Rate equations in stimulated light scattering

P. L. Kelley
Lincoln Laboratory, Massachusetts Institute of Technology, Lexington, Massachusetts 02173
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A set of rate equations which gives results not previously obtained has been proposed for stimulated light scattering. These equations are shown to be invalid under the conditions where the novel results appear.

Recently the well-known solutions to the coupled equations describing stimulated light scattering have been questioned. A new analysis based on an application of the "golden rule" has given solutions which show an additional instability.[1-5] It is the aim of this note to show why the second approach gives incorrect results for some values of the coupling and damping parameters.[6] In particular, the prediction of "enhanced" stimulated scattering is found to be wrong.

The starting point for the generally accepted calculations is a set of coupled partial differential equations for the amplitudes of the scattered electromagnetic field, the driving input field, and the material model (e.g., phonons).[7] In general, nonlinear equations cannot be readily solved. A number of approximations can be made, one of the most familiar is to linearize the problem by assuming the scattered-field and material-mode amplitudes remain small compared to the driving field. This is well justified both mathematically and physically as an approximation useful in describing the onset of the stimulated scattering instability and the growth of the scattered waves. Loudon[8] has shown that it is possible to avoid this approximation and obtain an analytic solution for the case of undamped driving and scattered electromagnetic waves and assuming the material amplitude is determined by an instantaneous dynamical equilibrium between the rate of growth due to the electromagnetic driving term and the damping. As one expects, when the depletion of the driving field becomes significant, the growth is less than the growth in the corresponding linearized regime.

The linearized equations have been solved for a wide variety of spatial and temporal boundary conditions. One early result was for purely spatial growth.[9] Kroll[10] extended the analysis to include both spatial and temporal growth using integral equations, an approach which has seen extended development.[11-14] A dispersion relation may also be found from the equations of motion and a steepest-descent analysis made to find the asymptotic solutions having maximum growth for various lengths of nonlinear medium, pulse durations, and pump intensities.[15] This method is very useful for material modes which give rise to complicated dispersion relations. The results of these methods of analysis are consistent with each other.

The approach which has given different results is due to Sparks[1-5] and involves rate equations for the intensity (square amplitudes) or density of quanta of the scattered electromagnetic and material fields. Note that Sparks' treatment also includes dynamical equations for the density of quanta of the input field. However, the assumption of a constant input field is valid for the initial buildup of the scattered fields. Although we will examine the problem under this condition, relaxing this constraint will not modify the conclusions. Two techniques have been used by Sparks to obtain the rate equations. One technique contains a mathematical error[16] which reduces the generality of the results. The second approach is to simply apply the golden rule of time-dependent perturbation theory to the calculation of the rate equations. While this is very appealing, care must be taken to use the golden rule in its domain of validity.

Generally speaking, when first-order transport equations are derived for a subset of the dynamical variables of a system, integrations must be performed which make the resulting equations temporally nonlocal (non-Markovian) and spatially nonlocal. Similarly, in quantum mechanics, when deriving equations of motion for a subset of the total density matrix, an integration must be performed in order to eliminate the unwanted subset and retain the apparent first-order character of the final equations. For certain constraints on the parameters of the system a local approximation may be used. The temporally local approximation is also called the Markovian or rate-equation approximation. As we shall see, even when the Markovian approximation is valid, a rate equation based on inappropriate application of the golden rule can lead to incorrect results.

The rate-equation approximation is often used in the description of laser amplifiers. It has recently been recognized[17,18] that this approximation breaks down for the amplifier under certain conditions, for example, in the case of a "swept-gain" amplifier. The polarization of the medium does not instantaneously follow the field, and the smallness of the deviation is a measure of the validity of the approximation. It is interesting to note that Serber and Townes[19] solved, in the linear approximation, the problem of purely temporal growth of an undamped amplifier, an extreme case of the breakdown of the rate-equation approximation. The mathematical description is identical to those of purely temporal parametric amplification and purely temporal undamped stimulated light scattering. In general, there is a formal similarity between the stimulated-light-scattering problem described here and the laser amplifier. In particular, the transient problems arising in the swept gain amplifier are very similar to the transient problems of stimulated scattering involving sufficiently short pulses of input radiation.[20] Thus we expect the formalism of Refs. 11–15 to be useful in the linearized swept-gain problem.

We now proceed from the dynamical equations for the classical amplitudes to derive equations of motion for the photon- and material-mode number densities. We could do the same calculation quantum mechanically, going from equations of motion for field operators to equations of motion for intensity operators. However, the classical problem demonstrates the essential features. The quantum problem adds inhomogeneous terms which do not affect the nature of the instabilities.

