Temporal and Spatial Gain in Stimulated Light Scattering*

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The theory of scattered light waves with exponential temporal and spatial gain is investigated in the linearized approximation for a number of different cases of material excitation. Regions of weakly coupled gain (the rate-equation approximation) and strongly coupled gain are found. A group velocity for the scattered wave is introduced to facilitate the analysis. Gain is found for the case where the material excitations lose their uncoupled character.

I. INTRODUCTION

Since the first observation of stimulated light scattering in 1962,[1,2] an extensive experimental literature has developed concerning stimulated Raman scattering, electrostrictive and absorptive Brillouin and entropy (Rayleigh-peak) scattering, and Rayleigh-wing scattering.[3] Part of this experimental work has been concerned with verification of the theoretical foundations. We wish to extend and amplify the theory of the growth of the scattered waves, and we hope these results suggest ways in which effective experimental verification may be obtained of some of the relatively untested theoretical conclusions.

Let us at the outset list the specific aspects of the theoretical treatment to which we shall address ourselves:

(a) the regimes of temporal and spatial growth of the scattered waves and the connection between these regimes, including the role of boundary conditions;

(b) the range of validity of the "rate-equation approximation" for the material motion, and characterization of the growth in the weak-coupling limit (rate-equation approximation, where the damping mechanism for the material motion balances the driving term due to the electromagnetic waves) and the strong-coupling limit (where the growth due to the driving term greatly exceeds the decay due to damping);

(c) the utility of the concept of a group velocity for the mixed material and scattered electromagnetic modes;

(d) the situation in which the input field is so intense that the material modes lose their uncoupled character.

A brief review of past theoretical treatments will give some perspective to this work. Most treatments of stimulated scattering (including the present work) linearize, in the scattered field and material coordinates, the system of coupled waves. This amounts to assuming the strong field is essentially unaffected by the growth of the scattered waves, a possibility which is realizable under certain experimental conditions. To further simplify the problem, the laser field is often taken to be a monochromatic plane wave, in which case the laser field amplitude is a fixed parameter in the linearized equations, hence the often used description of stimulated scattering as "a parametric process." The search for and characterization of exponentially growing solutions to the equations for the coupled linearized waves is described as "an instability analysis."

Exponential growth in stimulated scattering was pointed out by Bloembergen.[4] Garmire, Pandarese, and Townes[5] also considered exponential as well as linear gain and discussed the molecular model for stimulated scattering. The theory, further refined,[6] included the treatment of the Stokes–anti-Stokes interaction away from exact phase matching. Kroll[7] showed that in spatial growth one could include the effect of spatial nonlocality (the finite velocity of the material excitation) in the response of the material system, the locality assumption being similar to the rate-equation approximation in the time domain. The assumption of locality has been found to be an excellent approximation in spatial growth except for sound waves (Brillouin scattering), where incident field intensities could be achieved which gave rise to instabilities of the nonconvective type. The theory was also extended into the region where the growth depended on the time duration of the laser pulse. In this part of the treatment the assumption was made that the light velocity was effectively infinite, which leads to what will be defined as a strong-coupling space-time growth and the absence of a region of simple temporal growth.

Pure temporal growth was treated by Chiao[8] and Pine[9] in connection with the production of stimulated scattering in resonators. Their analyses gave both the strong- and weak-coupling limits. Brueckner and Jorna[10] theoretically proposed absorptive light scattering and found results similar to those of

Chiao and Pine in the strongly coupled limit, but in the absence of a resonator. They also introduced the group velocity concept for the scattered waves and obtained results analogous to those of Kroll for strongly coupled mixed space-time growth.

The problem of purely spatial growth for absorptively coupled scattering was analyzed by Herman and Gray.[11] They attempted to include the effect of laser linewidth. But as has been pointed out earlier,[12] the lack of monochromaticity in the laser does not necessarily decrease gain except through the connection between the lack of monochromaticity and temporal spiking behavior or laser pulse duration and the effect of color dispersion in causing a phase mismatch from one end of the spectral range to the other.[13] Furthermore, in the particular case of an uncertainty-limited laser pulse in which the linewidth is the reciprocal of the pulse duration and where the laser linewidth is greater than the linewidth of the material excitation, Herman and Gray would predict that the gain in an amplifier cell is proportional to the first power of the cell length times the pulse duration. This is in disagreement with earlier results[7,10] as well as the present work.

Further analysis has been given for certain cases of transient stimulated scattering for a square input pulse.[14,15] The connection between the various results described above has not been clearly developed, and it is one of the aims of the present work to carry out this development.

There are some obvious drawbacks to the linearized constant laser parameter treatment. To study the time-dependent problem we assume that the laser field is represented as a square pulse times an oscillating field. As indicated above, this makes the laser field a parameter in the coupled equations of the weak material and scattered electromagnetic waves. Nevertheless, the laser field is not monochromatic, but has its spectral intensity distributed like $(\sin x)^2/x^2$ about the central frequency. However, experimentally the laser pulse amplitudes are varying in a more or less smooth fashion, and the parameter coupling the weak-wave equations is time dependent. Therefore the weak-wave growth has to adjust itself to this parameter variation. To take this into account complicates the problem greatly; the equations must be treated by integral equation techniques[16,17] and numerical integration.[17] In the strongly coupled regime it is unlikely that smooth laser amplitude variations affect many of the features of the results, the scattered waves responding to the average field for a time equal to the pulse length. In the weakly coupled regime this is apparently not the case, particularly when the pulse gets to be shorter than the relaxation time.

Another drawback is the inappropriateness of this technique to the situation often encountered experimentally, where the laser field is markedly attenuated by the scattering process. The most reasonable situation in which to test the linearized theory is an amplifier experiment with very weakly scattered wave inputs, so that even if there is large gain, there is little attenuation of the laser. Treatments have been given[18] of the coupled-wave problem, including laser amplitude variation as well as its attenuation by stimulated scattering, which have been limited to the situation in which the rate-equation approximation is valid.

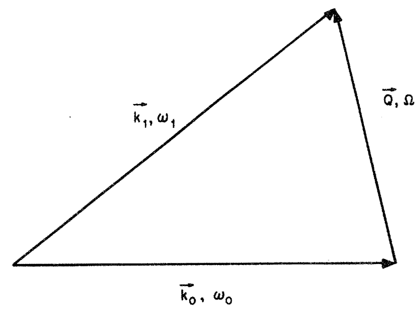

FIG. 1. Propagation vector diagram for light scattering.

II. KINEMATICS

For scattering angles (angles between the propagation directions of the incident and scattered light) larger than a few degrees we need consider only the scattering process shown in Fig. 1. Here, $\vec{k}_0$, ω_0 are the wave vector and frequency of the strong input wave; similarly, we have $\vec{k}_1$, ω_1 for the scattered electromagnetic wave, and $\vec{Q}$, Ω for the material excitation. The frequency- and momentum-matching conditions are

$$\omega_1 = \omega_0 + \Omega \, , \tag{2.1}$$

$$\vec{k}_1 = \vec{k}_0 + \vec{Q} \, . \tag{2.2}$$

When $\mathrm{Re}\,\Omega$ is positive we have anti-Stokes scattering, and when $\mathrm{Re}\,\Omega$ is negative we have Stokes scattering.

We note that when $\mathrm{Re}\,\Omega$ is positive, the material wave travels in the direction indicated by the arrow in Fig. 1, while when $\mathrm{Re}\,\Omega$ is negative, the material wave travels in the opposite direction. Thus in backscattering ($\vec{k}_1$ opposite to $\vec{k}_0$) the material wave excited in anti-Stokes scattering travels in a direction opposite to $\vec{k}_0$, while for Stokes scattering the material wave travels in the $\vec{k}_0$ direction.

It is often useful to write the scattered frequency and wave vector as

$$\omega_1 = \omega_1^0 + \nu \, , \tag{2.3}$$

$$\vec{k}_1 = \vec{k}_1^{\,0} + \vec{q} \, , \tag{2.4}$$

where

$$|\vec{k}_1^{\,0}| = k_1^0 = \omega_1^0 [\epsilon(\omega_1^0)]^{1/2}/c \, . \tag{2.5}$$

$\vec{k}_1^0$ and ω_1^0 satisfy the uncoupled (and undamped) electromagnetic dispersion relations, and $\vec{k}_1^0 - \vec{k}_0$ and $\omega_1^0 - \omega_0$ satisfy the uncoupled (and undamped) material dispersion relations. Since we have $q \ll k_1^0$, the scattering angle is approximately given by

$$\cos\theta \equiv \frac{\vec{k}_0 \cdot \vec{k}_1}{k_0 k_1} \simeq \frac{\vec{k}_0 \cdot \vec{k}_1^0}{k_0 k_1^0} \ . \tag{2.6}$$

It is useful to discuss the effect of a planar input boundary for the scattered waves. We suppose that the boundary conditions determine the direction of $\mathrm{Im}\vec{q}$ to be normal to the boundary. We can satisfy the phase variations at the boundary by properly choosing the component of $\vec{k}_1^0$ normal to $\mathrm{Im}\vec{q}$. Thus, without loss of generality, we can take $\mathrm{Re}\vec{q} \parallel \mathrm{Im}\vec{q}$. We indicate the angle ϕ between $\vec{q}$ and $\vec{k}_1^0$ as

$$\mu = \cos\phi = \mathrm{Im}\vec{q} \cdot \vec{k}_1^0 / |\mathrm{Im}\vec{q}\,| k_1^0 \ , \tag{2.7}$$

where, for all reasonable experimental applications, ϕ is zero or very small.

