Quantum Theory of Light Propagation in Amplifying Media*

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A simple model of a traveling-wave parametric amplifier is discussed in quantum-mechanical terms. The amplifier consists of a slab of Raman-active material, illuminated on one face by an intense laser beam that serves as the pump mode; an optical-phonon oscillation serves as the idler mode, and the scattered Stokes light occupies the signal mode. With only a few simplifying assumptions, it is possible to solve the equation of motion for the signal field operator exactly. For large times, both an amplified steady signal and the quantum noise contributed by the material assume simple steady states in an amplifier of finite length. The transient and asymptotic behaviors of the signal and noise intensities are found, together with the corresponding first-order correlation functions. The quasiprobability distribution which specifies the complete density operator for the electric field strength of the amplified signal is also derived.

I. INTRODUCTION

The development of lasers capable of producing intense, steady, and highly coherent beams of light has made possible the development of devices that amplify light beams. Among these is the optical parametric amplifier, whose microwave analog has been in use for some time.¹ Here we shall consider a simple, but fairly realistic, model of an optical parametric amplifier. Our discussion will be aimed at providing a simple and quantum-mechanically complete description of the output of the amplifier, including both the amplified signal and the intrinsic noise.

In a parametric device, amplification is achieved through the driven oscillations of one of the parameters describing the system. If a system has several modes of oscillation coupled through a nonlinear interaction, then, under the appropriate conditions, energy supplied to one of these modes may amplify oscillations present in the other modes. If the oscillations of the driven mode are strongly enough excited by an external force, the reaction on it due to coupling with the other modes may often be neglected. The amplitude of the driven oscillations may then be regarded as an externally specified parameter of the system rather than a dynamical variable. This approximation greatly simplifies the analysis of the parametric amplifier.

Usually three modes of oscillation are involved in a parametric amplifier. The driven mode is called the pump mode; the mode whose excitation carries the signal to be amplified is called the signal mode, and the remaining mode is called the idler mode.

At optical frequencies, there are many nonlinear interactions that can couple electromagnetic fields at two different frequencies with a material oscillation or with a third mode of the radiation field.²

Couplings of these types may be regarded as generating scattering processes in which an incident photon is inelastically scattered by the material or by another field mode undergoing a shift in frequency. In an amplifying device, photons of some given frequency are supplied by an external pump. These photons are scattered into modes with a lower frequency, the Stokes frequency; it is the light in the latter modes that is amplified. We shall refer to these modes as the signal modes since photons at the Stokes frequency can be introduced from outside the system as a species of signal; they then stimulate further inelastic scattering so that the incident signal is coherently amplified.

We shall consider below the particular case of inelastic scattering from molecular vibrations in a crystal, the coherent Raman effect.³ When an oscillating electromagnetic field is applied to a Raman active crystal, the resulting anharmonic molecular vibrations are able to transfer energy between oscillation modes of the radiation field whose frequencies differ by a resonant frequency of the molecules. The molecular vibrations here serve as the idler mode, while the two field modes are the pump and signal modes. Although the properties of the idler mode and the nature of nonlinear coupling may be rather different in other types of scattering processes, the corresponding amplification processes can, in a number of cases, still be treated by techniques similar to those we develop below.⁴

At optical frequencies, photons have enough energy to be individually detectable. The spontaneous emission of quanta, furthermore, is an important source of noise which cannot be incorporated into a classical discussion of the amplification process in any wholly satisfactory way. Thus, any thorough treatment of optical parametric amplification is necessarily a quantum-mechanical one. The mathematical analysis is simplified considerably by the

fact that only the signal and idler modes need be treated quantum mechanically; the pump mode may be assumed to be excited to classical intensities.

A simple model of the quantized parametric amplifier, which has been discussed in great detail in the literature,⁵ considers the nonlinear process to take place in a closed lossless cavity. Three of the normal modes are coupled by the interaction; one of them, the pump mode, is excited to a steady classical amplitude of oscillation. The equations of motion for the signal and idler modes are readily solved,^{5,6} and lead to an exponential growth of the signal- and idler-mode amplitudes. Such an exponential growth, however, cannot be sustained indefinitely. If the amplifier is completely lossless. the growth of the signal- and idler-mode amplitudes eventually demands more power than can be supplied by the pump, and the parametric approximation that its amplitude is fixed becomes untenable. When all three modes are treated as dynamical variables. the equations of motion are, of course, nonlinear, and energy supplied to the system oscillates back and forth between the modes.⁷

In most experimental situations, however, the amplifying system is neither closed nor lossless; its physical behavior cannot be adequately represented by the discrete mode models mentioned earlier. Open systems tend to achieve well-defined steady states in which the power supplied by the energy source is just equal to the power removed by damping and by radiation loss. Thus in an optical parametric amplifier the propagation of the amplified signal out of the active region carries with it a large fraction of the energy removed from the pump mode. The energy remaining behind as increased excitation of the idler mode is rapidly degraded to heat because of the strong damping experienced by phonon excitations.

Perhaps the simplest model that can take into account the effects of propagation and damping with the parametric approximation is an infinite uniform amplifying medium. In a semiclassical approximation the problem of the propagation of wave packets through the medium can readily be solved, and leads to well-behaved field amplitudes. ⁸ For fully quantized field amplitudes, however, the Stokes light emitted spontaneously at arbitrarily large distances from the point of observation is amplified indefinitely and leads, in the infinite medium, to signal intensities that diverge as a function of time. Only in a medium that is finite in extent can the field intensity reach a steady state.

We shall therefore consider a parametric amplifier consisting of a bounded Raman-active crystal illuminated by a powerful and steady laser beam which serves as the pump mode. In order to make the geometrical considerations as simple as possible, we shall assume that the crystal is a plane slab of material and that the laser beam is incident normally on one face. Any of the field modes into which Stokes scattering takes place can be considered to be a signal mode; thus there are signal modes with propagation vectors at all angles relative to the surface of the amplifying slab.

In order to provide the proper context for a quantum-mechanical description of the amplification, we discuss first, in Sec. II, the Hamiltonian for the system. We then construct, in Secs. III-V, the Heisenberg equations of motion for the signal and idler fields.

The interactions of the signal and idler fields with other excitations of the medium lead to damping of the fields; the optical phonons of the idler field, in particular, are strongly damped. We discuss this damping and the associated noise in Sec. VI; we neglect any direct damping of the signal field.

The resulting equation of motion for the radiation field⁹ is obtained in Sec. VII. This Heisenberg equation of motion for the signal field is identical in form to the classical equation in one spatial dimension x for an electric field E(x, t). In the classical context the exponential solutions of the initial value problem [E(x, 0) given] and of the boundary value problem [E(0, t) given] are equivalent; we discuss these solutions, which grow in time and in space, respectively, in Sec. VIII.

The quantum-mechanical problem, however, considered in dynamical terms is intrinsically an initial-value problem. We thus solve first the initialvalue problem for the homogeneous equation for arbitrary fields in Secs. IX and X, and then construct solutions for the inhomogeneous equations, using a Green's function, in Sec. XI. We finally consider the effects of the boundaries of the medium in Sec. XII. The solution for the signal field obtained in Sec. XII contains explicitly all the transient effects that obtain only for small times. We discuss the much simpler steady-state behavior of the amplified signal in Sec. XIII, and of the amplified noise in Sec. XIV.

Finally, in Sec. XV, we discuss the statistical properties of the output in terms of the quasiprobability distribution for the amplified field.

II. INTERACTION

In the phenomenological model of the Raman effect, ^{3,10} the molecular polarizability, which is in general a tensor α_{ij} , is regarded as a function of the positions of the nuclei within a molecule or a crystal unit cell. Since we shall be considering only one Raman line, we shall need to consider only one normal mode of molecular vibrations, i.e., one optical-phonon branch of the crystal. Thus, the polarizability is a function of the single vibrational

coordinate $\mathbf{\tilde{u}}(\mathbf{\tilde{r}}, t)$ which describes the oscillations in that mode of the molecule at $\mathbf{\tilde{r}}$.

The *u* dependence of α_{ij} implies that the susceptibility χ_{ij} and the dielectric constant ϵ_{ij} of the crystal are also functions of $\mathbf{u}(\mathbf{r}, t)$. Since the deviations of α , χ , or ϵ from their equilibrium (u = 0) values are quite small, it will suffice to retain only the lowest-order nonvanishing term in a power-series expansion. Thus, we consider only the linear contribution to α :

$$\alpha_{ij}(\mathbf{\tilde{r}},t) = \alpha_{ij} + \alpha_{ijk} u_k(\mathbf{\tilde{r}},t) ;$$

here i, j, k are vector indices, and we use the summation convention, intending a sum over all repeated indices.

In a crystal that has no center of inversion, the lowest-order contributions to χ and ϵ are again the ones linear in $\overline{u}(\mathbf{r}, t)$. In crystals that do have inversion symmetry, the linear contribution to the macroscopic parameters vanishes, and the lowestorder contributions come from terms quadratic in $\overline{u}(\mathbf{r}, t)$. In the following, we shall consider only crystals without a center of inversion, so that we may take

$$\chi_{ij}(\mathbf{\bar{r}}, t) = \chi_{ij} + \chi_{ijk} u_k(\mathbf{\bar{r}}, t) ,$$

$$\epsilon_{ij}(\mathbf{\bar{r}}, t) = \epsilon_{ij} + \chi_{ijk} u_k(\mathbf{\bar{r}}, t) .$$
(2.1)

We are using Heaviside-Lorentz units (i.e., rationalized Gaussian units) in which $\epsilon = 1 + \chi$. Since $\chi_{ij}(\vec{r}, t)$ is a symmetrical tensor, we have

 $\chi_{ijk} = \chi_{jik}$.

The constant term ϵ_{ij} of Eq. (2.1) may well be frequency dependent. We are, however, interested in only two bands of optical frequencies: One is the essentially monochromatic band of the pumping light, presumably provided by a laser; the other is the Raman line, centered about the Stokes frequency, whose width is roughly the same as the linewidth of the molecular vibrations.

It will therefore suffice to assume that ϵ_{ij} takes on constant, though possibly different, values over each of these frequency bands. We shall further assume that the linear dielectric constant is a scalar, $\epsilon_{ij} = \epsilon \delta_{ij}$.

The electric displacement within the crystal is

$$D_{i}(\mathbf{\ddot{r}}, t) = \epsilon_{ij}(\mathbf{\ddot{r}}, t) E_{j}(\mathbf{r}, t)$$
$$= \epsilon E_{i}(\mathbf{r}, t) + \chi_{ijk} E_{i}(\mathbf{\ddot{r}}, t) u_{k}(\mathbf{\ddot{r}}, t) .$$

In the absence of magnetic phenomena we have $\mu = 1$, so that the Hamiltonian density of the field within the crystal is

$$\frac{1}{2}D_{i}(\mathbf{\bar{r}}, t) E_{i}(\mathbf{\bar{r}}, t) + \frac{1}{2}B_{i}(\mathbf{\bar{r}}, t) B_{i}(\mathbf{\bar{r}}, t)$$

$$= \frac{1}{2}\epsilon E_{i}(\mathbf{\bar{r}}, t) E_{i}(\mathbf{\bar{r}}, t) + \frac{1}{2}B_{i}(\mathbf{\bar{r}}, t) B_{i}(\mathbf{\bar{r}}, t)$$

$$+ \frac{1}{2}\chi_{ijk}E_{i}(\mathbf{\bar{r}}, t) E_{j}(\mathbf{\bar{r}}, t) u_{k}(\mathbf{\bar{r}}, t) . \qquad (2.2)$$

The terms of this Hamiltonian density quadratic in the fields describe the linear response of the medium, which leads to reflection and refraction at the boundaries. These effects can be eliminated experimentally by immersing the crystal in an inactive medium with the same index of refraction. The linear response thus leads only to a phase velocity of the light different from c, and need not concern us further. The Hamiltonian density of Eq. (2. 2) can thus be regarded as the sum of a Hamiltonian density for the "freely" propagating field:

$$\mathcal{H}_{f} = \frac{1}{2} \epsilon [\vec{\mathbf{E}}(\vec{\mathbf{r}}, t)]^{2} + \frac{1}{2} [\vec{\mathbf{B}}(\vec{\mathbf{r}}, t)]^{2} , \qquad (2.3)$$

and a Hamiltonian density for the nonlinear interaction of the field and the crystal:

$$\mathcal{H}_{I} = \frac{1}{2} \chi_{ijk} E_{i}(\mathbf{\dot{r}}, t) E_{j}(\mathbf{\dot{r}}, t) u_{k}(\mathbf{\dot{r}}, t) . \qquad (2.4)$$

In the operation of the Raman-active crystal as a linear amplifier, the pump mode is strongly excited by, say, a laser beam to classical intensities, while the signal, i.e., Stokes light, and idler, i.e., phonon, modes remain weakly excited. Thus, all the states of the radiation field that we need to consider contain a strong coherent and nearly monochromatic excitation

$$\vec{\mathcal{E}}_L(r,t) \cong 2\vec{\mathcal{E}}_L \cos(\vec{\mathbf{k}}_L \cdot \vec{\mathbf{r}} - \omega_L t)$$

due to the laser field, in addition to whatever other excitations may be present. For a fairly wide range of laser field strengths the dynamical variations in the amplitude of the beam during its passage through the medium may be neglected. It is thus an excellent approximation, when the field modes occupied by the laser beam are in quantum states having a large coherent excitation, to regard the laser field not as a dynamical variable but as an externally given parameter of the system.

