex matters between the one explored here and those explored elsewhere. In any event and in most cases, they are of the order of magnitude unity; so that, in the absence of sufficiently precise measurement, we are unable to regard them as serious.

It is interesting to explore the implications of Eqs. (19) and (20) in the special case for which streaming is ignorable. In that instance it is seen that

$$I_i(t) + I_s(t) = I_0 \exp(-\nu t), \quad (27)$$

with the initial conditions

$$I_i(0) = I_0, \quad I_s(0) = 0. \quad (28)$$

Analytical solutions are readily obtained for the case of no absorption ( $\nu = 0$ ). Setting  $I_i = I_0 - I_s$  in Eq. (20) leads to

$$\frac{dI_s}{dt} = -GI_s^2 + (GI_0 - 2R)I_s + RI_0, \quad (29)$$

which integrates to

$$I_s(t) = I_0 \frac{\exp(Bt) - 1}{\exp(Bt) + GI_0/R}, \quad (30)$$

where  $B = GI_0 + R$ . Saturation due to pump depletion is evident. Equation (30) can be integrated to yield a reflectance, but the resulting formula is complicated and probably irrelevant due to the neglect of absorption. A reflectance of unity is predicted for the experiment reported by Mayer *et al.*<sup>15</sup> whereas they observe only forty to sixty percent reflection. We surmise that the difference is largely due to absorption.

## B. Neutral media case

Equations (19) and (20) may also be used for the study of stimulated Brillouin scattering by bound electrons in neutral media provided we appropriately modify the function  $f_{\lambda\lambda'}$  appearing in Eq. (2) to account for the effects of internal molecular degrees of freedom upon the scattering process. We appeal to Mountain<sup>9</sup> for this modification and find that

$$f_{\lambda\lambda'} = |\epsilon_{\lambda} \cdot \epsilon_{\lambda'}|^2 (m\alpha/e^2)^2 \omega^4, \quad (31)$$

for this case ( $\alpha$  is the polarizability of the scattering molecules). The transport coefficients in Eqs. (19) and (20) now read

$$R = NC \sigma_t f_{\lambda\lambda'} = (N\alpha^2 \omega_i^4 / c^2) |\epsilon_{\lambda} \cdot \epsilon_{\lambda'}|^2, \quad (32)$$

and

$$G = [(2\pi)^2 N \omega_a \omega_i \alpha^2 / c \theta \gamma_t] |\epsilon_{\lambda} \cdot \epsilon_{\lambda'}|^2, \quad (33)$$

and  $\nu$  (the net absorption rate coefficient) is negligibly small except near resonance. For optical photons, the Rayleigh scattering rate by bound electrons given by Eq. (32) is smaller than the Thomson scattering rate by free electrons by several orders of magnitude. The steady-state version of Eqs. (19) and (20) was derived by Tang,<sup>17</sup> employing very different arguments, and used in an investigation of stimulated Brillouin scattering by crystals.

## IV. SOME OTHER NONLINEARITIES

In addition to stimulated scattering, there are several other nonlinear optical phenomena that are conveniently described in the context of kinetic theory, Eq. (1). Bound-bound, two photon absorption introduces a sink term into the kinetic equation of the form,  $-\gamma F^2$ , i.e.,  $\nu \rightarrow \nu + \gamma F$ . It is this phenomenon that is believed to be responsible for gain saturation in Nd-glass laser amplifiers.<sup>18</sup> Light absorption by plasmas due to net inverse bremsstrahlung may become strongly nonlinear at high intensity.<sup>19</sup> According to a simple argument sketched in the Appendix, we find that that part of  $\nu$  descriptive of absorption by bremsstrahlung is modified according to,  $\nu \rightarrow \nu f(x)$ , where

$$f(x) = \exp(-x) [I_0(x) - I_1(x)],$$

and

$$x = e^2 E^2 / 4m \theta \omega^2 = e^2 \lambda^2 I / 4\pi m \theta c^3 = v_{osc}^2 / 4v_{th}^2.$$