For small values of the scattering angle θ we must consider the kinematics represented in Fig. 2, which shows the Stokes–anti-Stokes interaction. As the figure is drawn, $\vec{k}_1$, ω_1 is the upshifted scattered wave and $\vec{k}_2$, ω_2 is the downshifted scattered wave which satisfies the frequency and momentum conditions

$$\omega_2 = \omega_0 - \Omega \ , \tag{2.8}$$

$$\vec{k}_2 = \vec{k}_0 - \vec{Q} \ . \tag{2.9}$$

This small-angle interaction has important consequences for the gain in this region. We must treat the scattered waves referred to in the figure as part of a single coupled-wave problem. We also introduce the uncoupled frequency and wave vector magnitude for the second scattered wave:

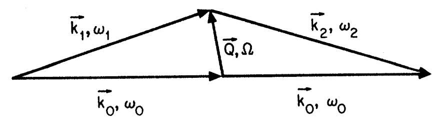

FIG. 2. Propagation vector diagram for light scattering, including the Stokes–anti-Stokes interaction.

$$\omega_2^0 = 2\omega_0 - \omega_1^0 \ , \tag{2.10}$$

$$k_2^0 = \omega_2^0 [\epsilon(\omega_2^0)]^{1/2}/c \ . \tag{2.11}$$

III. ELECTROMAGNETIC PART OF DISPERSION RELATION

We seek solutions of the propagation equations for the coupled system of electromagnetic waves and material excitations. We begin with the transverse Maxwell wave equation containing a nonlinear polarization $\vec{P}^{NL}$ cubic in the electric field:

$$-\vec{\nabla} \times (\vec{\nabla} \times \vec{E}) - \frac{1}{c^2}\frac{\partial^2 \vec{D}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 \vec{P}^{NL}}{\partial t^2} \ , \tag{3.1}$$

where $\vec{E}$ is the electric field and $\vec{D}$ the linear electric displacement vector. We restrict ourselves to situations where $\vec{P}^{NL}$, and the scattered and incident fields are parallel. This is, of course, always the case when the material excitations are hydrodynamic in nature (i.e., pressure and density change in liquids and gases), and the laser light is polarized perpendicular to the plane of the scattering. The electric field is written as

$$\vec{E} = \tfrac{1}{2}\hat{e}\left(\mathcal{E}_0 e^{i(\vec{k}_0\cdot\vec{r} - \omega_0 t)} + \mathcal{E}_1 e^{i(\vec{k}_1\cdot\vec{r} - \omega_1 t)} + \mathcal{E}_2 e^{i(\vec{k}_2\cdot\vec{r} - \omega_2 t)} + \text{c.c.}\right) \ , \tag{3.2}$$

$\mathcal{E}_0$ being the amplitude of the strong field and $\mathcal{E}_1$ and $\mathcal{E}_2$ the amplitude of the weak fields. Assuming $|\mathcal{E}_0| \gg |\mathcal{E}_1|, |\mathcal{E}_2|$, we may linearize (3.1) in the weak fields to obtain the dispersion relation

$$\begin{aligned}
&[(\vec{k}_0+\vec{Q})^2c^2 - (\omega_0+\Omega)^2\epsilon(\omega_0+\Omega) - 4\pi(\omega_0+\Omega)^2\chi^{(3)}(\vec{k}_0+\vec{Q}, \omega_0+\Omega)|\mathcal{E}_0|^2] \\
&\quad\times[(\vec{k}_0-\vec{Q})^2c^2 - (\omega_0-\Omega)^2\epsilon(\omega_0-\Omega) - 4\pi(\omega_0-\Omega)^2\chi^{(3)*}(\vec{k}_0-\vec{Q}, \omega_0-\Omega)|\mathcal{E}_0|^2] \\
&\quad= (4\pi)^2(\omega_0+\Omega)^2(\omega_0-\Omega)^2\chi^{(3)}(\vec{k}_0+\vec{Q}, \omega_0+\Omega)\chi^{(3)*}(\vec{k}_0-\vec{Q}, \omega_0-\Omega)|\mathcal{E}_0|^4 \ .
\end{aligned} \tag{3.3}$$

The third-order nonlinear susceptibility $\chi^{(3)}$ is defined by

$$2\frac{\mathcal{P}'_{NL}(\vec{k}_0 \pm \vec{Q}, \omega_0 \pm \Omega)}{\mathcal{E}(\vec{k}_0 \pm \vec{Q}, \omega_0 \pm \Omega)} = \chi^{(3)}(k_0 \pm Q, \omega_0 \pm \Omega)|\mathcal{E}_0|^2 \ , \tag{3.4}$$

where $\mathcal{P}'_{NL}(\vec{k}_0 \pm \vec{Q}, \omega_0 \pm \Omega)$ is the Fourier component of the nonlinear polarization arising from the $\vec{Q}$, Ω component of the material motion proportional to $\mathcal{E}(\vec{k}_0 \pm \vec{Q}, \omega_0 \pm \Omega)$. For the material excitations considered here we may make the reasonable approximation that

$$\chi^{(3)}(\vec{k}_0+\vec{Q}, \omega_0+\Omega) = \chi^{(3)*}(\vec{k}_0-\vec{Q}, \omega_0-\Omega) \ . \tag{3.5}$$

For simplicity, and since $\chi^{(3)}$ usually depends only weakly on ω_0 and k_0, we write

$$K(Q, \Omega) = 2\pi\chi^{(3)}(\vec{k}_0+\vec{Q}, \omega_0+\Omega)/\epsilon(\omega_0+\Omega) \ , \tag{3.6}$$

noting that the material excitations in isotropic materials depend on the magnitude of Q, not its direction. The coupling function $K(Q, \Omega)$ includes the dispersion relation for the material system and will be derived in Sec. V for various material excitations and couplings. In most situations treated in the literature Ω in $K(Q, \Omega)$ is taken to be real. For the transient or temporal growth case, this approach is no longer valid, as we shall see subsequently.

Assuming ν and q are small compared to optical frequencies and propagation constants, we may approximate (3.3) by

$$\left(\mu q - \frac{\nu}{v_1} - k_1 K(Q,\Omega)|\mathcal{E}_0|^2\right) \times \left(\frac{4k_1^0 k_0}{k_2^0}\sin^2\frac{\theta}{2} - \Delta k - \frac{(2\vec{k}_0 - \vec{k}_1^0)\cdot\vec{q}}{k_2^0} + \frac{\nu}{v_2} - k_2^0\frac{\epsilon(\omega_1^0)}{\epsilon(\omega_2^0)}K(Q,\Omega)|\mathcal{E}_0|^2\right) = \frac{k_1^0 k_2^0 \epsilon(\omega_1^0)}{\epsilon(\omega_2^0)} K^2(Q,\Omega)|\mathcal{E}_0|^4 , \quad (3.7)$$

where q, ν, and μ have been introduced using (2.3), (2.4), and (2.7); in addition, the electromagnetic group velocity v_i is given by

$$v_i = \frac{c\epsilon^{1/2}(\omega_i^0)}{\epsilon(\omega_i^0) + \frac{1}{2}\omega_i^0(\partial\epsilon/\partial\omega_i^0)} \quad (3.8)$$

and Δk is given by

$$\Delta k = [4k_0k_1^0 - 4k_0^2 + (k_2^0)^2 - (k_1^0)^2]/2k_2^0 . \quad (3.9)$$

Δk is positive or zero in all experimental situations studied to date.

For large angles such that

$$\frac{4k_1^0 k_0}{k_2^0}\sin^2\tfrac{1}{2}\theta \gg \Delta k + k_2^0\frac{\epsilon(\omega_1^0)}{\epsilon(\omega_2^0)}K(Q,\Omega)|\mathcal{E}_0|^2 ,$$

we may approximate (3.7) by

$$\mu q - \nu/v_1 - k_1^0 K(Q,\Omega)|\mathcal{E}_0|^2 = 0 ; \quad (3.10)$$

in other words the Stokes–anti-Stokes coupling is unimportant.