Since the behavior of the laser modes is predetermined in this approximation, it is convenient to separate the states and operators that refer to them from those that describe the dynamical behavior of the other modes. We can write the electric field, for example, as

$$\vec{\mathbf{E}}(\vec{\mathbf{r}},t) = \vec{\mathbf{E}}'(\vec{\mathbf{r}},t) + \vec{\mathbf{E}}_L(\vec{\mathbf{r}},t) , \qquad (2.5a)$$

where $\vec{\mathbf{E}}_{L}(\vec{\mathbf{r}}, t)$ is that part of the field which operates on states of the laser modes only, and $\vec{\mathbf{E}}'(\vec{\mathbf{r}}, t)$ operates on the states of all other modes of the field. It is the dynamics of the field $\vec{\mathbf{E}}'$ which interest us since it describes the scattered radiation. The laser field $\vec{\mathbf{E}}_{L}$ may, for all practical purposes, be replaced by its expectation value

$$\langle \vec{\mathbf{E}}_{L}(\vec{\mathbf{r}},t) \rangle = \hat{\mathcal{E}}_{L}(\vec{\mathbf{r}},t)$$
 (2.5b)

A corresponding separation may be made for the magnetic field \vec{B} as well.

The free-field Hamiltonian which corresponds to the Hamiltonian density given in Eq. (2.3) is

QUANTUM THEORY OF LIGHT PROPAGATION ···

$$\begin{split} H_f &= \int \mathcal{H}_f \, d^3 r \\ &= \int d^3 r \, \left\{ \frac{1}{2} \, \epsilon \big[\vec{\mathbf{E}'}(\vec{\mathbf{r}}, t) \big]^2 + \frac{1}{2} \big[\vec{\mathbf{B}'}(\vec{\mathbf{r}}, t) \big]^2 \right\} \\ &+ \int d^3 r \, \left\{ \frac{1}{2} \, \epsilon \big[\vec{\mathbf{E}}_L(\vec{\mathbf{r}}, t) \big]^2 + \frac{1}{2} \big[\vec{\mathbf{B}}_L(\vec{\mathbf{r}}, t) \big]^2 \right\} \end{split}$$

where we have made use of the fact that the laser modes are orthogonal to all other modes of the system. Since we shall use the Hamiltonian only to establish the equations of motion for the operators \vec{E}' and \vec{B}' , it will suffice to make use of a reduced form of the Hamiltonian that is the expectation value of H_f taken with respect to the state of the laser mode only. If the laser excitation is both large and coherent, as we have assumed, then in that expectation value we have, to an excellent approximation,

$$\langle [\vec{\mathbf{E}}_{L}(\vec{\mathbf{r}},t)]^{2} \rangle = [\vec{\mathcal{E}}_{L}(\vec{\mathbf{r}},t)]^{2} ,$$

$$\langle [\vec{\mathbf{B}}_{L}(\vec{\mathbf{r}},t)]^{2} \rangle = [\vec{\mathfrak{G}}_{L}(\vec{\mathbf{r}},t)]^{2} .$$

The reduced Hamiltonian defined in this way differs only by an additive c number from the Hamiltonian

$$H'_f = \int \mathcal{K}'_f \ d^3 \gamma$$

with

$$\mathcal{H}'_{f} = \frac{1}{2} \epsilon \left[\vec{E}'(\vec{r}, t) \right]^{2} + \frac{1}{2} \left[\vec{B}'(\vec{r}, t) \right]^{2} .$$
(2.6)

It is this form of the free-field Hamiltonian we shall use in Secs. IV and V to find the equation of motion for $\vec{\mathbf{E}}'(\vec{\mathbf{r}}, t)$.

It is, of course, also convenient to express the interaction Hamiltonian density in reduced form. From its definition, Eq. (2.4), and by making use of Eqs. (2.5a) and (2.5b) we see that this form is

$$3C_{I} = \frac{1}{2} \chi_{ijk} \mathcal{S}_{Li}(\vec{\mathbf{r}} t) \mathcal{S}_{Lj}(\vec{\mathbf{r}} t) u_{k}(\vec{\mathbf{r}} t) + \chi_{ijk} \mathcal{S}_{Li}(\vec{\mathbf{r}}, t) \mathcal{E}'_{j}(\vec{\mathbf{r}}, t) u_{k}(\vec{\mathbf{r}}, t) + \frac{1}{2} \chi_{ijk} \mathcal{E}'_{i}(\vec{\mathbf{r}}, t) \mathcal{E}'_{j}(\vec{\mathbf{r}}, t) u_{k}(\vec{\mathbf{r}}, t) .$$
(2.7)

The first term of Eq. (2.7) represents the forced motion of \vec{u} driven by the *c*-number laser field; the dc component of the driving force, i.e., of $\mathcal{E}_{Li}\mathcal{E}_{Lj}$, serves merely to provide a trivial shift of the zero point of molecular vibrations, while its remaining component at twice the laser frequency is so far from resonance that its effect is entirely negligible. The third term of Eq. (2.7) represents the nonlinear behavior of the Stokes light, and provides, for example, the generation of higher-order Stokes lines. However, unless the first Stokes line is amplified to a very large intensity indeed, these processes remain unimportant. That is, as long as the amplification of the Stokes light is not too great, it is true that

$$\langle [\vec{\mathbf{E}}']^2 \rangle \ll [\vec{\mathbf{\mathcal{E}}}_L]^2$$
,

so that the magnitude of the third term in \mathcal{H}_I is small compared to that of the second. For our purposes, then, we may take

$$\mathcal{K}_{I} \cong \chi_{ijk} \mathcal{E}_{Li}(\vec{\mathbf{r}}, t) E'_{j}(\vec{\mathbf{r}}, t) u_{k}(\vec{\mathbf{r}}, t) . \qquad (2.8)$$

Since we shall not need to refer further to the full electric field of Eq. (2.5), we shall in the following drop the primes from the dynamical field.

The interaction Hamiltonian

 $H_{I} = \int \mathcal{K}_{I} d^{3} \gamma$

given by Eq. (2.8) contains, in addition to slowly varying terms, terms that vary quite rapidly, oscillating at infrared, optical, and uv frequencies. It is well known that such rapidly oscillating terms tend to contribute little to physical results, since any contribution they make must come in higher orders of perturbation theory via energy-nonconserving virtual processes. In order to eliminate these terms from the Hamiltonian, it is convenient to write each of the fields $\vec{E}, \vec{\delta}_L$, and \vec{u} as a sum of positive- and negative-frequency components, where we define the positive-frequency parts $\vec{E}^{(*)}, \vec{\delta}_L^{(*)}$, and $\vec{u}^{(*)}$ to be those which vary as $e^{-i\omega t}$. Since the laser and phonon fields are nearly monochromatic, i.e.,

$$\vec{\mathcal{E}}_{L}^{(+)}(\vec{\mathbf{r}},t) \sim e^{-i\omega_{L}t}, \qquad \vec{\mathbf{u}}^{(+)}(\vec{\mathbf{r}},t) \sim e^{-i\omega_{0}t},$$

the only terms of \mathcal{H}_I that contain slowly varying, or resonant, contributions are

$$\begin{aligned} \mathcal{H}_{I}^{\prime} &= \chi_{ijk} \, \mathcal{E}_{Li}^{(+)}(\vec{\mathbf{r}}, t) E_{j}^{(-)}(\vec{\mathbf{r}}, t) u_{k}^{(-)}(\vec{\mathbf{r}}, t) + \mathrm{H.c.} \\ &+ \chi_{ijk} \, \mathcal{E}_{Li}^{(+)}(\vec{\mathbf{r}}, t) E_{j}^{(-)}(\vec{\mathbf{r}}, t) u_{k}^{(+)}(\vec{\mathbf{r}}, t) + \mathrm{H.c.} , \ (2.9) \end{aligned}$$

where H.c. represents the Hermitian conjugate of the previous term.

The first term of \mathcal{H}'_I describes the annihilation of a laser photon with the creation of a phonon and of a Stokes photon with frequency ω_s (aside from tiny line shifts associated with phonon damping $\omega_s \cong \omega_L$ $-\omega_0$; its Hermitian conjugate describes the inverse process in which a Stokes photon and a phonon are annihilated and a laser photon created. The second pair of terms describes the anti-Stokes process in which a laser photon and a phonon are annihilated (or created) and an anti-Stokes photon is created (or annihilated). Anti-Stokes light, of course, can never be emitted spontaneously; the emission must always be induced by the presence of optical phonons. Thus, unless the molecular vibrations are strongly excited, by, for instance, thermal processes or the Stokes process, the anti-Stokes process is unimportant. It is realistic then to neglect the anti-Stokes process entirely and to consider only the Hamiltonian density

$$\mathcal{H}_{s} = \chi_{ijk} \mathcal{E}_{Li}^{(+)}(\mathbf{r},t) E_{j}^{(-)}(\mathbf{r},t) u_{k}^{(-)}(\mathbf{r},t) + \mathrm{H.\,c.} \quad (2.10)$$

for the interaction of Stokes light with optical phonons.

In order to obtain a Hamiltonian density for the molecular vibrations, it will suffice to consider a simple phenomenological model of the crystal. At



FIG. 1. Typical optical-phonon spectrum for a crystal with lattice constant *a*. The resonance frequency of free molecules is ω_0 , and λ is an optical wavelength.

the comparatively long wavelengths involved in optical processes, the oscillations of molecules at neighboring lattice sites are nearly in phase. The intermolecular interactions that lead to a removal of the degeneracy of the crystal vibration frequencies for transverse oscillations are small in that case, and can be neglected. The resonance frequencies of all modes are then identical, and the crystal frequencies are independent of wave number. Optical phonon spectra, in other words, tend to be quite flat near $\vec{q} = 0$ (see Fig. 1); since optical wave vectors occupy only a very small volume about the origin of the reciprocal lattice, the dispersion curve can for all practical purposes be taken as flat.

Since we are neglecting the intermolecular interactions, the Hamiltonian of the material is just the sum of the free Hamiltonians for each unit cell:

$$H_{m} = \sum_{\alpha} \frac{1}{2} m \left[\left(\frac{\partial}{\partial t} \, \tilde{\mathbf{u}} \left(\tilde{\mathbf{r}}_{\alpha}, t \right) \right)^{2} + \omega_{0}^{2} \left[\tilde{\mathbf{u}} \left(\tilde{\mathbf{r}}_{\alpha}, t \right) \right]^{2} \right]$$

where m is an effective reduced mass for the normal mode of molecular oscillations we are considering, and α is a running index for the unit cells.