The amplitude equations[21] are

$$\left(\frac{\partial}{\partial t}+v\,\frac{\partial}{\partial z}\right)a=-\kappa b^* e^{i(\omega_a+\omega_b-\omega)t} \tag{1}$$

and

$$\left(\frac{\partial}{\partial t}+\gamma\right)b^*=-\kappa^* a e^{-i(\omega_a+\omega_b-\omega)t}\,, \tag{2}$$

where a is the Stokes amplitude, b is the material-mode amplitude, ω is the frequency of the input driving field, ω_a and ω_b are the frequencies of the Stokes and material fields, respectively, v is the velocity of Stokes field, and γ is the damping constant of the material excitation. Also, κ is the coupling coefficient which is linear in the input-field amplitude. We have assumed the Stokes field propagates in the $+z$ direction and the medium has negligible linear absorption at the input and Stokes frequencies. The neglect of Stokes damping makes the solutions generally unstable in the sense that there is always exponential growth for nonzero κ. We will also assume for simplicity that $\omega=\omega_a+\omega_b$.

We could solve the coupled linear amplitude equations by the methods described earlier. However, as stated above, we want to derive equations describing the evolution of the photon- and material-mode number densities. From (1) and (2), we obtain

$$\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial z}\right)N_a(t,z)=\left(\frac{\partial}{\partial t}+2\gamma\right)N_b(t,z) \tag{3}$$

$$=|\kappa|^2\int_0^t d\tau\, e^{-\gamma(t-\tau)}[a^*(\tau,z)a(t,z)+a^*(t,z)a(\tau,z)]\,, \tag{4}$$

where $N_a=a^*a$ and $N_b=b^*b$. In deriving (4) we have assumed the input-field amplitude varies sufficiently slowly in time that it can be taken outside the integral. We have also assumed that the initial- ($t=0$) value term which should appear on the right-hand side of (4) is zero or at least dominated by the integral term. Invoking the second requirement implies that we cannot use Eq. (4) (and hence the approximations to it made below) for the early evolution. It is an asymptotically valid approximation for times such that $\gamma t\gg 1$. Since the growth of the waves can generally be characterized by their behavior for $\gamma t\gg 1$, we concentrate our attention in this region. For example, in the case where the fields build up solely from material-mode noise, we may use (4) to describe the evolution after the Stokes field has established its dominance. It should be noted that Eq. (4) has nonlocal character. It is also apparent that (3) and (4) cannot be solved in general without first solving (1) and (2). However, we can obtain a soluble Markovian equation if we assume

$$\left|\frac{\partial}{\partial t}(\ln N_a)\right|\ll\gamma \tag{5}$$

in which case (4) becomes

$$\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial z}\right)N_a=\frac{2|\kappa|^2}{\gamma}N_a\,. \tag{6}$$

The solution to (6) for purely temporal growth[22] has an exponential growth constant

$$\Gamma=2|\kappa|^2/\gamma\,. \tag{7}$$

From (5) and (7) we see that

$$2|\kappa|^2\ll\gamma^2 \tag{8}$$

for (7) to describe the growth. Thus the purely temporal solution that is valid in the Markovian approximation is not correct for arbitrary values of the ratio, $|\kappa|/\gamma$. Also from (3) and (6) we find (repeating the Markovian approximation)

$$N_b=(|\kappa|^2/\gamma^2)N_a\,, \tag{9}$$

therefore $N_b\ll N_a$. In other words the growing mode is primarily Stokes in character. This asymmetry arises because we neglect the Stokes damping but not the phonon damping. Solving Eqs. (1) and (2) for purely temporal growth in the limit $|\kappa|/\gamma \gg 1$, the exponential growth constant becomes $2|\kappa|$, a well-known result for undamped parametric processes.[23] Note that for purely spatial growth the condition (8) no longer applies.

To see more clearly how the present calculation relates to the results of Sparks, we use (1), (2), and (4) to obtain

$$\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial z}\right)N_a(t,z)$$

$$=2|\kappa|^2\int_0^t d\tau\, e^{-\gamma(t-\tau)}\left[N_a(\tau,z)+N_b(\tau,z)+\frac{v}{2}\left(\frac{1}{\kappa}\frac{\partial a}{\partial z}(\tau,z)b(\tau,z)+\frac{1}{\kappa^*}\frac{\partial a^*}{\partial z}(\tau,z)b^*(\tau,z)\right)\right]. \quad (10)$$

In the Markovian approximation we have

$$\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial z}\right)N_a =\frac{2|\kappa|^2}{\gamma}\left\{N_a+N_b+\frac{v}{2}\left[\frac{1}{\kappa}\frac{\partial a}{\partial z}b+\frac{1}{\kappa^*}\frac{\partial a^*}{\partial z}b^*\right]\right\}. \quad (11)$$

The spatial derivatives on the right-hand side make the result differ from that of Sparks even in the Markovian approximation.[24] If the growth is purely temporal then (3) and (11) can be written

$$\frac{dN_a}{dt}=\left(\frac{d}{dt}+2\gamma\right)N_b=\frac{2|\kappa|^2}{\gamma}(N_a+N_b). \quad (12)$$

Since for the Markovian approximation we must have

$$\left|\frac{d}{dt}(\ln N_a)\right|,\ \left|\frac{d}{dt}(\ln N_b)\right|\ll\gamma, \quad (13)$$

we readily find, as before, that $2|\kappa|^2\ll\gamma^2$, $N_b \ll N_a$, and the growth constant is given by (7).