For Brillouin, entropy, and Rayleigh-wing scattering, ω_1^0, ω_2^0, and ω_0 are so nearly equal that we may take

$$\epsilon(\omega_1^0) = \epsilon(\omega_2^0) = \epsilon(\omega_0) .$$

This implies $\Delta k = 0$ and allows one to set $k_1^0 = k_2^0 = k_0$ and $\omega_1^0 = \omega_2^0 = \omega_0$ elsewhere in the expression. Equation (3.7) then takes the form

$$\left(\mu q - \frac{\nu}{v_0} - k_0 K(Q,\Omega)|\mathcal{E}_0|^2\right) \times \left(4k_0\sin^2\frac{\theta}{2} - \frac{(2\vec{k}_0 - \vec{k}_1^0)\cdot\vec{q}}{k_0} + \frac{\nu}{v_0} - k_0 K(Q,\Omega)|\mathcal{E}_0|^2\right) = k_0^2 K^2(Q,\Omega)|\mathcal{E}_0|^4 . \quad (3.11)$$

For the angles where the Stokes–anti-Stokes interaction is significant, $1 \gg \theta$, and we may further approximate the dispersion relation as

$$[q - \nu/v_0 - k_0 K(Q,\Omega)|\mathcal{E}_0|^2] \times [k_0\theta^2 - q + \nu/v_0 - k_0 K(Q,\Omega)|\mathcal{E}_0|^2] = k_0^2 K^2(Q,\Omega)|\mathcal{E}_0|^4 . \quad (3.12)$$

We assume, in order to make the interaction distance a maximum in this small-angle case, that $\vec{q}$ lies along $\vec{k}_0$ and that $\mu \approx 1$.

Finally, for large angles, we may approximate (3.10) for Brillouin, Rayleigh-wing, and absorptive Rayleigh as

$$\mu q - \nu/v_0 - k_0 K(Q,\Omega)|\mathcal{E}_0|^2 = 0 . \quad (3.13)$$

Note that when we replace v_1 by v_0 and k_1 by k_0, (3.10) becomes identical with (3.13).

IV. MATERIAL MOTION AND COUPLING TO SCATTERED ELECTROMAGNETIC WAVE

In this section we discuss various material motions which can be excited in light scattering in liquids, together with their coupling to the electromagnetic field. We will discuss electrostrictively and absorptively induced Brillouin and entropy (Rayleigh-peak) scattering, Rayleigh-wing scattering, and Raman scattering.

A. Electrostrictively and Absorptively Induced Brillouin and Entropy (Rayleigh-Peak) Scattering.

We begin with the standard equations of linearized hydrodynamics. The linearized equation for the increase in entropy density is written as

$$\rho_0 T_0 \frac{\partial s_1}{\partial t} = \frac{C_v(1-\gamma)}{\beta}\frac{\partial\rho_1}{\partial t} + \rho_0 C_v\frac{\partial\theta_1}{\partial t} + \rho_0 T\left(\frac{\partial\epsilon}{\partial T}\right)_\rho \frac{\partial}{\partial t}\left(\frac{E^2}{8\pi}\right) = \frac{\alpha E^2}{4\pi} n_0 c + \kappa\nabla^2\theta_1 . \quad (4.1)$$

In (4.1) ρ_1 is the induced density charge (to first order in E^2), ρ_0 the zeroth-order density, θ_1 the induced temperature change, κ the thermal conductivity, C_v (C_p) the specific heat at constant volume (pressure), $\gamma = C_p/C_v$, β the volume expansion coefficient, and α is the absorption constant per unit length. The first term on the right-hand side (rhs) represents the energy absorbed from the light wave per unit volume, while the second term is the heat flow into a volume element due to thermal conductivity. We also use the sound propagation equation

$$\left(\frac{\partial^2}{\partial t^2} - \frac{\eta}{\rho_0}\frac{\partial}{\partial t}\nabla^2 - \frac{v_s^2}{\gamma}\nabla^2\right)\rho_1 - \frac{\beta\rho_0 v_s^2}{\gamma}\nabla^2\theta_1 = -\rho_0\left(\frac{\partial\epsilon}{\partial\rho}\right)_T \nabla^2\frac{E^2}{8\pi} . \quad (4.2)$$

Here v_s is the velocity of sound, which is related to the other coefficients by

$$v_s^2 = (\gamma - 1)\, C_p/\beta \ ,$$

and η is the viscosity. The first term on the rhs of (4.2) is the gradient of the electrostrictive force. The dielectric constant change to first order in E^2 is given in terms of ρ_1 and θ_1 by

$$\delta\epsilon = \left(\frac{\partial\epsilon}{\partial\rho}\right)_T \rho_1 + \left(\frac{\partial\epsilon}{\partial T}\right)_\rho \theta_1 \ , \qquad (4.3)$$

which in turn gives rise to the nonlinear polarization

$$P^{\mathrm{NL}} = \frac{\delta\epsilon\, E}{4\pi}\left(\frac{\epsilon_0+2}{3}\right)^2 . \qquad (4.4)$$

Fourier transforming (4.1)–(4.4), and using (3.4) and (3.6), we find

$$16\pi\epsilon_0 D(Q,\ \Omega)\, K(Q,\ \Omega) = \left(\frac{\partial\epsilon}{\partial\rho}\right)_T \left\{\frac{\beta\rho_0 v_s^2}{\gamma}\left[\alpha n_0 c + \frac{i\Omega}{2}\rho_0 T\left(\frac{\partial\epsilon}{\partial T}\right)_\rho\right] - \frac{1}{2}(\kappa Q^2 - i\rho_0 C_v\Omega)\rho_0\left(\frac{\partial\epsilon}{\partial\rho}\right)_T\right\} Q^2$$
$$- \left(\frac{\partial\epsilon}{\partial T}\right)_\rho \left\{\left(\frac{v_s^2 Q^2}{\gamma} - \Omega^2 - \frac{i\Omega Q^2\eta}{\rho_0}\right)\left[\alpha n_0 c + \frac{i\Omega}{2}\rho_0 T\left(\frac{\partial\epsilon}{\partial T}\right)_\rho\right] - \frac{i\Omega C_v(\gamma-1)}{2\beta}\rho_0\left(\frac{\partial\epsilon}{\partial\rho}\right)_T Q^2\right\}\left(\frac{\epsilon_0+2}{3}\right)^2 , \qquad (4.5)$$

where

$$D(Q,\ \Omega) = \left[\frac{i\Omega C_v(\gamma-1)}{\gamma}\rho_0 v_s^2 Q^2 - (\kappa Q^2 - i\rho_0 C_v\Omega) \times\left(\frac{v_s^2 Q^2}{\gamma} - \Omega^2 - \frac{i\Omega Q^2\eta}{\rho_0}\right)\right] . \qquad (4.6)$$

If only a fraction f of the absorbed energy becomes thermalized (rather than reradiated) and if a relaxation time τ is required for the absorbed flux to be transferred to thermal kinetic energy, then a factor $f/(1+i\Omega\tau)$ should multiply α.

B. Rayleigh-Wing Scattering

We consider next the alignment of axially symmetric anisotropically polarizable molecules. The fractional alignment $s = \langle\cos^2\theta - \frac{1}{3}\rangle$ (where θ is the angle between the axis of symmetry and the electric field) obeys the equation

$$\tau\frac{ds}{dt} + s = \frac{4}{45}\frac{\Delta a E^2}{k_B T}\left(\frac{\epsilon_0+2}{3}\right)^2 , \qquad (4.7)$$

where τ is one-third the Debye relaxation time, Δa is the difference in polarizability between the axis of symmetry and the directions normal to it, k_B is Boltzmann's constant, and each $\frac{1}{3}(\epsilon_0+2)$ is a local-field correction factor. The change in dielectric constant due to alignment is given by

$$\delta\epsilon = 4\pi N[\tfrac{1}{3}(\epsilon_0+2)]^2\Delta a s \ , \qquad (4.8)$$

where N is the number of molecules per unit volume Fourier transforming (4.4), (4.7), and (4.8) and using (3.4) and (3.6), we find

$$K(Q,\ \Omega) = \frac{4\pi}{45}\frac{N(\Delta a)^2}{\epsilon_0 k_B T}\left(\frac{\epsilon_0+2}{3}\right)^4 \frac{1}{1-i\Omega\tau} . \qquad (4.9)$$

C. Stimulated Raman Scattering

In stimulated Raman scattering, we may write the equation of motion for the vibrational coordinate x as

$$\frac{d^2x}{dt^2} + 2\Gamma\frac{dx}{dt} + \omega_v^2 x = \frac{1}{2m}\frac{\partial a}{\partial x_0}E^2\ \frac{\epsilon_0+2}{3}\ \frac{\epsilon_1+2}{3} \ , \qquad (4.10)$$

where Γ is the damping constant, ω_v is the vibrational frequency, m is the mass, and $\partial a/\partial x_0$ is the variation in the polarizability with respect to the vibrational coordinate at the equilibrium position x_0. The nonlinear polarization induced by the vibration is given by

$$P^{\mathrm{NL}} = Nx\frac{\partial a}{\partial x_0}E\ \frac{\epsilon_0+2}{3}\ \frac{\epsilon_1+2}{3} \ . \qquad (4.11)$$

Fourier transforming (4.10) and (4.11) and once again using (3.4) and (3.6), we find

$$K(Q,\ \Omega) = \frac{\pi N}{2\epsilon_0 m}\left(\frac{\partial a}{\partial x_0}\right)^2\left(\frac{\epsilon_0+2}{3}\right)^2\left(\frac{\epsilon_1+2}{3}\right)^2 \times\frac{1}{\Omega^2 + 2i\Gamma\Omega - \omega_v^2} . \qquad (4.12)$$

V. GENERAL APPROACH TO INSTABILITIES

In this section we give a general treatment of the problem of temporal as well as spatial instabilities in stimulated light scattering.[19] This is done in order to understand the transient region of growth of the scattered waves and the transition from this region to the steady state, where the growth is purely spatial.