On the scale of optical wavelengths, the lattice may be regarded as a continuum, so that the sum over lattice sites \vec{r}_{α} can be replaced by an integral of a Hamiltonian density over the volume of the crystal:

$$H_m = \int d^3 \gamma \mathcal{H}_m,$$

with

$$\Im \mathcal{C}_{m} = \frac{1}{2} \rho \left(\left| \frac{\partial}{\partial t} \, \vec{u}(r, t) \right|^{2} + \omega_{0}^{2} \left| \vec{u}(r, t) \right|^{2} \right), \qquad (2.11)$$

The effective density ρ is simply the effective mass *m* divided by the volume of a unit cell.

The total Hamiltonian for the Raman process is

thus

$$H=\int \left(\mathfrak{H}_{f}'+\mathfrak{H}_{m}+\mathfrak{H}_{s}\right) d^{3}r,$$

where the field Hamiltonian density \mathscr{K}'_f is given by Eq. (2.6), the material Hamiltonian density \mathscr{K}_m is given by Eq. (2.11), and the interaction Hamiltonian density for Stokes-light generation \mathscr{K}_s is given by Eq. (2.10).

III. REDUCTION TO A ONE-DIMENSIONAL PROBLEM

The considerations of Secs. I and II have served to establish the energy density associated with the interaction of the electric and phonon fields. Which of the normal modes of oscillation of these fields actually interact must be determined by the boundary conditions imposed on the fields and nature of the volume within which the fields interact.

In case the amplifying medium is unbounded, translational symmetry requires that momentum be conserved by the interaction, i.e., that the sum of the wave vectors \vec{k} of the Stokes photon and \vec{q} of the phonon generated by the scattering be precisely equal to the wave vector \vec{k}_L of the laser light:

$$\vec{\mathbf{k}} + \vec{\mathbf{q}} = \vec{\mathbf{k}}_L \,. \tag{3.1}$$

Thus each electromagnetic propagation mode only interacts strongly with phonons having a single wave vector. If one further takes into account the fact that oscillating terms in the Hamiltonian contribute little to the observed scattering, i.e., that energy is conserved, one sees that the values of \vec{k} are restricted to those for which the frequency of the Stokes photons, $\omega(\vec{k})$, the frequency of the molecular vibrations, ω_0 , and the laser frequency, ω_L are related by

$$\omega(\mathbf{\bar{k}}) + \omega_0 = \omega_L \quad . \tag{3.2}$$

The observed wave vectors for the Stokes light generated in an infinite medium thus lie on the surface of constant $\omega(\vec{k})$ defined by Eq. (3.2); we will call this surface the Stokes sphere. The wave vectors of the corresponding optical phonons are determined by Eq. (3.1) (see Fig. 2).

The introduction of boundaries destroys the translational invariance of the medium, and momentum is no longer precisely conserved in the interaction. Each plane wave of Stokes light generated in a finite medium is thus coupled to a certain finite range of optical phonons whose wave vectors may differ from that given by Eq. (3.1) by amounts of the order of the reciprocal of the dimensions of the medium.

A typical experimental situation is shown in Fig. 3; the signal consists of Stokes light scattered in the direction labeled S. As we have noted earlier, we consider the amplifying medium to be a plane slab which extends to infinity in the direction transverse to the propagation of the laser beam, but only extends for a finite distance along the direction of

propagation of the beam.

Let us establish coordinates such that the laser beam propagates in the +x direction, and that the medium extends from x=0 to x=l, and to $y=\pm\infty$, $z=\pm\infty$. Further, let the laser beam be incident normally on the slab, so that

 $\vec{\mathcal{E}}_{L}^{(+)}\left(\vec{\mathbf{r}}, t\right) = \vec{\mathcal{E}}_{L}^{(+)}\left(x, t\right) .$

For this configuration, translational invariance requires that the y and z components of momentum be conserved exactly, while the x component need only be conserved, crudely speaking, to terms of $O(\hbar/l)$. Thus, Stokes photons with wave vector \mathbf{k} are coupled to phonons whose wave vectors \mathbf{q} have the same transverse components as $\mathbf{k} (q_y = -k_y)$, q_x $= -k_x$), but whose longitudinal components may lie within a diffuse region around $q_x = k_L - k_x \pm O(1/l)$ (see Fig. 4). Among the processes characterized by these vectors the energy-conserving ones are again those for which \mathbf{k} satisfies Eq. (3. 2); that condition and the precision with which it must be satisfied are, of course, in no way connected with the size of the medium.

With our choice of coordinates, the Hamiltonian of the interaction that leads to Raman scattering becomes

$$H_{s} = \int_{\mathbf{m} \cdot \mathbf{d} \cdot \mathbf{i} \mathbf{u} \mathbf{m}} \mathcal{K}_{s} d^{3} \gamma$$

= $\int d^{3} \gamma \lambda(x) \chi_{ijk} \mathcal{E}_{Li}^{(-)}(x, t) E_{J}^{(-)}(\mathbf{r}, t) u_{k}(\mathbf{r}, t)$
+ $H_{s} c_{s}$. (3.3)

where the function $\lambda(x)$ is the characteristic function of the medium:

$$\lambda(x) = 1$$
, $0 < x < l$
 $\lambda(x) = 0$, $x < 0$ or $x < l$. (3.4)



FIG. 2. Wave vector relationships in an infinite medium: \vec{k}_L is the laser wave vector, \vec{k} the wave vector of the generated Stokes light, and \vec{q} the wave vector of the optical phonon created by the Raman process.



FIG. 3. Arrangements to investigate stimulated Raman emission using (a) off-axis scattering and (b) forward scattering; L represents the laser beam, S the signal, and $M_{1/2}$ a half-silvered mirror. In (b), the amplified signal must be separated spectroscopically.

In order to make use of the fact that the transverse components of momentum are conserved, we express the electric and phonon fields in terms of their transverse Fourier transforms. To simplify later notation we consider the Fourier transform of the electric field in terms of propagating waves rather than stationary plane waves:

$$E_{i}^{(*)}(\mathbf{\vec{r}}, t) = \int \left[d^{2}K/(2\pi)^{2} \right] E_{i}^{(*)}(\mathbf{\vec{K}}; x, t) e^{i(\mathbf{\vec{K}}\cdot\mathbf{\vec{r}}_{T}-\omega_{T}t)},$$

$$u_{i}^{(*)}(\mathbf{\vec{r}}, t) = \int \left[d^{2}Q/(2\pi)^{2} \right] u_{i}^{(*)}(\mathbf{\vec{Q}}; x, t) e^{i\mathbf{\vec{Q}}\cdot\mathbf{\vec{r}}_{T}},$$
(3.5)

where

$$\vec{\mathbf{K}} = (k_{\mathbf{y}}, k_{\mathbf{g}}), \quad \vec{\mathbf{Q}} = (q_{\mathbf{y}}, q_{\mathbf{g}}), \quad \vec{\mathbf{r}}_T = (y, z) \quad (3.6)$$

are purely transverse vectors.

The frequencies $\omega(\vec{k})$ which are improtant for the amplification process all tend to lie close to the Stokes frequency, and the corresponding wave vectors \vec{k} all lie close to the Stokes sphere (see Fig. 4). As we shall show in more detail in Sec. V the frequency function $\omega(\vec{k})$ can, in the vicinity of the Stokes sphere, be written as the sum of two terms:

$$\omega(\mathbf{\bar{k}}) = \omega_{\mathbf{k}} + \omega_{\mathbf{T}} \quad . \tag{3.7}$$

The frequency ω_k depends only on k, the longitudinal component of \vec{k} , while ω_T depends only on the constant transverse wave vector \vec{K} . The explicit functional forms of these frequencies are derived in Eqs. (5.3)-(5.5). For the present we shall simply use Eq. (3.7) to make explicit the separation of the longitudinal and transverse parts of the field.

By introducing the expressions for the fields from Eqs. (3.5) into Eq. (3.3) and performing the integration over \mathbf{r}_T , we obtain

$$\begin{split} H_{s} &= \int \left[d^{3}K / \chi_{ijk} \mathcal{E}_{Li}^{(-)}(x, t) \right. \\ &\times \int \left[d^{2}K / (2\pi)^{2} \right] \left[d^{2}Q / (2\pi)^{2} \right] E_{j}^{(+)}(\vec{\mathbf{K}}; x, t) e^{-i\omega Tt} \\ &\times u_{k}^{(+)}(\vec{\mathbf{Q}}; x, t) (2\pi)^{2} \delta^{2}(\vec{\mathbf{K}} + \vec{\mathbf{Q}}) + \mathrm{H.\,c.} \\ &= \int dx \,\lambda(x) \,\chi_{ijk} \,\mathcal{E}_{Li}^{(-)}(x, t) \int \left[d^{2}K / (2\pi)^{2} \right] \\ &\times E_{j}^{(-)}(\vec{\mathbf{K}}; x, t) \,u_{k}^{(-)}(-\vec{\mathbf{K}}; x, t) e^{-i\omega Tt} + \mathrm{H.\,c.} \end{split}$$

The interaction Hamiltonian can thus be written as a sum of independent terms

$$H_{s} = \int d^{2}K H_{s} (\mathbf{K}) ,$$

. .



FIG. 4. Wave-vector relationships in a slab of thickness l; notation as in Fig. 2. Stokes light with wave vector \mathbf{k} is coupled to phonons whose wave vectors \mathbf{q} start anywhere within the diffuse segment centered around the tip of \mathbf{k} . Transverse components of \mathbf{k} and \mathbf{q} are equal in magnitude and opposite in direction. The energy-conserving processes are those for which \mathbf{k} terminates on the surface given by $\omega(\mathbf{k}) = \omega_L - \omega_0$.

where

$$H_{s}(\vec{\mathbf{K}}) = (\chi_{ijk}/4\pi^{2}) \int dx \,\lambda(x) \,\mathcal{S}_{Li}^{(-)}(x, t)$$
$$\times E_{j}^{(*)}(\vec{\mathbf{K}}; x, t) u_{k}^{(*)}(-\vec{\mathbf{K}}; x, t) e^{-i\omega Tt} + \text{H.c.}$$
(3.8)

The three-dimensional problem specified by H_s can thus be separated into a set of independent onedimensional problems corresponding to precise values of the transverse wave vector K. The Hamiltonians for these one-dimensional problems, $H_s(K)$, are all quite similar in structure, and it will suffice to consider a single such one-dimensional problem corresponding to a particular choice of K. We shall in the following, therefore, drop all reference to the transverse wave vector K. It is possible at any stage to return to the consideration of the full set of coupled modes by using, in effect, a bundle of one-dimensional solutions to solve the three-dimensional problem.

For purposes of quantization, it will be convenient to consider the crystal to have a large but finite area A; the values of \vec{K} are then not continuous but discrete, and integrals over \vec{K} must be replaced by $(4\pi^2/A)\sum_{K}$. The Hamiltonian then becomes

$$H_{s} = \sum_{K} H_{s}(\vec{K}),$$

where
$$H_{s}(\vec{K}) = A^{-1} \chi_{ijk} \int dx \lambda(x) \delta_{Li}^{(-)}(x, t) E_{j}^{(+)}(x, t)$$
$$\times u_{k}^{(+)}(x, t) e^{-i\omega Tt} + \text{H. c.}$$
(3.9)

IV. COMMUTATION RELATIONS

We now proceed to determine the equations of motion generated by the Hamiltonian of Eqs. (2.2), (2.4), and (3.9). Since the fields \vec{E} and \vec{u} are quantized, it will be convenient to expand them in terms of normalized boson operators. We shall suppose that the electric field propagates in a region of total length $L \gg l$ whose ends impose periodic boundary conditions. The amplifying medium, as we have said, extends from x=0 to x=l; we shall suppose that its boundaries are such that the normal modes of the phonon field are standing waves. The fields \vec{E} and \vec{u} can then be written as

$$\vec{\mathbf{E}}^{(+)}(x,t) = \sum_{k\lambda} i \left(\frac{\hbar\omega(\vec{k})A}{2L\epsilon}\right)^{1/2} \vec{\mathbf{e}}^{\lambda}(\vec{k}) a_{k\lambda}(t) e^{i(kx-\omega_k t)}, \quad (4.1)$$
$$\vec{\mathbf{u}}^{(+)}(x,t) = \sum_{q\mu} \left(\frac{\hbar A}{l\rho\omega_0}\right)^{1/2} \vec{\mathbf{g}}^{\mu}(\vec{\mathbf{q}}) b_{q\mu}(t) \sin qx e^{-i\omega_0 t} \lambda(x). \quad (4.2)$$

Here $\vec{e}^{\lambda}(\vec{k})$ and $\vec{g}^{\mu}(\vec{q})$ are unit polarization vectors, and their superscripts are polarization indices. The allowed wave numbers k are

 $k = 2\pi n/L$, $n = 0, \pm 1, \pm 2, \ldots$

The allowed values of q are

$$q = \pi n/l$$
, $n = 0, \pm 1, \pm 2, \ldots$

The factor \sqrt{A} , where A is the (large) transverse area of the medium which appears in the normalization factor in Eqs. (4.1) and (4.2) arises from the definition of $\vec{E}(\vec{K}; x, t)$, here written as $\vec{E}(x, t)$.