The functions,  $I_0$  and  $I_1$ , are the modified Bessel functions of the first kind. This nonlinear correction factor is in good agreement with the one presented in Ref. 15, Fig. 1. In a lasing medium there is a source term in the kinetic equation of the form,  $cgf$ , where  $g(\text{cm}^{-1})$  is the small signal gain coefficient. This gain coefficient may be displayed as  $g = (n_u - n_l)\sigma$ , where  $n_u$  and  $n_l$  are the atomic densities in the upper and lower lasing states, respectively, and  $\sigma$  is the atomic cross section for the  $u \neq l$  transition. The atomic densities  $n_u$  and  $n_l$  may become intensity dependent (particularly  $n_u$ , e.g., gain depletion) as may also the cross section itself due to intensity-dependent broadening of the upper lasing level-power broadening.<sup>20,21</sup> In the latter instance,  $g \rightarrow g_0 / (1 + \kappa I)$  at line center, where  $g_0$  is the gain coefficient at zero intensity and

$$\kappa = \frac{8\pi^3 c^2 \gamma_r}{\hbar \omega^4 \gamma_l (\Delta\omega_l / \omega)} = 1.8 \times 10^{12} \lambda^4 \frac{\gamma_r}{\gamma_l} \frac{\omega}{\Delta\omega_l} \text{ cm}^2 / \omega.$$

In this formula,  $\gamma_r$  is the zero intensity radiative width of the upper lasing level,  $\gamma_l$  is the width of the lower lasing level, and  $(\Delta\omega_l / \omega)$  is the ratio of laser bandwidth to laser frequency. The nonlinearity induced by power broadening is universal for lasers because it is simply a consequence of the fact that stimulated emission from the upper level must shorten its lifetime and, hence, increase its width. Because  $\kappa$  is proportional to  $\lambda^4$ , we expect this phenomenon to be relatively more important for long wavelength lasers. For example, estimating  $\gamma_r / \gamma_l = 10^{-9}$  and  $\omega / \Delta\omega_l \approx 10^3$ , we find for the CO<sub>2</sub> laser,  $\kappa \approx 2 \times 10^{-6} \text{ cm}^2 / \omega$ . Thus, we would expect that CO<sub>2</sub> amplifiers might begin to exhibit gain saturation at intensities of the order of  $10^6 - 10^7 \text{ W/cm}^2$ .

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## APPENDIX

Here, we sketch an argument leading to the formula for the correction factor for the absorption coefficient for net inverse bremsstrahlung displayed in the text. We set

$$\frac{dN\theta}{dt} = \left\langle \frac{m_e u^2}{2} NR \right\rangle \equiv kI, \quad (\text{A1})$$

where  $m_e u^2 / 2$  is the quivering energy of an electron in an electric field,  $E [u = (eE/m\omega)\cos\omega t]$ ,  $N$  is the electron density,  $R$  is an average electron-ion collision rate, and  $k$  is the absorption coefficient per centimeter. The symbol  $\langle \rangle$  means time average over a period of the electromagnetic wave. The collision rate may be approximately displayed as

$$R = n^i \int d^3v P(\mathbf{v})v\sigma_{ei}(v), \quad (\text{A2})$$

where  $P(\mathbf{v})d^3v$  is the probability of finding an electron with a velocity in  $d^3v$  about  $\mathbf{v}$ . This probability may be estimated to be a Maxwellian in a reference frame moving with the quivering velocity of the electron, i.e.,

$$P(\mathbf{v}) \approx M(\mathbf{v} - \mathbf{u}) = \exp\left(-\frac{m_e u^2}{2\theta}\right) \exp\left(\frac{m_e \mathbf{u} \cdot \mathbf{v}}{\theta}\right) M(v). \quad (\text{A3})$$

Neglecting the factor,  $\exp(m_e \mathbf{u} \cdot \mathbf{v} / \theta)$ , Eq. (A2) becomes

$$R = R_0 \exp(-m_e u^2 / 2\theta), \quad (\text{A4})$$

where  $R_0$  is the collision rate at zero intensity. Performing the time average in Eq. (A1) we find that

$$kI = k_0 \exp(-x) [I_0(x) - I_1(x)] I, \quad (\text{A5})$$

where  $k_0$  is the absorption coefficient at zero intensity,  $x = (v_{osc} / 2v_T)^2$ , and  $I_0$  and  $I_1$  are the modified Bessel functions of the first kind. The correction factor,  $e^{-x} [I_0(x) - I_1(x)]$ , is the one discussed in the text.

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