The study of these instabilities begins with Eq. (3.7), often referred to as the dispersion relation. It determines q as a function of ν. Both q and ν are taken to be complex and we write

$$q = q' + iq'' \ , \qquad (5.1)$$

$$\nu = \nu' + i\nu'' \ . \qquad (5.2)$$

For discussing steady-state problems one assumes $\nu'' = 0$. Spatial growth of instabilities occurs for those values of ν' for which $q''(\nu')$ is negative, and one typically seeks a maximum growth by maximizing $|q''(\nu')|$ as a function of ν'. The steady-state gain yields an appropriate measure of instability only when the effective interaction time is sufficiently long to permit the disturbance of highest spatial gain to propagate from one end of the stimulated scattering region to the other. For shorter interaction times we proceed as follows.

Equation (3.7) can be thought of as determining q', q'' as functions of ν', ν''. For reasons which will become clear later, the additional condition

$$\frac{\partial q''}{\partial \nu'} = 0 \tag{5.3}$$

is imposed. This condition together with Eq. (3.7) determines q', q'', and ν'' as functions of ν'. Alternatively, it is often convenient to introduce a group velocity

$$v_g \equiv \left(\frac{\partial q'}{\partial \nu'}\right)^{-1} , \tag{5.4}$$

and to regard all four variables q', q'', ν', and ν'' as functions of v_g. Equation (5.4) then implies

$$v_g = \frac{d\nu''}{dv_g}\bigg/\frac{dq''}{dv_g} . \tag{5.5}$$

The response (scattered electromagnetic field or material excitation) at a distance z and time interval t after an initial disturbance localized in space and time can be expressed in the following simplified manner:

$$\mathcal{R}(z, t) = \int d\nu \int dq \frac{e^{i(qz-\nu t)}}{\mathcal{D}(q, \nu)} . \tag{5.6}$$

$\mathcal{D}(q, \nu)$ vanishes when q, ν satisfy the dispersion relation (3.7) for the coupled scattered electromagnetic and material systems. We have factored out the rapid variations proportional to ω_0 and k_0 and the variation in the plane perpendicular to $\vec{q}$, assuming it is determined only by boundary conditions. Integration over q gives an expression of the form

$$\mathcal{R}(z, t) = \int_{c_\nu} d\nu\, e^{i[q(\nu)z-\nu t]}\mathcal{G}(\nu) , \tag{5.7}$$

where $\mathcal{G}(\nu)$ is generally a function of ν slowly varying compared to the variations in ν of the exponent for some asymptotic region of the z, t plane.

It is convenient to study the asymptotic behavior of $\mathcal{R}(z, t)$ along a line in the z, t plane, denoted for the moment by $t = \alpha z$. The asymptotic behavior along this ray is then determined by the saddle-point condition

$$\frac{d}{d\nu}[q(\nu) - \alpha\nu]z = 0 . \tag{5.8}$$

In other words, along the line $t = \alpha z$, the dominant asymptotic contribution to the amplitude of the scattered field comes from modes in a region of q, ν space near the mode defined by (5.8) and the dispersion relation (3.7). Hence, the scattered intensity is related to the growth constant of this mode. This condition, together with the Cauchy-Riemann relations, yields

$$\alpha = \frac{\partial q''}{\partial \nu''} = \frac{\partial q'}{\partial \nu'} = v_g^{-1} , \qquad \frac{\partial q'}{\partial \nu''} = \frac{\partial q''}{\partial \nu'} = 0 . \tag{5.9}$$

We conclude, therefore, that the gain G for the intensity of the scattered light along the line $z = v_g t$ in the z, t plane is given by

$$\ln G = 2(\nu'' t - q'' z) , \tag{5.10}$$

where ν'' and q'' are functions of v_g determined by Eqs. (3.7), (5.3), and (5.4). The dominant instability is found by maximizing Eq. (5.10) as a function of v_g, subject to the constraint that values of z and t be chosen consistent with the requirements that growth take place within the stimulated scattering region while the laser pulse is present. The instability is convective when v_g is finite, in other words, for a fixed spatial point the dominant disturbance can decay with time. The constraints are thus characterized by $z \le l_c$, where l_c is the cell length and $t \le t_e$, where t_e is the effective interaction time. The effective interaction time t_e is different from the pulse duration t_p because of the finite rate of propagation of the laser pulse through the medium. Thus, for example, a disturbance initiated by the head of the laser pulse and propagating in the same direction will be driven until overtaken by the tail of the pulse (or until it leaves the medium). We have, therefore,

$$t_e = \frac{v_0 t_p}{v_0 - v_g \cos\theta} . \tag{5.11}$$

There are several possible asymptotic regions in which it is interesting to examine the saddle-point exponent. These regions are shown schematically in Fig. 3. One limit occurs when the pulse is long compared with the time it takes a disturbance to propagate over the dimensions of the system. In this limit $t_e > l_c v_g^{-1}$, in which case we write $t = z v_g^{-1}$, and for maximum gain, let z be its maximum value l_c. Therefore, we obtain

$$\ln G = 2(\nu'' v_g^{-1} - q'') l_c . \tag{5.12}$$

This result reflects the fact that in this limit t is restricted to the time it takes a disturbance to propagate from one end of the nonlinear medium to the other. This is the so-called steady-state region.

Further maximization is obtained by requiring the factor $(\nu'' v_g^{-1} - q'')$ to be an extremum. Differentiating with respect to v_g, we have, using (5.5),

$$\frac{d}{dv_g}(\nu'' v_g^{-1} - q'') = -\frac{\nu''}{v_g^2} , \tag{5.13}$$

so that the point $\nu'' = 0$ is an extremum. Differentiating once more, we find

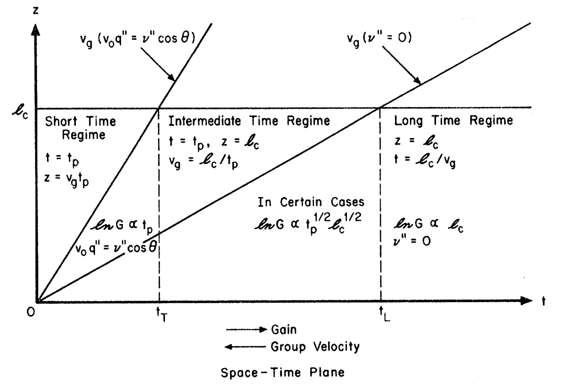

FIG. 3. Time domains in stimulated light scattering for temporal, spatial, and mixed temporal and spatial growth. The case of strong coupling is shown, so that t_T and t_L are well separated.

$$\frac{d^2}{dv_g^2}(\nu'' v_g^{-1} - q'')\bigg|_{\nu''=0} = -\frac{1}{v_g^2}\frac{d\nu''}{dv_g}\bigg|_{\nu''=0} . \qquad (5.14)$$

We then have that the condition $d\nu''/dv_g|_{\nu''=0} > 0$ has a maximum in gain at $\nu'' = 0$. Note that in the steady state we require that $t_e > l_c v_{g,L}^{-1}$, where

$$v_{g,L} = v_g(\nu'' = 0) . \qquad (5.15)$$

Therefore, we can define a steady-state time

$$t_L = \frac{v_0 - v_{g,L}\cos\theta}{v_0 v_{g,L}}\, l_c \qquad (5.16)$$

which t_p must exceed in order to be in the steady-state region.

Another limit occurs when the length of active medium in the z direction is large compared to the distance a disturbance propagates during the laser pulse, in other words $l_c > v_g t_e$. In this case we write $z = v_g t$ in the saddle-point exponent, and for maximum gain let $t = t_e$, so that

$$\ln G = 2(\nu'' - v_g q'')t_e . \qquad (5.17)$$

This we call the short-time or extreme-transient region. Further maximization is obtained by requiring $(\nu'' - q'' v_g)t_e$ to be a maximum. Differentiating this factor with respect to v_g, we obtain

$$\frac{d}{dv_g}(\nu'' - q'' v_g)t_e = \frac{(\nu''\cos\theta - q'' v_0)t_e}{v_0 - v_g\cos\theta} , \qquad (5.18)$$

using (5.5). Thus, we find

$$v_0 q'' = \nu''\cos\theta \qquad (5.19)$$

for an extremum, and (5.17) can be written

$$\ln G = 2\nu'' t_p . \qquad (5.17')$$

Differentiating (5.18) once more, we have

$$\frac{d^2}{dv_g^2}(\nu'' - q'' v_g)t_e\bigg|_{v_0 q'' = \nu''\cos\theta} = -\frac{1}{v_g}\frac{d\nu''}{dv_g}t_e , \qquad (5.20)$$

again using (5.5). Thus we have a maximum when

$$d\nu''/dv_g|_{v_0 q'' = \nu''\cos\theta} > 0 .$$

In the short-time region, since $t_e > l_c/v_{g,T}$, where $v_{g,T} \equiv v_g(v_0 q'' = \nu''\cos\theta)$, we may define a maximum time

$$t_T = \frac{v_0 - v_{g,T}\cos\theta}{v_0 v_{g,T}}\, l_c , \qquad (5.21)$$

below which the short-time results are correct.