We have included explicitly in Eqs. (4.1) and (4.2) the time dependence of the free fields. That is, $\vec{E}^{(+)}(x, t)$ and $\vec{u}^{(+)}(x, t)$ are Heisenberg-picture operators, but the Heisenberg-picture boson amplitude operators are written as

$$a_{k\lambda}e^{-i\omega(\tilde{\mathbf{k}})t} = a_{k\lambda}e^{-i(\omega_k+\omega_T)t},$$

$$b_{q\mu}e^{-i\omega_0t}.$$

In the absence of couplings, then, the operators $a_{\lambda\lambda}$ and $b_{q\mu}$ are time independent. In writing Eq. (4.1) we have made use of Eq. (3.7). Since the factor $e^{-i\omega_T t}$ has already been included in our definition of $\vec{E}(\vec{K}; x, t)$ [Eq. (3.5)] it does not appear in Eq. (4.1). The boson operators of Eqs. (4.1) and (4.2) are the canonical operators, so that we have

$$\left[a_{k\lambda}(t), a_{k'\lambda'}^{\dagger}(t)\right] = \delta_{kk'} \delta_{\lambda\lambda'}, \qquad (4.3)$$

$$\left[b_{q\mu}(t), b_{q'\mu'}^{\dagger}(t)\right] = \delta_{qq'} \delta_{\lambda\lambda'}, \qquad (4.4)$$

while all other commutators vanish. The field commutators can be computed directly from these. For the electric field, we have

$$[E_i^{(+)}(x,t), E_j^{(-)}(x',t)]$$

$$=\frac{\hbar A}{2L\epsilon}\sum_{k}\omega(\vec{k})\delta_{ij}^{T}e^{ik(x-x')}, \qquad (4.5)$$

where δ_{ij}^{T} is the transverse part of the Kronecker δ :

$$\delta_{ij}^{T} = \sum_{\lambda=1}^{2} e_{i}^{\lambda} (\mathbf{k}) e_{j}^{\lambda} (\mathbf{k})^{*} = \delta_{ij} - k_{i} k_{j} / \mathbf{k}^{2} , \qquad (4.6)$$

which has the property

$$\delta_{ij}^T e_j^{\lambda}(\vec{\mathbf{k}}) = e_i^{\lambda}(\vec{\mathbf{k}}) .$$
(4.7)

For the phonon field, we find

$$\begin{bmatrix} u_i^{(+)}(x,t), u_j^{(-)}(x',t) \end{bmatrix}$$
$$= \frac{\hbar A}{l\rho\omega_0} \lambda(x)\lambda(x') \sum_q \delta_{ij}^U \sin qx \sin qx', \qquad (4.8)$$

where, in analogy with δ_{ij}^{T} ,

$$\delta^{U}_{ij} = \sum_{\mu} g^{\mu}_{i}(\mathbf{q}) g^{\mu}_{j}(\mathbf{q})^{*}, \qquad (4.9)$$

$$\delta^{U}_{ij}g^{\mu}_{j}(\mathbf{\bar{q}}) = g^{\mu}_{i}(\mathbf{\bar{q}}); \qquad (4.10)$$

since the phonon field $\tilde{u}(x,t)$ describes only one optical-phonon branch, and since longitudinal and transverse optical phonons are, in general, not degenerate, the sum of Eq. (4.9) does not extend over a complete set of polarizations, so that $\delta_{ij}^U \neq \delta_{ij}$.

Because of the orthogonality of the various mode functions over their respective volumes, the free Hamiltonians, given by the densities of Eqs. (2.3) and (2.11), assume a simple form in terms of the boson operators:

$$H'_{f} = A^{-1} \int_{L} \mathfrak{K}_{f} dx = \hbar \sum_{k}' \omega(\mathbf{k}) a_{k}^{\dagger}(t) a_{k}(t) , \qquad (4.11)$$

$$H_m = A^{-1} \int_I \mathcal{K}_m \, dx = \hbar \sum_q \omega_0 b_q^{\dagger}(t) \, b_q(t) \, . \tag{4.12}$$

We have dropped the zero-point contributions to these Hamiltonians, and \sum_{k}' sums over all modes other than the laser modes.

Since the functions e^{ikx} and $\sin qx$ are in general not orthogonal over the volume of the medium, the interaction Hamiltonian of Eq. (3.9) assumes a somewhat more complex form:

$$H_{s} = A^{-1} \int \mathcal{H}_{s} dx$$

$$= i\hbar \frac{\chi_{i \ im}}{(2Ll \epsilon \rho)^{1/2}} \sum_{\substack{kq \\ \mu\lambda}} \left(\frac{\omega(\tilde{k})}{\omega_{0}} \right)^{1/2}$$

$$\times e_{j}^{\lambda}(\tilde{k}) g_{m}^{\mu}(q) a_{k\lambda}(t) b_{q\mu}(t) \int dx \lambda(x) \sin qx$$

$$\times \mathcal{S}_{Li}^{(-)}(x, t) e^{ikx - i(\omega_{k} + \omega_{0} + \omega_{T})t} + \text{H.c.} \qquad (4.13)$$

As we mentioned in Sec. III, the integral gives nonvanishing contributions for a range of pairs of values for k and q, even when the laser field is perfectly monochromatic.

The equations of motion of the fields are, of course, determined by the commutators of the fields with the total Hamiltonian. These commutators can be found readily from the expressions above.

For the molecular vibrations, one may use either Eqs. (4.12) and (4.9) or Eqs. (3.9) and (4.8) together with the orthogonality of the functions $\sin qx$ to obtain

$$[u_i^{(+)}(x,t), H_m] = \hbar \omega_0 u_i^{(+)}(x,t) .$$
(4.14)

For the electric field, the corresponding expression is

$$\begin{bmatrix} E_i^{(*)}(x,t), H_f \end{bmatrix}$$

= $i\hbar \sum_{k\lambda} \omega(\vec{k}) \left(\frac{\hbar \omega(\vec{k})A}{2L\epsilon}\right)^{1/2} e_i^{\lambda}(\vec{k}) a_{k\lambda}(t) e^{i(kx-\omega_k t)}.$
(4.15)

These commutators give rise to the free-field equations of motion.

The commutator of the electric field with the interaction Hamiltonian is, from Eqs. (4.5) and (3.9),

$$\begin{bmatrix} E_j^{(+)}(x,t), H_s \end{bmatrix} = (\hbar/2\epsilon) \int dx' \lambda(x') \chi_{ilm}^* \mathcal{S}_{\ell i}^{(+)}(x',t) \\ \times u_m^{(-)}(x',t) e^{*i\omega_T t} \sum_k (1/L) \omega(\mathbf{k}) \delta_{jl}^T e^{ik(\mathbf{x}-\mathbf{x}')}. \quad (4.16)$$

Now, as we have argued before, the only photon modes which contribute appreciably to the emitted Stokes light are those whose frequencies are within a few Raman linewidths of the Stokes frequency ω_s [apart from tiny line shifts to be considered in Sec. VI, $\omega_s = \omega_L - \omega_0$; see also Eq. (7.6)]. Thus, the factor $\omega(\mathbf{k})$ in Eq. (4.16) is nearly a constant, ω_s for all those terms that contribute to the process. Furthermore, for any given transverse wave vector $\mathbf{\vec{k}}$ the direction of the emitted Stokes light is quite sharply defined by Eq. (3.2), so that the transverse part of the Kronecker δ may also be taken as constant. The sum thus reduces to one that can be performed using the completeness relationship

$$\sum_{k} e^{ik(x-x')} = L \,\delta(x-x'),$$

$$|x| < \frac{1}{2}L, \quad |x'| < \frac{1}{2}L. \quad (4.17)$$

The commutation relationship then becomes

$$[E_j^{(+)}(x,t),H_s]$$

$$= \hbar(\omega_s/2\epsilon)\overline{\chi}^*_{ijm}\mathcal{E}^{(+)}_{Li}(x,t)u^{(-)}_m(x,t)\lambda(x)e^{i\,\omega_T t}, \quad (4.18)$$

where we have introduced the abbreviation

$$\overline{\chi}_{ijm} = \chi_{ilm} \delta_{jl}^T$$

The commutator of \vec{u} with the interaction Hamiltonain is, from Eqs. (3.9) and (4.8),

$$\begin{bmatrix} u_k^{(*)}(x,t), H_s \end{bmatrix} = (\hbar/\rho\omega_0)\chi_{ijm}^* \int dx'\lambda(x)\lambda(x')$$
$$\times \mathcal{E}_{Li}^{(*)}(x',t)\mathcal{E}_{j}^{(-)}(x',t)e^{i\omega_T t} \sum_q \delta_{km}^U \operatorname{sin} qx \operatorname{sin} qx'.$$

Now, just as δ_{ij}^T is a function of k, δ_{ij}^U is a function of q. However, the direction of the phonon wave vectors is nearly as well defined as that of the photon wave vectors, so that δ_{ij}^U may be taken as con-

stant for all those values of q that contribute to the Stokes process. The sum can then be performed using the completeness relationship

$$\lambda(x)\lambda(x')\sum_{a}\sin qx\sin qx'=\frac{1}{2}l\lambda(x)\delta(x-x'). \quad (4.19)$$

Thus, by writing

 $\tilde{\chi}_{ijk} = \chi_{ijm} \delta^U_{km} ,$

we may express the commutator for the phonon field as

$$[u_k^{(+)}(x,t),H_s]$$

= $(\hbar/l\rho\omega_0)\tilde{\chi}^*_{ijk}\mathcal{E}_{Li}^{(+)}(x,t)E_j^{(-)}(x,t)\lambda(x)e^{+i\omega_T t}$. (4.20)
V. EQUATIONS OF MOTION

The equations of motion of the fields are determined by the commutators of Eqs. (4.14), (4.15), (4.18), and (4.20). For the phonon field, we have

$$\frac{\partial}{\partial t} u_i^{(+)}(x,t) = \frac{1}{i\hbar} \left[u_i^{(+)}(x,t), H_m + H_s \right]$$
$$= -i \omega_0 u_i^{(+)}(x,t) - \frac{i}{2\rho \omega_0} \tilde{\chi}^*_{mji} \mathcal{E}_{Lm}^{(+)}(x,t) \times E_j^{(-)}(x,t) e^{i\omega_T t} \lambda(x) .$$
(5.1)