From (12) we can write the material-mode rate equation as

$$\frac{dN_b}{dt}=\frac{2|\kappa|^2}{\gamma}(N_a+N_b)-2\gamma N_b. \quad (14)$$

It has been stated[25] that this equation shows the explosive growth of the material excitation can occur with $N_a\ll N_b$. For this to be correct we would require that $\gamma^2<|\kappa|^2$, a result inconsistent with the derivation.

For the case of pure spatial growth the term in square brackets in (11) cancels the N_b term for all $|\kappa|/\gamma$. It can also be shown that one can neglect the term in square brackets relative to the N_a term when $2|\kappa|^2\ll\gamma^2$. Without making the consistent approximation $N_b\ll N_a$, Eq. (11) becomes

$$v\frac{dN_a}{dz}=2\gamma N_b=\frac{2|\kappa|^2}{\gamma}(N_a+N_b). \quad (15)$$

This is Sparks' result for spatial growth and leads to an exponential growth constant α of the form

$$\alpha=(2|\kappa|^2/v\gamma)/(1-|\kappa|^2/\gamma^2). \quad (16)$$

Since we have the constraint that $2|\kappa|^2\ll\gamma^2$, (16) becomes

$$\alpha=2|\kappa|^2/v\gamma, \quad (17)$$

which is also the exact solution to the spatial-growth problem. The supposed singularity in α when $|\kappa|/\gamma=1$ does not occur since this ratio of parameters is inconsistent with the derivation of (15).

In some cases the material excitation spectrum is inhomogeneously broadened. If the Markovian approximation used in deriving (6) obtains and the inhomogeneous linewidth is much wider than γ, the inhomogeneous gain may be found by multiplying the right-hand side of (6) by $\gamma g(\omega_b)$, where g is the inhomogeneous line-shape function.

Even without damping of the material excitation, inhomogeneous broadening can lead to a similar result. The nature of this calculation is described in what follows. Each phonon frequency has a different time dependence in the kernel of the non-Markovian rate equation for the Stokes photon density. If the frequencies of the material modes are sufficiently closely spaced the sum of the oscillating terms gives rise to a strongly time localizing kernel, the rate equation becomes Markovian and the transition rate is proportional to the number of Stokes photons. This is, of course, the essence of the traditional golden rule transport equation calculation based on the assumption that one subsystem (Stokes field) weakly perturbs another subsystem with a continuous energy spectrum (material modes).

ACKNOWLEDGMENT

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[21]It is apparent these equations are appropriate for dispersionless or weakly dispersive phonons in the harmonic approximation. They can also be used to describe stimulated scattering from a noninteracting set of degenerate two-level atoms (the second-order coupling term is assumed to involve nonresonant intermediate states) whose response can be linearized in the driving term. In this case b is proportional to the induced polarizability of the atoms. The constraint that the atoms' response remains linear requires that N_b be much smaller than the density of the initial population difference between upper and lower states.

[22]The appropriate space-time character of the growth is determined by the shape of the input field pulse and the initial conditions on the Stokes and material fields. For the present purposes we assume that these conditions can be adjusted so as to give either pure temporal or pure spatial growth. The conclusions made here are not changed for cases where mixed space-time growth occurs.

[23]It is often useful to describe the situation when $|\kappa| < \gamma$ as the moderately coupled regime and when $|\kappa| > \gamma$ as the strongly coupled regime. If the Stokes damping, γ_a, is not neglected but $\gamma \gg \gamma_a$ the case when $\gamma_a \gamma > |\kappa|^2$ can be called weak coupling and the case when $\gamma^2 > |\kappa|^2 > \gamma_a \gamma$ can be called moderate coupling. The instability threshold then occurs when $\gamma_a \gamma = |\kappa|^2$.

[24]If it is assumed, as Sparks does, that the golden rule is valid then we find the total rate term consists of the difference between the rate of emission of Stokes photons and material mode quanta and the rate of absorption of these quanta. This difference is of the form

$$(N_a + 1)(N_b + 1)N_l - N_a N_b (N_l + 1),$$

where N_l is the input photon density. When $N_l \gg N_a + N_b \gg 1$, the above reduces to $N_l (N_a + N_b)$ and we can absorb N_l into the rate constant.

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