We now consider the intermediate region, in which $t_T < t_p < t_L$. Note that $t_T < t_L$ implies that

$$v_{g,T} > v_{g,L} .$$

Furthermore, we expect the group velocity to vary smoothly between these two limits as t_p varies in the intermediate-time region:

$$v_{g,T} > v_g > v_{g,L} .$$

Now consider $(\nu'' v_g^{-1} - q'')l_c$ subject to the constraint

$$v_g \geq l_c/t_e > v_{g,L} . \qquad (5.22)$$

Equation (5.13) and the positive sign of ν'' imply $(\nu'' v_g^{-1} - q'')l_c$ is maximum at the limits of the constraint, namely, when

$$v_g = \frac{l_c}{t_p + (l_c\cos\theta)/v_0} , \qquad (5.23)$$

and that its value decreases monotonically as t_p decreases. Similar considerations show that the expression $(\nu'' - q'' v_g)t_e$ subject to the constraint

$$v_g \leq l_c/t_e < v_{g,T} \qquad (5.24)$$

is also maximized when (5.23) holds. The result for the gain in the intermediate region is

$$\ln G = 2[\nu''(v_g = l_c/t_e)t_e - q''(v_g = l_c/t_e)l_c] . \qquad (5.25)$$

When a system has a nonconvective instability (sometimes called an absolute instability), a critical power level exists at which $v_{g,L}$ vanishes; then, according to (5.16), t_L is infinite. There then exists no steady state either at or above this critical power level. The formulas for the intermediate region then hold for all values of $t > t_T$. An example will be provided by electrostrictive Brillouin scattering.

VI. GROWTH IN REGION OF NEGLIGIBLE STOKES AND ANTI-STOKES COUPLING

Away from the region where the Stokes and the anti-Stokes are simultaneously phase matched we may neglect the coupling between these two waves via the material excitation.[20] The extent of the region over which they are coupled depends on the size of the nonlinearity. However, in reasonable situations, its angular extent is at most one or two degrees.

A. Electrostrictive Brillouin and Entropy Fluctuation Effects

We obtain from (3.13) and (4.5), assuming α and

$(\partial\epsilon/\partial T)_\rho$ can be neglected and that $\mu = 1$ and the dispersion relation

$$q - \frac{\nu}{v_0} + k_0 \frac{AQ^2(\kappa Q^2 - i\rho_0 C_v \Omega)}{D(Q,\ \Omega)} = 0\ , \qquad (6.1)$$

where

$$A = \left(\frac{\partial\epsilon}{\partial\rho}\right)_T^2 \frac{\rho_0 |\mathcal{E}_0|^2}{32\pi} \left(\frac{\epsilon_0 + 2}{3}\right)^2 \qquad (6.2)$$

and $D(Q,\ \Omega)$ is given by (4.6). In order to simplify further discussion we introduce

$$Q_0 = |\vec{k}_1^0 - \vec{k}_0| = 2k_0 \sin\tfrac{1}{2}\theta \qquad (6.3)$$

and assume that $Q_0 \gg |q|$.

1. *Brillouin Region*

In this region we have

$$|\Omega| \approx v_s Q_0 \gg \kappa Q_0^2/\rho_0 C_v\ ,$$

so that the dispersion relation (6.1) becomes approximately

$$\left(q - \frac{\nu}{v_0}\right) - \frac{C_b k_0 \Gamma}{v_s' q + \nu + i\Gamma} = 0\ , \qquad (6.4)$$

where we have taken $\Omega = -v_s Q_0 + \nu$,

$$v_s' = v_s Q_0 / 2k_0\ , \qquad (6.5)$$

and introduced the dimensionless coupling constant

$$C_b = \left(\frac{\partial\epsilon}{\partial\rho}\right)_T^2 \frac{\rho_0 Q_0 |\mathcal{E}_0|^2}{64\pi v_s \Gamma}\ , \qquad (6.6)$$

where Γ is the damping constant

$$\Gamma = \frac{Q_0^2}{2\rho_0}\left(\eta + \frac{\kappa(\gamma - 1)}{C_p}\right)\ . \qquad (6.7)$$

Taking the derivative with respect to ν' of the dispersion relation and using the saddle-point condition (5.9) as well as the definition of v_g given by (5.4), we obtain

$$\frac{1}{v_g} - \frac{1}{v_0} + \frac{C_b k_0 \Gamma (v_s'/v_g + 1)}{(v_s' q + \nu + i\Gamma)^2} = 0\ . \qquad (6.8)$$

Solving (6.4) and (6.8) for ν and q, we obtain

$$\nu = \frac{-iv_0}{v_0 + v_s'}\left[\Gamma \pm B\left(\frac{1}{v_s'} + \frac{1}{v_g}\right)^{-1}\left(\frac{1}{v_s'} + \frac{1}{v_g} - \frac{1}{v_0}\right)\right], \qquad (6.9)$$

$$q = \frac{-i}{v_0 + v_s'}\left[\Gamma \pm B\left(\frac{1}{v_s'} + \frac{1}{v_g}\right)^{-1}\left(\frac{2}{v_s'} + \frac{1}{v_g} - \frac{v_0}{v_s' v_g}\right)\right] \qquad (6.10)$$

where

$$B = [C_b k_0 \Gamma v_0 (v_s' + v_g)/(v_0 - v_g)]^{1/2}\ . \qquad (6.11)$$

a. Short times $t_p \leq t_T$. Using (5.19), we can solve for the group velocity at the point of maximum gain for the growing root

$$v_g = \tfrac{1}{2}\{[(v_0 - v_s' + 2v_\alpha v_\beta/v_{cb}') + (v_0 - v_s' + 2v_\alpha v_\beta/v_{cb}')^2 + 4v_0(v_s' - v_\beta^2/v_{cb}')(1 + v_\alpha^2/v_0 v_{cb}')]^{1/2}\} \times (1 + v_\alpha^2/v_0 v_{cb}')^{-1}, \qquad (6.12)$$

where

$$v_{cb}' = v_{cb}(1 - \cos\theta)^2 = 4v_{cb}\sin^4\tfrac{1}{2}\theta \qquad (6.13)$$

$$v_{cb} = \Gamma/C_b k_0\ , \qquad (6.14)$$

$$v_\alpha = 2v_0 - (v_0 - 2v_s')\cos\theta\ , \qquad (6.15)$$

$$v_\beta = v_0 + v_s'(2\cos\theta - 1)\ . \qquad (6.16)$$

Noting that $v_0 \gg v_s$, (6.12) reduces to

$$v_g = \frac{v_0}{2[1 + (2 - \cos\theta)^2 v_0/v_{cb}']}\{1 + 2(2 - \cos\theta)v_0/v_{cb}' + [1 + 4(1 - \cos\theta)v_0/v_{cb}']^{1/2}\}, \qquad (6.17)$$

which is independent of v_s. There are two interesting limits:

(i) weak coupling, when $v_0 \ll v_{cb}'$,

$$v_g \approx v_0(1 - v_0/v_{cb})\ ; \qquad (6.18)$$

(ii) strong coupling, when $v_0 \gg v_{cb}$,

$$v_g \approx \frac{v_0}{(2 - \cos\theta)}\left(1 + \frac{[v_{cb}(1 - \cos\theta)^3/v_0]^{1/2}}{2 - \cos\theta}\right)\ . \qquad (6.19)$$

Using (5.17), (6.7), and (6.8), and using $v_0 \gg v_s$, we have for the gain exponent

$$\ln G = 2\Gamma\left(1 - \frac{v_g}{v_0}\right)\left[2\left(\frac{v_g v_0}{v_{cb}(v_0 - v_g)}\right)^{1/2} - 1\right]t_e\ . \qquad (6.20)$$

We have, in the weak-coupling limit,

$$\ln G = 2C_b k_0 v_0 t_e = \frac{2C_b k_0 v_0 t_p}{1 - \cos\theta + v_0\cos\theta/v_{cb}} \qquad (6.21)$$

and, in the strong-coupling limit,

$$\ln G = \frac{4}{2 - \cos\theta}[(1 - \cos\theta)C_b \Gamma v_0 k_0]^{1/2} t_e = 2\left(\frac{v_0 k_0 \Gamma C_b}{1 - \cos\theta}\right)^{1/2} t_p\ . \qquad (6.22)$$

For the time t_T we have, in the weak-coupling limit,

$$t_T = \left(\frac{1 - \cos\theta}{v_0} + \frac{1}{v_{cb}}\right) l_c\ , \qquad (6.23)$$

while, in the strong-coupling limit,

$$t_T = 2(1 - \cos\theta) l_c / v_0\ . \qquad (6.24)$$

It should be noted that the approximation $|\Omega| \approx v_s Q_0$ implies $\nu \ll v_s Q_0$. On the other hand, significant gain occurs only when $\nu t_e > 1$. Hence the useful range of the above formulas is confined to $t_e \gg 1/v_s Q_0$. For very short pulses, which violate this restriction, it is necessary to return to (6.1) and make approximations appropriate to this region (see Sec. VIC).