Because of our separation of the factor $e^{-i\omega_T t}$, the field $E_j^{(*)}(x, t)$ is explicitly as well as implicitly time dependent, so that

$$\frac{\partial}{\partial t} E_{j}^{(*)}(x,t) = \frac{1}{i\hbar} \left[E_{j}^{(*)}(x,t), H_{f} + H_{s} \right] - i\omega_{T} E_{j}^{(*)}(x,t)$$

$$= \sum_{k\lambda} \omega(\vec{k}) \left(\frac{\hbar \omega(\vec{k}) A}{2L\epsilon} \right)^{1/2} e_{j}^{(\lambda)}(k) a_{k\lambda}(t)$$

$$\times e^{i(kx - \omega_{k}t)} - i\omega_{T} E_{j}^{(*)}(x,t) - \frac{i\omega_{s}}{2\epsilon} \vec{\chi}_{ijm}^{*}$$

$$\times \mathcal{S}_{Li}^{(*)}(x,t) u_{m}^{(-)}(x,t) e^{i\omega_{T}t} \lambda(x) .$$

Since $\omega(\vec{k}) = \omega_{\tau} + \omega_{\nu}$, this becomes

$$\frac{\partial}{\partial t} E_{j}^{(+)}(x,t) = \sum_{k\lambda} \omega_{k} \left(\frac{i \overline{v} \omega(\overline{k}) A}{2L \epsilon}\right)^{1/2} e_{j}^{(\lambda)}(k) a_{k\lambda}(t)$$

$$\times e^{i (kx - \omega_{k} t)} - \frac{i \omega_{s}}{2\epsilon} \overline{\chi}_{ijm}^{*} \mathcal{E}_{Li}^{(+)}(x,t)$$

$$\times u_{m}^{(-)}(x,t) e^{i \omega T^{t}} \lambda(x) . \qquad (5.2)$$

In order to simplify Eq. (5.2), it is necessary to specify a dispersion law for the Stokes light. Since the only frequencies of interest lie within a small number of Raman linewidths of the Stokes frequency, we may assume a linear relationship between the frequency and the wave vector. Furthermore the dielectric constant does not change over the range of frequencies involved, so that

$$[\omega(\vec{k})]^2 = (1/\epsilon) c^2 (k^2 + \vec{K}^2) , \qquad (5.3)$$

where k is the longitudinal component of the wave

vector \vec{k} , and \vec{K} is its fixed transverse component from Eq. (3.8). For $\omega(\vec{k})$ very near ω_s , the magnitude of k is very nearly

$$k_{s} = (\epsilon \omega_{s}^{2} / c^{2} - \vec{\mathbf{K}}^{2})^{1/2} .$$
 (5.4)

If we expand $\omega(\vec{k})$ as given by Eq. (5.3) to lowest order in $|k| - k_s$, we obtain

$$\omega(\vec{\mathbf{k}}) = \omega_s + (c^2 k_s / \epsilon \omega_s) \left(\left| k \right| - k_s \right)$$
$$= (c^2 / \epsilon) \left[\vec{\mathbf{k}}^2 / \omega_s + \left| k \right| k_s / \omega_s \right]$$

The constant term representing the transverse propagation is thus

$$\omega_T = c^2 \mathbf{\vec{K}}^2 / \epsilon \omega_s ,$$

while the term associated with the longitudinal propagation is

$$\omega_{k} = v \left| k \right| , \qquad (5.5)$$

where we have denoted by

$$v = (c/\epsilon) (k_s/\omega_s)$$

the x component of the wave velocity of Stokes light in the crystal.

With the dispersion law of Eq. (5.5), the sum over k, λ in Eq. (5.2) becomes

$$\frac{1}{i\hbar} \left[E_{j}^{(+)}(x,t), H_{f} \right]$$

$$= \sum_{k\lambda} v \left| k \right| \left(\frac{\hbar \omega(\vec{k})A}{2L \epsilon} \right)^{1/2} e_{j}^{\lambda}(k) a_{k\lambda}(t) e^{i(kx - \omega_{k}t)}$$

$$= v \sum_{k>0; \lambda} k \left(\frac{\hbar \omega(\vec{k})A}{2L \epsilon} \right)^{1/2} e_{j}^{\lambda}(k) a_{k\lambda}(t) e^{ik(x - vt)}$$

$$- v \sum_{k<0; \lambda} k \left(\frac{\hbar \omega(\vec{k})A}{2L \epsilon} \right)^{1/2} e_{j}^{\lambda}(k) a_{k\lambda}(t) e^{ik(x + vt)}.$$
(5.6)

The form of Eq. (5.6) suggests a separation of the electric field into a forward-traveling component

$$\vec{\mathbf{E}}_{F}^{(*)}(x,t) = \sum_{k>0,\lambda} i \left(\frac{\hbar \omega(\vec{\mathbf{k}})A}{2L\epsilon}\right)^{1/2} \vec{\mathbf{e}}^{\lambda}(k) a_{k\lambda}(t)$$
$$\times e^{ik(x-vt)} , \qquad (5.7)$$

and a backward-traveling component

$$\vec{\mathbf{E}}_{B}^{(*)}(\mathbf{x},t) = \sum_{k<0,\lambda} i \left(\frac{\hbar \omega(\vec{\mathbf{k}})A}{2L\epsilon}\right)^{1/2} \vec{\mathbf{e}}^{\lambda}(k) a_{k\lambda}(t) \\ \times e^{ik(x+\nu t)} .$$
(5.8)

The commutator of Eq. (5.6) can then be written

$$\frac{1}{i\hbar} \left[E_j^{(*)}(x,t), H_f \right] = -v \frac{\partial}{\partial \chi} E_{Fj}^{(*)}(x,t) +v \frac{\partial}{\partial \chi} E_{Bj}^{(*)}(x,t) .$$
(5.9)

The separation of the field into forward- and backward-propagating parts suggests the approximation of writing the equation of motion for the electric field, Eq. (5.2), as a pair of uncoupled first-order differential equations, one for each part. In the case of the noninteracting field this separation is exact and leads to the individual equations of motion

$$\frac{\partial}{\partial t} E_F^{(+)}(x,t) = -v \frac{\partial}{\partial x} E_F^{(+)}(x,t) ,$$
$$\frac{\partial}{\partial t} E_B^{(+)}(x,t) = +v \frac{\partial}{\partial x} E_B^{(+)}(x,t) .$$

In order to write first-order equations of motion for the interacting fields, it is necessary to separate the interaction term also into forward- and backward-propagating parts. Now as we saw earlier, the only values of k for which there is an appreciable contribution to the Stokes light are those for which

$$k \cong \pm k_s$$
,

with k_s given by Eq. (5.4). Because of the finite thickness of the medium, each mode k of the Stokes light is coupled to a range of phonon modes whose wave vectors q are equal to $k_L - k \pm O(1/l)$. Each of these phonons modes is in turn coupled to a similarly diffuse set of photon modes. Thus, we have a set of photon modes with

 $k = +k_s$,

which is coupled to a set of phonon modes with

$$q \cong k_L - k_s \pm O(1/l)$$

and another set of photon modes with

 $k \cong -k$

coupled to phonon modes with

 $q \cong k_L + k_s \pm O\left(1/l\right)$

(see Fig. 5). As long as the direction of observation is not perpendicular to the laser beam and as long as l is sufficiently large, the two sets of photon modes are completely disjoint, as are the two sets of phonon modes. That is, there is no appreciable coupling between photons near $+k_s$ and phonons near $k_L + k_s$, and, conversely, no coupling between photons near $-k_s$ and phonons near $k_L - k_s$.

Thus, if we define a separation of the phonon field into the parts¹¹

$$\vec{\mathbf{u}}_{F}^{(*)}(x,t) = \sum_{q < k_{L}; \mu} \left(\frac{\hbar A}{l\rho\omega_{0}}\right)^{1/2} \vec{\mathbf{g}}^{\mu}(q) b_{q\mu}(t) e^{-i\omega_{0}t}$$

$$\times \sin q x \lambda(x)$$
, (5.10)

$$\vec{\mathbf{u}}_{B}^{(+)}(x,t) = \sum_{q > k_{L}; \mu} \left(\frac{\hbar A}{l \rho \omega_{0}}\right)^{1/2} \vec{\mathbf{g}}^{\mu}(q) b_{q\mu}(t) e^{-i\omega_{0}t}$$

$$\times \operatorname{sing} x \lambda(x) \tag{5.11}$$

$$\sin q x \lambda(x) , \qquad (5.11)$$



FIG. 5. Wave-vector relationships for forward- and backward-propagating Stokes light. Stokes light with wave vectors \mathbf{k}_{F} and \mathbf{k}_{B} is emitted along and against the laser light of wave vector \bar{k}_L . The Stokes light in these two beams is coupled to optical phonons whose wave vectors originate in the diffuse segments around \mathbf{q}_F and \mathbf{q}_B , respectively, indicated in the figure. The magnitude of the x component of \vec{k}_F and of \vec{k}_B is k_s , given by Eq. (5.4).

then \overline{u}_F contains the contributions from modes with $q \cong k_L - k_s$, while \vec{u}_B contains the contributions from modes with $q \cong k_L + k_s$, so that u_F is coupled only to $\mathbf{\tilde{E}}_{F}$, and \mathbf{u}_{B} is coupled only to $\mathbf{\tilde{E}}_{B}$.

With the separations of the fields given by Eqs. (5.7), (5.8), (5.10), and (5.11), the equations of motion, Eqs. (5.1) and (5.2), become, for E_F and u_F ,

$$\frac{\partial}{\partial t} u_{Fi}^{(*)}(x,t) = -i \omega_0 u_{Fi}^{(*)}(x,t) - i \frac{1}{2\rho \omega_0} \tilde{\chi}_{mji}^* \mathcal{E}_{Lm}^{(*)}(x,t) E_{Fj}^{(*)}(x,t) \lambda(x) e^{i\omega} \tau^t ,$$
(5.12)

$$\frac{\partial}{\partial t} E_{Fi}^{(+)}(x,t) = -v \frac{\partial}{\partial x} E_{Fi}^{(+)}(x,t)$$
$$-i \frac{\omega_s}{2\epsilon} \overline{\chi}_{mji}^* \mathcal{S}_{Lm}^{(+)}(x,t) u_{Fi}^{(-)}(x,t) \lambda(x) e^{i\omega_T t} ; \quad (5.13)$$

and for E_B and u_B they are

$$\frac{\partial}{\partial t} u_{Bi}^{(+)}(x,t) = -i \omega_0 u_{Bi}^{(+)}(x,t) - i \frac{1}{2\rho \omega_0} \tilde{\chi}_{mji}^* \mathcal{E}_{Lm}^{(+)}(x,t) E_{Bj}^{(-)}(x,t) \lambda(x) e^{i\omega_T t},$$
(5.14)

$$\frac{\partial}{\partial t} E_{Bj}^{(+)}(x,t) = v \frac{\partial}{\partial x} E_{Bj}^{(+)}(x,t) - i \frac{\omega_s}{2\epsilon} \overline{\chi}_{mji}^* \mathcal{S}_{Lm}^{(+)}(x,t) u_{Bi}^{(-)}(x,t) \lambda(x) e^{i\omega \tau t} .$$
(5.15)

Since these pairs of equations differ only in the sign of v, we shall consider only the forward-propagating case, Eqs. (5.12) and (5.13). Our results can be rewritten for the backward-propagating case simply

by changing the sign of v and appropriately adjusting given equation of motion of the form¹² the boundary conditions.

VI. PHONON DAMPING

Our equations of motion [Eqs. (5.12) and (5.13)] are not yet complete. As we mentioned in the Introduction, there are two important mechanisms for removing energy from the interacting modes and thus preventing an explosive growth of the amplified fields: the radiation of Stokes light out of the crystal, and phonon damping. The traveling wave model we are considering treats the escape of Stokes light quite naturally. Phonon excitations, however, do not escape from the crystal; in the absence of damping they tend to accumulate within it. The increased scattering they stimulate prevents a steady state from being reached, and we must therefore include a damping mechanism for the phonons in our model.