b. Intermediate-time region $t_L \ge t_p \ge t_T$. In this region

$$\ln G = 2\Gamma t_e\left[\frac{2B'}{\Gamma}\left(\frac{1}{v_s'}+\frac{t_e}{l_c}\right)^{-1}\left(\frac{1}{v_s'}+\frac{t_e}{l_c}-\frac{l_c}{v_s' v_0 t_e}\right) - \left(1-\frac{l_c}{v_0 t_e}\right)\right], \quad (6.25)$$

where

$$B' = \left[\frac{k_0 \Gamma C_b v_0 (v_s' + l_c/t_e)}{v_0 - l_c/t_e}\right]^{1/2}, \quad (6.26)$$

$$t_e = t_p + (l_c \cos\theta)/v_0 . \quad (6.27)$$

If we have $v_0 \gg l_c/t_e \gg v_s'$, then

$$\ln G = 4(C_b k_0 l_c \Gamma t_p)^{1/2} - 2\Gamma t_p , \quad (6.28)$$

which is a strong-coupling result.

c. Long-time region $t_p > t_L$. In this region the gain is given by

$$\ln G = \frac{2\Gamma l_c}{v_0}\left[1 + \frac{B}{\Gamma}\left(\frac{1}{v_s'}+\frac{1}{v_g}\right)^{-1}\left(\frac{v_0}{v_s' v_g} - \frac{2}{v_s'} - \frac{1}{v_g}\right)\right], \quad (6.29)$$

where

$$v_g = \tfrac{1}{2}\{(v_{cb} - 4v_s') + 2[(v_{cb} - 4v_s')v_s'(1 + v_{cb}/v_0)]^{1/2}\} \times (1 + v_{cb}/v_0)^{-1} . \quad (6.30)$$

An alternative form for the gain is

$$\ln G = \frac{4k_0 l_c C_b}{(1 - 4v_s' C_b k_0/\Gamma)^{1/2} + 1} , \quad (6.31)$$

so that when $v_{cb} \gg 4v_s'$ we have

$$\ln G = 2C_b k_0 l_c , \quad (6.32)$$

$$v_g = v_0 v_{cb}/(v_0 + v_{cb}) , \quad (6.33)$$

$$t_L = \left(\frac{1-\cos\theta}{v_0} + \frac{1}{v_{cb}}\right) l_c , \quad (6.34)$$

which is identical with t_T for the weak-coupling case given by (6.23).

As v_{cb} approaches v_s', v_g approaches zero, and consequently t_L becomes infinite. As noted before, this implies the existence of a nonconvective instability. There is no steady state, and for $v_{cb} \le 4v_s'$ the formulas for the intermediate-time region apply for all $t > t_T$. Nonconvective instabilities in stimulated Brillouin scattering were discussed in Ref. 7. The possibility of their occurrence was, in Ref. 7, restricted to larger than right angle scattering, but in the present treatment there is no such restriction. The difference lies in the fact that Im$\vec{q}$ was taken parallel to $\vec{k}_0$ in Ref. 7, but parallel to $\vec{k}_1^0$ in the present treatment.

2. *Entropy or Rayleigh-Peak Region*

In this region we have $|\Omega| \approx \kappa Q_0^2/\rho_0 C_v \ll v_s Q_0$, so that the dispersion relation becomes, with $\nu = \Omega$,

$$q - \frac{\nu}{v_0} - \frac{2C_e k_0}{(\gamma-1)} - \frac{2C_e k_0}{1 - i\nu\tau} = 0 , \quad (6.35)$$

where

$$C_e = (\gamma - 1)A/2v_s^2 , \quad (6.36)$$

$$\tau = \Gamma^{-1} = \rho_0 C_p/\kappa Q_0^2 , \quad (6.37)$$

noting that A is given by (6.2).

Again taking the derivative of (6.35) with respect to ν' using (5.4) and (5.3), we have

$$\frac{1}{v_g} - \frac{1}{v_0} - \frac{i2C_e k_0 \tau}{(1 - i\nu\tau)^2} = 0 . \quad (6.38)$$

Solving (6.38) for ν gives

$$\nu = -i/\tau \pm (1 - i)B , \quad (6.39)$$

where

$$B = \left(\frac{C_e k_0}{\tau(1/v_g - 1/v_0)}\right)^{1/2} . \quad (6.40)$$

Inserting (6.39) into (6.35), we have

$$q = \frac{2C_e k_0}{(\gamma-1)} - \frac{i}{v_0\tau} \pm (1-i)B\left(\frac{2}{v_0} - \frac{1}{v_g}\right) . \quad (6.41)$$

a. Short-time regime $t_p < t_T$. The group velocity in this regime is given using (5.19) and (6.17), replacing v_{be}' by v_{ce}', where

$$v_{ce}' = v_{ce}(1 - \cos\theta)^2 = 4v_{ce}\sin^4\tfrac{1}{2}\theta , \quad (6.42)$$

$$v_{ce}^{-1} = C_e \tau k_0 . \quad (6.43)$$

We have as before strong- and weak-coupling limits:

(i) weak coupling:

$$v_{ce} \gg v_0, \quad v_g \approx v_0 ,$$

$$\ln G = \frac{2C_e k_0 v_0 t_p}{1 - \cos\theta + v_0 \cos\theta/v_{ce}} , \quad (6.44)$$

with a frequency shift

$$\nu' = -1/\tau , \quad (6.45)$$

where t_T is given by (6.23) with v_{cb} replaced by v_{ce};

(ii) strong coupling:

$$v_{ce} \ll v_0, \quad v_g \approx v_0/(2 - \cos\theta) ,$$

$$\ln G = 2\left(\frac{C_e k_0 v_0 \tau}{1 - \cos\theta}\right)^{1/2} \frac{t_p}{\tau} , \quad (6.46)$$

with a frequency shift

$$\nu' = \frac{-1}{\tau}\left(\frac{2C_e k_0 v_0 \tau}{1 - \cos\theta}\right)^{1/2} = \frac{-\ln G}{2t_p} , \quad (6.47)$$

where t_T is given by (6.24).

b. Intermediate-time regime $t_L \ge t_p \ge t_T$. Here the gain is given by

$$\ln G = 2t_e(2B' - \tau^{-1})\left(1 - \frac{l_c}{v_0 t_e}\right) , \quad (6.48)$$

where B' is now

$$B' = \left(\frac{C_e v_0 k_0 l_c}{(v_0 t_e - l_c)\tau} \right)^{1/2} . \tag{6.49}$$

If we have $v_0 \gg l_c / t_e$, then

$$\ln G = 4(C_e k_0 l_c t_p / \tau)^{1/2} - 2 t_p / \tau . \tag{6.50}$$

c. Long-time regime $t_p \geq t_L$. Here we have

$$\ln G = 2 C_e k_0 l_c , \tag{6.51}$$

$$v_g = v_0 v_{ce} / (v_0 + v_{ce}) ; \tag{6.52}$$

also t_L is given by (6.34) with v_{cb} replaced by v_{ce}, and the frequency shift is

$$\nu' = -1/\tau . \tag{6.53}$$

The results in this subsection are valid provided the nonlinearity does not cause complex frequency and propagation constant changes larger than the separation between the Brillouin and entropy modes. We will discuss the breakdown of this condition in Sec. VI C.

B. General Results

We may treat absorptive Brillouin, absorptive entropy, Raman, and Rayleigh-wing scattering in a manner similar to the electrostrictive Brillouin and entropy scattering treatment in Sec. VI A. It is sufficient to note that for small nonlinearities the dispersion relation for all these processes can be written in the approximate form

$$q - \frac{\nu}{v_1} + \frac{M}{v_{ex} q + \nu + i\Gamma} = H ,$$

with all parameters independent of q and ν. This relation leads to solutions of similar character for all these cases. We therefore simply write the results in general form for the three time regions with the constants for the various processes given in Table I.

To simplify the results we introduce the quantities

$$v_c = \Gamma / C k_1 , \tag{6.54}$$

$$v'_{ex} = v_{ex} Q_0 / 2 k_1 , \tag{6.55}$$

$$\mathrm{Re}\, \Omega = \Omega_0 + p \nu' . \tag{6.56}$$

Note that for all cases except Raman scattering it is a good approximation to replace k_1 and v_1 by k_0 and v_0, respectively.

1. *Short-Time Regime* $t_p \leq t_T$

For short times we have the following results in the weak-coupling limit $v_c \gg v_0$:

$$\ln G = \frac{2 C k_1 v_1 t_p}{1 - v_1 \cos\theta \, (1 - v_1 / v_c) / v_0} , \tag{6.57}$$

$$t_T = \left(\frac{1}{v_1} + \frac{1}{v_c} - \frac{\cos\theta}{v_0} \right) l_c , \tag{6.58}$$

$$v_g = v_1 (1 - v_1 / v_c) , \tag{6.59}$$

$$\nu' = C v_0 k_0 = \Gamma v_0 / v_c . \tag{6.60}$$

In the strong-coupling limit $v_0 \gg v_c$ we have

$$\ln G = 2 \left(\frac{v_1 C k_1 \Gamma}{1 - (v_1 \cos\theta)/v_0} \right)^{1/2} t_p , \tag{6.61}$$

$$t_T = 2 \left(1 - \frac{v_1 \cos\theta}{v_0} \right) \frac{l_c}{v_1} , \tag{6.62}$$

TABLE I. Constants for various physical processes to be used in conjunction with the general equations in Sec. VI B.