Molecular vibrations in a crystal are strongly damped through their coupling to a vast number of other crystal excitation modes; these may include modes of acoustical vibrations, electronic excitations, other species of molecular vibrations, and so forth. We shall assume that these other excitation modes serve as a thermal reservoir for the optical-phonon modes described by the field $\mathbf{\tilde{u}}(\mathbf{\tilde{r}}, t)$. These other excitations, we assume, are in a state of thermal equilibrium; the strong interaction of the optical phonons with these modes tends to establish thermal equilibrium between them, while the amplification process tends to upset thermal equilibrium. One of our aims is to describe the way in which a nonequilibrium steady state is eventually reached. It will suffice for our purposes to consider a thermal reservoir consisting of any large number of modes of excitation of the crystal, having a sufficiently dense spectrum of frequencies that energy transferred to the heat bath from the optical phonons is then shared among so many excitation modes that it does not return to reexcite the modes from which it originally came. Detailed models having these properties have been considered extensively in the literature.¹²

However, no thermal reservoir can be an entirely passive absorber. The statistical fluctuations inherent in any large system themselves serve as sources of excitation for the optical phonons. That is, the damping mechanism can heat up the optical phonon modes as well as cool them down; the introduction of a damping mechanism inevitably introduces fluctuations as well.

Thus, if the free phonon field $\overline{u}(x, t)$ satisfies the Heisenberg equation of motion

$$\left(\begin{array}{c}\frac{\partial}{\partial t}+i\omega_0\end{array}\right)\mathbf{\bar{u}}^{(+)}(x,\,t)=0$$

then the damped phonon field must satisfy a Lan-

$$\left(\frac{\partial}{\partial t} + i\omega_0\right) \mathbf{\tilde{u}}^{(*)}(x,t) = -(\gamma + i\nu) \mathbf{\tilde{u}}^{(*)}(x,t) + \mathbf{\tilde{v}}^{(*)}(x,t) .$$
(6.1)

Here γ is the real positive damping constant, ν is the associated small frequency shift, and $\mathbf{\bar{v}}^{(*)}(x, t)$ is an operator representing the positive-frequency part of the fluctuating forces exerted by the heat bath on the optical phonons.

The dynamical properties of $\vec{v}(x, t)$ can, for our purposes, be given by specifying the commutator at unequal times:

$$[v_i^{(-)}(x,t), v_j^{(+)}(x',t')]$$

The thermal reservoir contains a vast number of modes of excitation, and we shall assume that its spectrum of frequencies and wave numbers is essentially continuous and flat in the vicinity of ω_0 . The commutator tends then to be quite sharply peaked in both the space and time coordinates. We shall further assume that the commutator is a c number and effectively takes the form

$$\begin{bmatrix} v_i^{(-)}(x, t), v_j^{(+)}(x', t') \end{bmatrix}$$

= $(\hbar/A) \Gamma \delta_{ij} \delta(x - x') \delta(t - t')$. (6.2)

The constant Γ is a measure of the strength of the fluctuating forces. Equation (6.2) can, indeed, be the exact one when the reservoir modes have nonpropagating boson excitations. By using the commutator of Eq. (6.2) we are, in effect, using the Markoffian or short-relaxation-time approximation for the fluctuating forces.¹²

Since the damping process is a linear one, its effect can simply be added to those of the coupling between phonons and Stokes light. The equations of motion for the forward Stokes processes thus are

$$\left(\frac{\partial}{\partial t} + i\overline{\omega_0} + \gamma\right) u_{Fi}^{(*)}(x, t)$$

$$= -i(1/2\rho\omega_0) \widetilde{\chi}_{mji}^* \mathcal{S}_{Lm}^{(*)}(x, t) E_{Fj}^{(-)}(x, t)\lambda(x) e^{i\omega_T t}$$

$$+ v_i^{(*)}(x, t) , \qquad (6.3)$$

$$\left(\frac{\partial}{\partial t} + v\frac{\partial}{\partial x}\right) E_{Fj}^{(*)}(x,t)$$
$$= -i\frac{\omega_s}{2\epsilon} \overline{\chi}_{mji}^* \mathcal{S}_{Lm}^{(*)}(x,t) u_{Fi}^{(*)}(x,t) \lambda(x) e^{i\omega_T t} , \qquad (6.4)$$

where

 $\omega_0 = \omega_0 + \nu$.

VII. REDUCED EQUATIONS OF MOTION

In practice, the dynamical behavior of the phonons is not observed directly, and it is therefore convenient to rewrite the pair of coupled

equations for the electric and phonon fields as a single equation for the dynamical behavior of the electric field alone. To that end we assume that the operator $\tilde{u}_{F}^{(+)}(x, t)$ is known at some initial time t = 0. We can then integrate Eq. (6.3) from t = 0 to find that

$$u_{Fi}^{(*)}(x,t) = u_{Fi}^{(*)}(x,0)e^{-(\gamma+i\overline{\omega}_0)t} - (i/2\rho\omega_0)\tilde{\chi}_{mji}^*$$
$$\times \int_0^t dt' \exp\left[-(\gamma+i\overline{\omega}_0)(t-t') + i\omega_T t'\right]$$

$$\times \lambda(x) \mathcal{S}_{Lm}^{(*)}(x, t') E_{Fj}^{(-)}(x, t')$$

$$+ \int_{0}^{t} dt' \ e^{-(\gamma + i\omega_{0})(t-t')} v_{i}^{(*)}(x, t') .$$
(7.1)

By using this expression for $\tilde{u}_{F}^{(*)}(x, t)$ in the equation of motion for the electric field [Eq. (6.4)] we bring that equation into a closed form; it becomes the linear inhomogeneous integro-differential equation

$$\begin{pmatrix} \frac{\partial}{\partial t} + v \frac{\partial}{\partial x} \end{pmatrix} E_{Fj}^{(*)}(x,t) = -\frac{i\omega_s}{2\epsilon} \overline{\chi}_{mjl}^* \lambda(x) \mathcal{S}_{Lm}^{(*)}(x,t) u_{Fl}^{(-)}(x,0) e^{-(\gamma - i\overline{\omega}_0 - i\omega_T)t} \\ - \frac{i\omega_s}{2\epsilon} \overline{\chi}_{mjl}^* \lambda(x) \mathcal{S}_{Lm}^{(*)}(x,t) \int_0^t dt' \exp[-(\gamma - i\overline{\omega}_0)(t-t') + i\omega_T t] v_l^{(*)}(x,t') \\ + \frac{\omega_s}{4\epsilon\rho\omega_0} \overline{\chi}_{mjl}^* \overline{\chi}_{nil} \lambda(x) \mathcal{S}_{Lm}^{(*)}(x,t) \int_0^t dt' \exp[-(\gamma - i\overline{\omega}_0 - i\omega_T)(t-t')] \mathcal{S}_{Ln}^{(-)}(x,t') E_{Fi}^{(*)}(x,t') .$$

$$(7.2)$$

Now, the fourth-rank tensor $\overline{\chi}_{mj1}^* \overline{\chi}_{ni1}$ is a nonlinear susceptibility which couples $\vec{\mathcal{G}}_L^{(+)}$, $\vec{\mathcal{G}}_L^{(-)}$, $\vec{E}_F^{(+)}$, and $\vec{E}_F^{(-)}$. In a homogeneous and isotropic medium, such a tensor can have only three independent components¹³:

$$\overline{\chi}_{mj1}^* \overline{\chi}_{ni1} = a \delta_{mn} \delta_{ji} + b \delta_{mj} \delta_{ni} + c \delta_{mi} \delta_{nj} .$$
 (7.3)

In that case, if the laser field is linearly polarized, the equations for the three components of $\mathbf{E}_{F}^{(*)}$ can be decoupled by properly orienting the y and z axes. When either of these axes is taken to lie in the direction of polarization, the equation of motion for $E_{Fx}^{(*)}$ contains no terms in $E_{Fy}^{(*)}$ or $E_{Fz}^{(*)}$, and the equations for the other components are similarly simplified. If the laser field is circularly or elliptically polarized, the situation is more complex, but it is always possible to find elliptical polarizations which again obey decoupled equations of motion.

Let us for simplicity, then, assume that the laser beam is linearly polarized in, say, the y direction, and that, further,

$$\vec{\mathcal{E}}_{L}^{(+)}(x,t) = \mathcal{E}_{L} \hat{\mathcal{Y}} e^{i(k_{L}x - \omega_{L}t)} .$$
(7.4)

With Eqs. (7.3) and (7.4), the equations of motion for the x and z components become identical, with the tensor $\overline{\chi}_{mjl}^* \chi_{nll}$ replaced by the coefficient a of Eq. (7.3), while in the equation for the y component the tensor is replaced by a+b+c.

Since the equations for the different components are identical in form, it will suffice in the sequel to consider only one component of the electric field, say the *j*th component. To simplify Eq. (7.2)for that component, let

$$\kappa^{2} = \sum_{i} \overline{\chi}^{*}_{jii} \, \overline{\chi}_{jji} \left| \mathcal{S}_{L} \right|^{2} (\omega_{s} / 4 \epsilon \rho \omega_{0}) , \qquad (7.5)$$

in which repeated indices are not summed except as indicated. The coefficient κ plays the role of a coupling constant in Eq. (7.2); its dimensions are (time)⁻¹. Further, let

$$\omega_s = \omega_L - \overline{\omega_0} = \omega_L - \omega_0 - \nu, \qquad (7.6)$$

$$\beta = \gamma + i(\omega_s - \omega_T); \qquad (7.7)$$

here note that ω_T equals 0 in the forward direction and increases until $\omega_T \cong \omega_s$ for Stokes light propagating at a right angle to the laser beam. The contributions from the initial states of the phonons and from the heat bath can be expressed concisely by defining the operators:

$$U(x) = -(i\omega_s/2\epsilon)\overline{\chi}^*_{yjl} \mathcal{E}^*_L e^{-ik_L x} u^{(+)}_{Fl}(x,0)\lambda(x), (7.8)$$
$$V(x,t) = -(i\omega_s/2\epsilon)\overline{\chi}^*_{yjl} \mathcal{E}^*_L$$
$$\times \exp[-ik_L x - i(\omega_L - \omega_T)t]$$
$$\times v^{(+)}_l(x,t)\lambda(x) \qquad (7.9)$$

(we are again using the summation convention). And, finally, we shall deal exclusively below with the single component $E_{Ff}^{(*)}(x,t)$, and we shall simplify the notation yet further, dropping all subscripts and superscripts:

$$E_{F_i}^{(*)}(x,t) \rightarrow E(x,t)$$
 (7.10)

With the abbreviations permitted by Eqs. (7.4)-(7.10), the equation of motion for the *j*th component of the positive-frequency part of the forward-propagating electric field becomes

$$\left(\frac{\partial}{\partial t} + v \frac{\partial}{\partial x}\right) E(x,t) = \kappa^2 \lambda(x) \int_0^t dt' e^{-\beta (t-t')} E(x,t') + U^{\dagger}(x) e^{-\beta t} + \int_0^t dt' V^{\dagger}(x,t') e^{-\beta (t-t')}.$$
(7.11)

Of the three terms on the right-hand side of Eq. (7.11), the last two represent the contributions of sources within the material; together they represent the dielectric polarization (with frequency near ω_s) that is induced in the amplifying medium by the laser beam. These terms are present whether the

Stokes light is amplified or not, and they can serve as sources of Stokes light even in the absence of any excitation in the phonon modes or the thermal reservoir due to the zero-point fluctuations of the fields u and v.

The first term on the right-hand side of Eq. (7, 11)describes the amplification of any Stokes light as it passes through the medium and is amplified by the radiation it stimulates. The source of the Stokes light may be either an external source (signal) or the spontaneous emission within the medium described by the other two terms of Eq. (7.11) (noise). The amplification process itself, like the more familiar ones of refraction and absorption, is not an inherently quantum-mechanical process, and can be described as well classically as quantum mechanically. On the other hand, it is precisely the quantum-mechanical properties of the noise terms which are the most interesting; the spontaneous emission effects which they describe are not present in any classical version of the theory.

By differentiating Eq. (7.11) with respect to time, we can reduce it to the simple differential equation

$$\begin{pmatrix} \frac{\partial}{\partial t} + \beta \end{pmatrix} \left(\frac{\partial}{\partial t} + v \frac{\partial}{\partial x} \right) E(x, t)$$

= $\kappa^2 E(x, t) \lambda(x) + V^{\dagger}(x, t) .$ (7.12)

To determine E(x, t) completely, Eq. (7.12) must, of course, be supplemented by appropriate initial conditions. The initial condition implied by Eq. (7.11) is

$$\left. \left(\frac{\partial}{\partial t} + v \frac{\partial}{\partial x} \right) E(x, t) \right|_{t=0} = U^{\dagger}(x) . \qquad (7.13)$$

The initial-value problem for the electric field is defined by specifying the field at t = 0:

$$E(x,t) \Big|_{t=0} = E_0(x), \tag{7.14}$$

and asking for the solution at later times. The electric field operator is completely determined for all times $t \ge 0$ by Eq. (7.11) with Eq. (7.14), or, equivalently by Eq. (7.12) with Eqs. (7.13) and (7.14).

VIII. INDEX OF REFRACTION

The amplification process itself, as we have argued, may be described as well classically as quantum mechanically. The distinctly quantum-mechanical aspect of the stimulated Raman effect, aside from the general quantum properties of the fields, lies in the production of noise. Once the noise has been generated, however, it, too, is amplified in an essentially classical manner. It is thus useful to investigate first the classical process by which any field is amplified, and to consider later the introduction of quantum-mechanical souces of noise.

If we remove the noise sources from the equations of motion Eqs. (7.12)-(7.14), they become

$$\left(\frac{\partial}{\partial t} + \beta\right) \left(\frac{\partial}{\partial t} + v \frac{\partial}{\partial x}\right) E(x, t) = \kappa^2 E(x, t) \lambda(x), \qquad (8.1)$$

$$E(x, 0) = E_0(x),$$
 (8.2)

$$\left(\frac{\partial}{\partial t} + v \frac{\partial}{\partial x}\right) E(x, t) \Big|_{t=0} = 0 .$$
 (8.3)

In an infinite medium we have $\lambda(x) = 1$ everywhere, and the differential equation [Eq. (8.1)] has an exponential solution

$$E(x, t) = E_0 e^{i(kx - \omega t)} , \qquad (8.4)$$

in which k and ω are related by

$$vk = \omega - i\kappa^2 / (\beta - i\omega) . \qquad (8.5)$$

The field of Eq. (8.4) is, with k real and ω complex, a solution to an initial-value problem with $E(x, 0) = E_0 e^{ikx}$. If, on the other hand, ω is taken to be real and k complex, the same field may be regarded as the response of the medium to a disturbance at x = 0 that oscillates with a frequency ω and produces there a field $E(0, t) = E_0 e^{-i\omega t}$.

Taking the latter point of view, we obtain a frequency-dependent complex index of refraction

$$n(\omega) = ck/\omega$$

= n'(\omega) + in''(\omega) . (8.6)

Let us define an effective index of refraction at the Stokes frequency as

$$n = c/v , \qquad (8.7)$$

so that we have

$$n(\omega) = n\left(1 - \frac{i\kappa^2}{\omega(\beta - i\omega)}\right)$$
.

For frequencies near ω_s we may approximate the slowly varying factor of $1/\omega$ by a constant¹⁴ so that

$$n(\omega) \cong n\left(1 - \frac{i\kappa^2}{\omega_s(\beta - i\omega)}\right).$$
 (8.8)

Thus, the real and imaginary parts of the index of refraction are (see Fig. 6)¹⁵

$$n'(\omega) = n \left(1 + \frac{\kappa^2 \Delta \omega}{\omega_s (\gamma^2 + \Delta \omega^2)} \right) \quad , \tag{8.9}$$

$$n''(\omega) = -n \frac{\kappa^2 \gamma}{\omega_s \left(\gamma^2 + \Delta \omega^2\right)} , \qquad (8.10)$$

where

$$\Delta \omega = \omega + \omega_T - \omega_s \quad . \tag{8.11}$$

Since the light is being amplified rather than absorbed, $n''(\omega)$ is negative. That it is negative for all frequencies, rather than just a narrow band, is chiefly due to the fact that we have neglected absorption of Stokes light as it passes through the medium.

It is well known that microcausality requires that



FIG. 6. Real and imaginary parts of the complex index of refraction of an amplifying medium. Symbols are as in text.

the real and imaginary parts of the index of refraction be Hilbert transforms of each other. Thus, the fact that $n''(\omega)$ is negative requires that the dispersion be anomalous $\{d[n(\omega)]/d\omega < 0\}$ for all frequencies except those close to the center of the line. Since the group velocity v_g is related to the dispersion by

$$\frac{c}{v_g} = n(\omega) + \omega \frac{dn(\omega)}{d\omega} ,$$

the group velocity can exceed c in those regions where the dispersion is anomalous. For the amplifying medium, this is in the wings rather than in the center of the line.

IX. SOLUTION FOR INITIAL PLANE WAVES

Using the solution of the equations of motion given by Eqs. (8.4) and (8.5), we may determine the behavior of the field if it has a known frequency ω or is a known superposition of such fields. However, a more common situation, particularly in quantummechanical contexts, is that in which the spatial behavior of the field is specified at a given time, as in Eqs. (8.2) and (8.3), and the behavior of the field at subsequent times is to be determined. Thus, we will need to know the frequency associated with a given wave number k.

Now, although the wave number k is a single-valued function of the frequency, given by Eq. (8.5):

$$vk = \omega + \kappa^2/(\omega + i\beta)$$

the frequency is a double-valued function of the wave number. The two solutions for ω as a function of k from Eq. (8.5) are

$$\omega_{\pm}(k) = \frac{1}{2} (vk - i\beta) \pm \frac{1}{2} (vk + i\beta) [1 - 4\kappa^2 / (vk + i\beta)^2]^{1/2} .$$
(9.1)

These frequencies can be characterized by the fact that

$$\operatorname{Im}\omega_{+}(k) > 0, \quad \operatorname{Im}\omega_{-}(k) < 0$$

The frequencies $\omega_*(k)$ and $\omega_-(k)$ are the frequencies of the two normal modes of oscillation that exist for each value of k. When the coupling between photons and phonons is weak, these normal modes are easily recognized to be photonlike and phonon-like in their behavior. Thus, when either (i) phase matching is poor, so that

$$|vk - \omega_s + \omega_T| \gg \kappa$$
,

or (ii) the coupling constant itself is small, so that

 $\kappa \ll \gamma$,

$$\left| 2\kappa/(vk-\omega_s+\omega_T+i\gamma) \right|^2 \ll 1$$

The radical of Eq. (9.1) may then be expanded to give

$$\omega_{\star}(k) \cong vk - \kappa^2 / [i\gamma + (vk - \omega_s + \omega_T)], \qquad (9.2)$$

$$\omega_{k}(k) \cong \omega_{s} - \omega_{T} - i\gamma + \kappa^{2} / [i\gamma + (vk - \omega_{s} + \omega_{T})] .$$
(9.3)

Clearly, the growing solution $\omega_*(k)$ is associated with the propagating photonlike mode, while the decaying solution is associated with the stationary phononlike mode.

In the case of strong coupling, i.e.,

 $\kappa \geq \gamma$, $|vk - \omega_s + \omega_T| \leq \kappa$,

no such identification is possible: The two original modes are thoroughly mixed by the coupling. In fact, for

$$|2\kappa/(vk-\omega_s+\omega_T+i\gamma)|\gg 1,$$

we have

$$\omega_{\pm}(k) \cong \frac{1}{2} (vk + \omega_s - \omega_T - i\gamma) \pm i\kappa , \qquad (9.4)$$

so that both normal modes partake equally of a photonlike and a phononlike character. This situation, however, would be difficult to obtain, as the observed values¹⁶ of Stokes gain $\kappa^2/\gamma v$ are on the order of 10^{-3} cm⁻¹ for laser-beam intensities of 1 MW/cm², while vibrational linewidths γ/v are on the order of several cm⁻¹. Thus, $\kappa^2/\gamma^2 \sim 10^{-3}$ or less for 1 MW/cm², and even for laser power levels of 100 MW/cm², we find $\kappa^2/\gamma^2 \sim 10^{-1}$.

Let us now assume that the material is permeated at t = 0 by a plane-wave electric field

$$E_k(x, 0) = e^{ikx} . (9.5)$$

The field $E_k(x, t)$ at arbitrary times is then a superposition of the two normal modes for that k:

$$E_{k}(x, t) = e^{-ikx} \left[A_{+}(k) e^{-i\omega_{+}(k)t} + A_{-}(k) e^{-i\omega_{-}(k)t} \right] ,$$
(9.6)

where, of course, from Eq. (9.5), we find

$$A_{+}(k) + A_{-}(k) = 1$$
 (9.7)

The other boundary condition on the (classical) electric field [Eq. (8.3)]

$$\left(\begin{array}{cc} \frac{\partial}{\partial t} + v & \frac{\partial}{\partial x} \end{array}\right) E(x, 0) = 0 \ ,$$

determines the ratio of the constants to be

$$\frac{A_{\star}(k)}{A_{\star}(k)} = -\frac{vk - \omega_{\star}(k)}{vk - \omega_{\star}(k)} .$$
 (9.8)

With the constants $A_{\pm}(k)$ determined by the initial conditions [Eqs. (9.7) and (9.8)], the electric field becomes

$$E_{k}(x,t) = e^{ikx} \left(\frac{\omega_{-}(k) - vk}{\omega_{-}(k) - \omega_{+}(k)} e^{-i\omega_{+}(k)t} + \frac{\omega_{+}(k) - vk}{\omega_{+}(k) - \omega_{-}(k)} e^{-i\omega_{-}(k)t} \right) .$$
(9.9)

In order to facilitate the interpretation of Eq. (9.9), let us again consider the case of weak coupling between photons and phonons. In that case, Eq. (9.9) can be written with Eqs. (9.2) and (9.3) as

$$E_{k}(x,t) \cong e^{ikx} \times \left(e^{-i\omega_{*}(k)t} + \kappa^{2} \frac{\left(e^{-i\omega_{*}(k)t} - e^{-i\omega_{*}(k)t}\right)}{\left(vk - \omega_{s} + \omega_{T} + i\gamma\right)^{2}} \right).$$
(9.10)

Since $\omega_*(k)$ is very nearly vk, the first term within the large parentheses simply represents the propagation of the initial field through the medium, with a small correction in the index of refraction due to the interaction with the phonons. The second term vanishes at t = 0, and represents an additional contribution to the field due to stimulated emission; its amplitude is large only when $k \cong (\omega_s - \omega_T)/v$, i.e., when phase matching prevails approximately.

Such a separation into contributions from the simple forward propagation of the initial field on the one hand, and from the amplification due to stimulated emission on the other can be made in general. The rate at which the electric field is amplified as it passes through the medium is given by the comoving derivative

$$\left(\frac{\partial}{\partial t} + v \frac{\partial}{\partial x}\right) E(x,t)$$

For $E_k(x, t)$, as given by Eq. (9.9), that rate is

$$\left(\frac{\partial}{\partial t} + v \frac{\partial}{\partial x}\right) E_{k}(x, t)$$
$$= \frac{i\kappa^{2}}{\omega_{*}(k) - \omega_{-}(k)} e^{ikx} (e^{-i\omega_{*}(k)t} - e^{-i\omega_{-}(k)t}), \quad (9.11)$$

where we have made use of the fact that

$$[\omega_{+}(k) - vk][\omega_{-}(k) - vk] = \kappa^{2} . \qquad (9.12)$$

The form of Eq. (9.11) may be interpreted in part through the following considerations. First, the two normal modes must contribute with equal magnitude and opposite sign, since the initial condition [Eq. (8.3)] requires that there be no amplification at t = 0. Second, the combination

$$\omega_{+}(k) - \omega_{-}(k) = \left[(vk - \omega_{s} + \omega_{T} + i\gamma)^{2} - 4\kappa^{2} \right]^{1/2} \quad (9.13)$$

has a zero near phase matching; thus, the factor $\kappa^2/[\omega_*(k) - \omega_*(k)]$ takes into account the enhancement of the coupling between photons and phonons that exists near phase matching.