Process \ Constant	$C / \lvert \mathcal{E}_0 \rvert^2 \left(\frac{\epsilon_0 + 2}{3} \right)^3$	Γ	v_{ex}	Ω_0	p
Electrostrictive Brillouin	$\frac{Q_0 \rho_0}{64 \pi v_s \epsilon_0 \Gamma} \left(\frac{\partial \epsilon}{\partial \rho} \right)_T^2$	$\frac{Q_0^2}{2\rho_0} \left(\eta + \frac{\kappa(\gamma - 1)}{C_p} \right)$	v_s	$-v_s Q_0$	0
Electrostrictive entropy	$\frac{\gamma(\gamma - 1)\rho_0}{64 \pi v_s^2 \epsilon_0} \left(\frac{\partial \epsilon}{\partial \rho} \right)_T^2$	$\frac{\kappa Q_0^2}{\rho_0 C_p}$	0	0	-1
Absorptive Brillouin	$\frac{\alpha c \beta}{32 \pi C_p n_0 \Gamma} \left(\frac{\partial \epsilon}{\partial \rho} \right)_T$	$\frac{Q_0^2}{2\rho_0} \left(\eta + \frac{\kappa(\gamma - 1)}{C_p} \right)$	v_s	$+v_s Q_0$	± 1
Absorptive entropy	$\frac{\alpha c \beta}{32 \pi C_p n_0 \Gamma} \left(\frac{\partial \epsilon}{\partial \rho} \right)_T$	$\frac{\kappa Q_0^2}{\rho_0 C_p}$	0	0	1
Raman	$\frac{\pi N}{4 m \omega_v \epsilon_1 \Gamma} \left(\frac{\partial \alpha}{\partial x_0} \right)^2 \left(\frac{\epsilon_1 + 2}{3} \right)^2$	$\Delta \omega_v$	0	$-\omega_v$	0
Rayleigh wing	$\frac{2 \pi N (\Delta \alpha)^2}{45 k_B T \epsilon_0} \left(\frac{\epsilon_0 + 2}{3} \right)^2$	$\frac{1}{\tau} = \frac{3}{\tau_{\mathrm{Debye}}}$	0	0	-1

$$v_g = \frac{v_1}{2-(v_1\cos\theta)/v_0}\ , \qquad (6.63)$$

$$\nu' = \left(\frac{Cv_1\Gamma k_1}{1-(v_1\cos\theta)/v_0}\right)^{1/2}\ . \qquad (6.64)$$

2. *Intermediate-Time Regime* $t_T \le t_p \le t_L$

For intermediate times we have the following results provided $l_c/t_e \gg v'_{ex}$:

$$\ln G = 2\Gamma t_e\left(\frac{2B'}{\Gamma}-1\right)\left(1-\frac{l_c}{v_1 t_e}\right)\ , \qquad (6.65)$$

where

$$B' = \left(\frac{C\Gamma v_1 k_1 l_c}{v_1 t_e - l_c}\right)^{1/2}\ , \qquad (6.66)$$

$$t_e = t_p + (l_c\cos\theta)/v_0\ , \qquad (6.67)$$

$$v_g = l_c/t_e\ , \qquad (6.68)$$

$$\nu' = B'\ . \qquad (6.69)$$

When $v_0 \gg l_c/t_e$ and $t_L \gg t_T$, these results reduce to

$$\ln G = 4(Ck_1 l_c\Gamma t_p)^{1/2} - 2\Gamma t_p\ , \qquad (6.70)$$

$$\nu' = (Ck_1 l_c\Gamma/t_p)^{1/2}\ . \qquad (6.71)$$

3. *Long-Time Regime* $t_p \ge t_L$.

When $v_c \ge 4v'_{ex}$ we have

$$\ln G = \frac{4Ck_1 l_c}{(1-4v'_{ex}Ck_1/\Gamma)^{1/2}+1}\ , \qquad (6.72)$$

$$v_g = \frac{\frac{1}{2}\{(v_c - 4v'_{ex}) + [v_c(v_c-4v'_{ex})]^{1/2}\}}{1+v_c/v_1}, \qquad (6.73)$$

$$\nu' = 1/\Gamma; \qquad (6.74)$$

when $v_c \gg 4v'_{ex}$ we have

$$G = 2Ck_1 l_c, \qquad (6.75)$$

$$v_g = v_1 v_c/(v_1+v_c), \qquad (6.76)$$

$$\nu' = 1/\Gamma, \qquad (6.77)$$

$$t_L = \left(\frac{1}{v_1}+\frac{1}{v_c}-\frac{\cos\theta}{v_0}\right) l_c. \qquad (6.78)$$

The extension of the tabulated formulas to include electrostrictive and absorptive effects simultaneously is straightforward, but has not been carried out here.

C. Extreme Nonlinear Growth

As pointed out earlier, we assumed in Secs. VI A and VI B that the material motions were not strongly perturbed by the nonlinearity from their natural frequencies. The breakdown of this condition is most important in the case of the hydrodynamic modes (Brillouin and entropy waves) and is of lesser significance in Raman scattering. For spatial growth in the case of the hydrodynamic modes the condition for the natural material propagation constants to be weakly perturbed by the nonlinearity is

$$Q_0 \gg k_0 C_b,\ \ k_0 C_e\ , \qquad (6.79)$$

where, as before, b denotes Brillouin waves and e denotes entropy waves. For electrostriction we note that $C_b k_0 \gg C_e k_0$, and we may rewrite the condition (6.79) for typical liquids as

$$I_L \ll 2\times 10^6 \sin^2\tfrac{1}{2}\theta\ \text{MW/cm}^2, \qquad (6.80)$$

where I_L is the laser intensity. For absorption (6.79) becomes

$$\alpha I_L \ll 2\times 10^5 \sin^3\tfrac{1}{2}\theta\ \text{MW/cm}^3. \qquad (6.81)$$

To violate these conditions we must use powers somewhat in excess of those which are currently available for times as long as the steady-state times in the hydrodynamic scattering processes.

For temporal growth we have a similar condition for the validity of the results in Secs. VI A and VI B:

$$v_s^2 Q_0^2 \gg C_b k_0\Gamma_b v_0,\ \ C_e k_0\Gamma_e v_0, \qquad (6.82)$$

which, in the electrostrictive case, can be rewritten as

$$I_L \ll 10^2 \sin\tfrac{1}{2}\theta\ \text{MW/cm}^2\ , \qquad (6.83)$$

while for the absorptive case we have

$$\alpha I_L \ll 2\times 10^2 \sin^2\tfrac{1}{2}\theta\ \text{MW/cm}^2. \qquad (6.84)$$

These last conditions can be easily violated with Q-switched lasers. We will therefore examine the temporal growth when the conditions given by (6.83) and (6.84) are strongly violated. Under these conditions the Brillouin and entropy modes are no longer distinct.

1. *Electrostrictive Case*

For electrostrictive scattering we readily obtain from (6.1), neglecting damping and the propagation vector in the material propagator,

$$q - \frac{\nu}{v_0} + \frac{F}{\nu^2} = 0, \qquad (6.85)$$

where

$$F = \left(\frac{\partial\epsilon}{\partial\rho}\right)_T^2 \left(\frac{\epsilon_0+2}{3}\right)^2 \frac{\rho_0\,|\mathcal{E}_0|^2 Q_0^2 k_0}{8\pi\epsilon_0}\ . \qquad (6.86)$$

Taking the derivative with respect to ν' and using (4.3) and (4.4), we obtain

$$\left(\frac{1}{v_g}-\frac{1}{v_0}\right) = \frac{2F}{\nu^3}\ , \qquad (6.87)$$

which has three roots:

$$\nu = [\cos(\tfrac{2}{3}\pi n) + i\sin(\tfrac{2}{3}\pi n)]H, \qquad (6.88)$$

where n runs 1–3 and

$$H=\left(\frac{2F}{(1/v_g-1/v_0)}\right)^{1/3} . \quad (6.89)$$

From (6.85) we have for q

$$q=\frac{\nu}{2}\left(\frac{3}{v_0}-\frac{1}{v_g}\right) . \quad (6.90)$$

a. Short-time regime $t_p<t_T$. Using (5.19) we solve for the group velocity and find

$$v_g=v_0/(3-2\cos\theta) . \quad (6.91)$$

We have for the gain

$$\ln G=2(\sin\tfrac{2}{3}\pi n)\left[-\frac{Fv_0}{1-\cos\theta}\right]^{1/3} t_p, \quad (6.92)$$

and for t_T

$$t_T=3l_c(1-\cos\theta)/v_0 . \quad (6.93)$$

The maximum in gain occurs when $n=1$ and is

$$\ln G=\sqrt{3}\left(\frac{Fv_0}{1-\cos\theta}\right)^{1/3} t_p , \quad (6.94)$$

and corresponds to a frequency shift

$$\nu'=-\frac{1}{2}\left(\frac{Fv_0}{1-\cos\theta}\right)^{1/3} . \quad (6.95)$$

b. Intermediate-time regime $t_p>t_T$. From (6.88) and (6.90) and setting $v_g=l_c/t_e$, we have

$$\ln G=3\sin\left(\frac{2\pi n}{3}\right)\left(\frac{2F}{t_e/l_c-1/v_0}\right)^{1/3}\left(t_e-\frac{l_c}{v_0}\right), \quad (6.96)$$

where

$$t_e=t_p+(l_c\cos\theta)/v_0 . \quad (6.97)$$

If $t_p\gg l_c/v_0$, the gain reduces to

$$\ln G=3(\sin\tfrac{2}{3}\pi n)(2Fl_ct_p^2)^{1/3} , \quad (6.98)$$

which has a maximum value of

$$\ln G=\tfrac{3}{2}\sqrt{3}(2Fl_ct_p^2)^{1/3} , \quad (6.99)$$

when $n=1$.

2. *Absorptive Scattering*

In the case of absorptive scattering we obtain from (3.13) and (4.5)

$$q=\frac{\nu}{v_0}+\frac{iJ}{\nu^3}=0 , \quad (6.100)$$

where

$$J=k_0|\mathcal{E}_0|^2\left(\frac{\partial\epsilon}{\partial\rho}\right)_T\frac{\beta v_s^2\alpha n_0cQ_0^2}{4\pi C_p\epsilon_0}\left(\frac{\epsilon_0+2}{3}\right)^2 . \quad (6.101)$$

Differentiating with respect to ν' we have

$$\left(\frac{1}{v_g}-\frac{1}{v_0}\right)=\frac{3iJ}{\nu^4} , \quad (6.102)$$

which gives the following roots for ν :

$$\nu=[\cos(\tfrac{1}{2}n\pi+\tfrac{1}{8}\pi)+i\sin(\tfrac{1}{2}n\pi+\tfrac{1}{8}\pi)]L , \quad (6.103)$$

where n runs 1–4 and

$$L=\left(\frac{3J}{1/v_g-1/v_0}\right)^{1/4} . \quad (6.104)$$

From (6.100) we have for q

$$q=\frac{\nu}{3}\left(\frac{4}{v_0}-\frac{1}{v_g}\right) . \quad (6.105)$$

a. Short-time regime $t_T>t_p$. From (5.19) and (6.105) we obtain

$$v_g=v_0/(4-3\cos\theta) , \quad (6.106)$$

in which case L becomes

$$L=\left(\frac{Jv_0}{1-\cos\theta}\right)^{1/4} , \quad (6.107)$$

$$\ln G=2[\cos(\tfrac{1}{2}n\pi+\tfrac{1}{8}\pi)+i\sin(\tfrac{1}{2}n\pi+\tfrac{1}{8}\pi)]L \quad (6.108)$$

and the time t_T becomes

$$t_T=4l_c(1-\cos\theta)/v_0 . \quad (6.109)$$

The maximum gain occurs when $n=1$ and is given by

$$\ln G=(2+\sqrt{2})^{1/2}\left(\frac{Jv_0}{1-\cos\theta}\right)^{1/4} . \quad (6.110)$$

Thus the gain goes as the square root of the electric field.

b. Intermediate-time regime $t_p\geq t_T$. In this time region, using (6.103) and (6.105), and replacing v_g by l_c/t_e, we have for the gain

$$\ln G=\tfrac{8}{3}\sin(\tfrac{1}{2}n\pi+\tfrac{1}{8}\pi)t_e\left(\frac{3J}{t_e/l_c-1/v_0}\right)^{1/4}\left(1-\frac{l_c}{v_0t_e}\right) , \quad (6.111)$$

where as usual

$$t_e=t_p+(\cos\theta)l_c/v_0 . \quad (6.112)$$

When $l_c/t_e\ll v_0$,

$$\ln G=\tfrac{8}{3}\sin(\tfrac{1}{2}n\pi+\tfrac{1}{8}\pi)(3Jt_p^3l_c)^{1/4} , \quad (6.113)$$

which gives a maximum gain of

$$\ln G=\tfrac{4}{3}(2+\sqrt{2})^{1/2}(3Jt_p^3l_c)^{1/4}, \quad (6.114)$$

where $n=1$.

VII. CONSIDERATION OF EXPERIMENTS AND CONCLUSIONS

Some of the features of the analysis presented here are well known and understood experimentally, other aspects are not. A number of authors have observed apparent steady-state or long-time regime gain, for example, Walder and Tang,[21] Hagenlocker *et al.*,[22] Maier,[23] and Denariez and Bret.[24] Some authors have found situations in which the steady-state theory does not apply. We discuss some of these results below.

Brewer[25] has measured gain as a function of cell length and observed transient or time-dependent gain effects. However, there is little agreement between the experimental results and the theoretical analysis[7] which is applied. This analysis is limited to the strongly coupled long- and intermediate-time regimes and does not include the short-time regime or the possibility of weak coupling. Since no input scattered power is sent into the cell, these results also include the effects of growth from spontaneous noise. Saturation of gain due to depletion of the laser by the backward-traveling scattered light may also play a role in these results.

Pine,[9] using an off-axis resonator, was able to observe growth which had temporal character and found results which were apparently consistent with the strong-coupling regime.

Hagenlocker *et al.*[22] made careful, quantitative studies of gain in an amplifier in both the long-time (or steady-state) and in the time-dependent regimes. In their experiment they hold the gain fixed throughout by varying the laser intensity, compute the steady-state gain from this intensity, and plot the ratio as a function of Γ^{-1}. These results are purported to confirm the theory[7]; however, the method of data analysis tends to throw some doubt on this. Following Kroll, these authors write the gain in the strongly coupled intermediate-time regime as [see (6.70)]

$$\ln G = 4(Ck_0 l_c \Gamma t_p)^{1/2} - 2\Gamma t_p . \tag{7.1}$$

They then reexpress this as

$$\frac{\ln G_{ss}}{\ln G} = \frac{(\ln G + 2\Gamma t_p)^2}{16 \ln G\, \Gamma t_p} , \tag{7.2}$$

where G_{ss} is to steady-state gain given by (6.75), namely,

$$\ln G_{ss} = 2Ck_0 l_c . \tag{7.3}$$

Equation (7.2) gives the appropriate linear dependence on Γ^{-1} when $\frac{1}{2}\ln G\, \Gamma^{-1} \gg t_p$ as shown in their Fig. 5. It does not, however, give the steady-state result [the left-hand side of (7.2) becomes unity in the steady state], contrary to the assertion of these authors. Furthermore, the linear dependence on Γ^{-1} when $\frac{1}{2}\ln G\, \Gamma^{-1} \gg t_p$ is also a consequence of the short-time strongly coupled regime (6.61). In addition, if following Herman and Gray[11] and Denariez and Bret [24] we write the gain as

$$\ln G = \frac{\Gamma \ln G_{ss}}{\Gamma + t_p^{-1}} , \tag{7.4}$$

assuming an uncertainty limited laser pulse, or equivalently

$$\frac{\ln G_{ss}}{\ln G} = 1 + (\Gamma t_p)^{-1} , \tag{7.5}$$

we then note that the solid curve in Fig. 5 of Hagenlocker and Minck is described reasonably well by (7.5) given the proper choice of t_p. There is some question about the correct value t_p in this experiment.

Denariez and Bret[24] have also obtained results concerning the dependence of gain on the damping Γ, which again shows (see their Fig. 4) a deviation from steady state for small Γ. The present theory in the strongly coupled limit would predict the observed onset of deviation from the steady state if $\ln G$ is about 25 and the pulse is of the order of a few times 10^{-8} sec long. However, the expression for the gain used by these authors [which is effectively (7.5)] would also give a similar result assuming a t_p of the order of 10^{-9} sec. A further extension of this approach has been made by Bret and Weber,[26] in evaluating experimental data on stimulated scattering with picosecond pulses.

Pohl *et al.*[27] observed in stimulated Brillouin scattering a dependence of gain on the ratio of the rise time of the signal to be amplified to the damping time Γ^{-1}. The gains observed are small and cannot be interpreted in terms of the present theory.

A number of authors[28] have observed stimulated entropy (Rayleigh-peak) scattering with short laser pulses. Transient effects could play a role in these observations because of the long relaxation time of the entropy waves.

Shapiro *et al.*[29] and others [26,30] observed stimulated Raman scattering in the picosecond regime where transient gain reduces stimulated Brillouin scattering. Maier and co-workers[30] analyzed their experimental results on backward Raman scattering in terms of a model involving the temporal and spatial interaction of the laser pulse with a backward-traveling stimulated Raman pulse. Their analysis was limited to the rate-equation approximation. The effects of pulse shape on transient gain and the generation, in the transient gain regime, of Raman-Stokes pulses shorter than the laser pulse have been studied in liquids by Carman *et al.*[31] The results can be compared to transient gain theory, having many of the features presented here, but modified to include a variety of input pulse shapes in addition to the square pulse.[17] Observations[32] have also been made in gases with picosecond pulses of transient rotational and vibrational scattering. The influence of gain on the linewidth of stimulated Raman scattered light has been observed recently by Akhmanov *et al.*[33] Transient effects have been incorporated into their theoretical analyses of these results.

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