The total electric field at x and t is the sum of the initially given field that has propagated to x, $E_k(x - vt, 0)$, and the further contribution from the radiation stimulated at all points between x and x - vt. That contribution is just the sum of all the contributions to each constant phase point, as given by Eq. (9.11). If

$$\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial x}\right)E_{k}(x,t)=\mathfrak{p}(x,t),$$

then, since the contribution made at the point x'must be made at t' = t - (x - x')/v to reach x at t, that sum is

$$\int_0^t dt' \, \mathfrak{p}(x-vt+vt',t')$$

Thus, the total field at x is

$$E_{k}(x,t) = E_{k}(x-vt,0) + \int_{0}^{t} dt' \, \mathfrak{p}(x-vt+vt',t') \; .$$

With $\mathfrak{p}(x, t)$ given by Eq. (9.11), this becomes

$$E_{k}(x, t) = e^{ik(x-vt)} \left(1 + \frac{i\kappa^{2}}{\omega_{+}(k) - \omega_{-}(k)} \int_{0}^{t} dt' e^{ivkt'} \times (e^{-i\omega_{-}(k)t'} - e^{-i\omega_{+}(k)t'}) \right).$$
(9.14)

It is worth noting that the terms of Eq. (9.14) are not completely analogous to those of Eq. (9.10): The first term of Eq. (9.14) represents a freely propagating wave without correction to the index of refraction, in contrast to that of Eq. (9.10).

X. SOLUTION FOR ARBITRARY INITIAL FIELDS

In order to find the electric field E(x, t) for general initial fields $E_0(x)$, we use a superposition of the solutions for initial plane waves. The Fourier component of $E_0(x)$ which varies as $\exp(ikx)$ becomes, at later times, the solution $E_k(x, t)$ of Eq. (9.14). Thus, the electric field which satisfies the initial conditions of

$$E(x, 0) = E_0(x)$$
, (8.2)

$$\left(\frac{\partial}{\partial t} + v \frac{\partial}{\partial x}\right) E(x, t) \Big|_{t=0} = 0, \qquad (8.3)$$

is

$$E(x, t) = \int (dk/2\pi) \left[\int dx' e^{-ikx'} E_0(x') \right] E_k(x, t)$$

= $\int dx' E_0(x') \int (dk/2\pi) E_k(x - x', t) .$ (10.1)

If, as we have assumed, the field E_0 contains only waves propagating in the x direction, and, indeed only Fourier components with wave numbers near $+k_s$, the solutions E_k for k < 0 make no contribution to Eq. (10.1). It is quite convenient to introduce a compact notation for the kernel in Eq. (10.1) that transfers the initial value of the field at x' to a later value at x at time t'. In a formal sense the kernel is just the field which evolves according to Eqs. (8.1)-(8.3) from an initial δ -function pulse. The field defined by

$$E_{\delta}(x,t) = \int (dk/2\pi) E_{k}(x,t) , \qquad (10.2)$$

in other words, satisfies the classical equation of motion, Eq. (8.1), together with the initial conditions

$$E_{\delta}(x, 0) = \delta(x),$$

$$\left(\frac{\partial}{\partial t} + v \frac{\partial}{\partial x}\right) E_{\delta}(x, t) \Big|_{t=0} = 0.$$
(10.3)

In order to write $E_{\delta}(x, t)$ in a more useful form, it is most convenient to start with Eq. (9.14) for $E_{k}(x, t)$. In that expression, the integrand can be written as a contour integral:

$$i\kappa^{2} \frac{e^{-i\omega_{-}(k)t'} - e^{-i\omega_{+}(k)t'}}{\omega_{+}(k) - \omega_{-}(k)} = \int_{c-i\infty}^{c+i\infty} \frac{ds}{2\pi i} \frac{\kappa^{2} e^{st'}}{[s + i\omega_{+}(k)][s + i\omega_{-}(k)]}, \qquad (10.4)$$

where the value of the real constant c is such as to place the contour to the right of both poles in the integrand. Since $\text{Im}\omega_{-}(k) < 0$, and since

$$\operatorname{Im}\omega_{*}(k) \leq \operatorname{Im}\omega_{*}(\omega_{s}/v) = -\frac{1}{2}\gamma + \frac{1}{2}(\kappa^{2} + \gamma^{2})^{1/2}$$
$$\leq \frac{1}{2}\kappa,$$

it will suffice if

 $C > \frac{1}{2} \kappa$.

The denominator of the integrand in Eq. (10.3) can also be written as

$$[s + i\omega_{*}(k)][s + i\omega_{-}(k)] = (s + \beta)(s + ivk) - \kappa^{2}.$$
(10.5)

Thus, using Eqs. (9.14), (10.4), and (10.5), we can write the response of the medium to an initial δ -function pulse as

$$E_{\delta}(x,t) = \delta(x-vt) + \kappa^{2} \int_{0}^{t} dt' \int \frac{dk}{2\pi}$$
$$\times \int_{c-i\infty}^{c+i\infty} \frac{ds}{2\pi i} \frac{\exp[ik(x-vt+vt')+st']}{(s+\beta)(s+ivk)-\kappa^{2}} \quad (10.6)$$

It is worth noting that the unmodified response of the medium, $\delta(x - vt)$, and, indeed, the entire solution (10.6), propagates in the +x direction, rather than showing the behavior symmetrical between the +x and -x directions which is physically to be expected when the initial field is a δ function. This spatial asymmetry has been introduced by our use of approximations which are accurate only for waves propagating in the forward direction, with wave numbers near $+k_s$. The solution is not therefore the actual physical field which follows from an initial δ function; it is simply a field which has the correct Fourier components in the neighborhood of $k = k_s$. That fact alone would make it a useful result, but as we shall see it is also useful in finding the Green's function for Eq. (7.11). If on the other hand we desire a symmetrical form for the field which ensues from an initial δ function, we can find it from Eq. (10.6) by restricting the integration to positive values of k and adding to that solution the terms which represent an identical solution for waves propagating in the negative direction.

The integral over wave numbers in Eq. (10.6) can now be performed as a contour integral in the complex plane. When x - vt + vt' < 0, the path of integration can be closed in the lower half-plane; since there are no singularities there, the integral vanishes. When x - vt + vt' > 0, the contour can be closed in the upper half-plane; there is a simple pole at

$$ivk = -s + \kappa^2/(s+\beta)$$
,

and there are no other singularities. Thus, with a change of variables from t' to t - t', we have

$$E_{\delta}(x, t) = \delta(x - vt) + \frac{\kappa^2}{v} \int_0^t dt' \int_{c - i^\infty}^{c + i^\infty} \frac{ds}{2\pi i} \frac{\eta(x - vt')}{s + \beta}$$
$$\times \exp\left(-s(x/v - t) - \frac{\kappa^2(x/v - t')}{s + \beta}\right) \quad ; \quad (10.7)$$

the function $\eta(x)$ is the Heaviside unit step function,

 $\eta(x) = 0$ for x < 0

= 1 for x > 0.

It may be worth noting that Eq. (10.7) can also be obtained by using Laplace-transform techniques to solve the equation of motion, Eq. (8.1), with the initial conditions of Eq. (10.3).

The remaining contour integral can be simplified by a change of variables from s to $s' = s + \beta$. The only singularity of the integrand is then at the origin, and a deformation of the path of integration transforms the integral into a well-known representation of a modified Bessel function¹⁷:

$$\eta(u)I_0(2(uv)^{1/2}) = \frac{1}{2\pi i} \int_{-\infty}^{(0_*)} \frac{ds'}{s'} e^{us' + v/s'} . \quad (10.8)$$

The path of integration extends from $-\infty$, around

the origin in the positive sense, and returns to $-\infty$. By using Eq. (10.8) we find that the response of the medium to a pulsed initial field can be written as

$$E_{\delta}(x, t) = \delta(x - vt) + (\kappa^{2}/v) \int_{0}^{t} dt' \eta(x - vt') \eta(vt - x) e^{-\beta(t - x/v)} I_{0} (2\kappa [(t - x/v)(x/v - t')]^{1/2})$$

$$= \delta(x - vt) + (\kappa^{2}/v) \eta(vt - x) \eta(x) e^{-\beta(t - x/v)} \int_{0}^{x/v} dt' I_{0} (2\kappa [t'(t - x/v)]^{1/2})$$

$$= \delta(x - vt) + (\kappa/v) \eta(vt - x) \eta(x) e^{-\beta(t - x/v)} [x/(vt - x)]^{1/2} I_{1} (2(\kappa/v) [x(vt - x)]^{1/2}).$$
(10.9)

Thus, each pulse that travels through the medium generates a delayed response field which propagates with a phase velocity v. The electric field $E_{\delta}e^{i\omega_T t}$ has, according to Eq. (7.7), a narrow spectrum of frequencies near ω_s as long as the coupling constant κ is not too large. The modulus of the delayed-response field is

$$f(x, t) = (\kappa/v) e^{-\gamma (t-x/v)} \times [x/(vt-x)]^{1/2} I_1(2(\kappa/v) [x(vt-x)]^{1/2}),$$

which extends from the current position of the pulse to its initial position. For small times $\kappa t \ll 1$, we find

$$f(x, t) \cong (\kappa^2/v^2) \cdot x \bar{e}^{\gamma(t-x/v)}$$

For large times the behavior is considerably more complicated. In Fig. 7, we show a few examples of the modulus of the delayed response field f(x, t) as a function of the dimensionless parameters

$$\tau = \kappa t, \quad \xi = \kappa x/v, \quad \alpha = \gamma/\kappa;$$

in terms of these variables the modulus is given by

$$f(x, t) = e^{-\alpha(\tau-\xi)} \left[\xi/(\tau-\xi) \right] I_1(2[\xi(\tau-\xi)]^{1/2})$$

Using Eq. (10.9), we can now write a general solution for the noise-free equation of motion [Eq. (8.1)] as

$$E(x, t) = \int dx' E_0(x') E_6(x - x', t). \qquad (10.10)$$

After changing variables of integration from x' to t'' = t - (x - x')/v, we obtain

$$E(x, t) = E_0(x - vt) + \kappa \int_0^t dt'' E_0(x - vt + vt'') e^{-\beta t''} [(t - t'')/t'']^{1/2} I_1(2\kappa [t''(t - t'')]^{1/2})$$

= $E_0(x - vt) + \kappa^2 \int_0^t dt' \int_0^{t - t'} dt'' E_0(x - vt + vt'') e^{-\beta t''} I_0(2\kappa (t't'')^{1/2}),$ (10.11)

where the two forms arise from the last two of the alternate expressions for $E_6(x, t)$ in Eq. (10.9).

It is to be emphasized that Eq. (10.11) describes the propagation of fields in an infinite and noisefree medium. The solution of Eq. (10.11) will require some modification before it describes the propagation of fields through a finite and noisy medium as determined by the equations of motion, Eqs. (7.12)-(7.14).

XI. GREEN'S FUNCTION FOR AMPLIFIED FIELD

The distinctly quantum-mechanical contributions to the equation of motion, represented by the terms in $U^{\dagger}(x)$ and $V^{\dagger}(x, t)$ in Eq. (7.15), can be considered as inhomogeneous additions to the homogeneous equation of motion

$$\left(\frac{\partial}{\partial t} + v \frac{\partial}{\partial x}\right) E(x, t) = \kappa^2 \int_0^t dt' E(x, t') e^{-\beta(t-t')}.$$
(11.1)

The general solution of Eq. (11.1) is, of course, given by Eq. (10.11). The inhomogeneous equation, Eq. (7.11), can be solved if a Green's func-

tion for Eq. (11.1) can be found.

It is easy to see that such a function is given by

$$G(x, t) = \eta(t) E_{\delta}(x, t).$$
 (11.2)

To show that it is a Green's function, let us first note that its derivatives satisfy

$$\left(\frac{\partial}{\partial t} + v \frac{\partial}{\partial x}\right) \mathfrak{g}(\mathbf{x}, t) = \delta(t) E_{\delta}(\mathbf{x}, t)$$
$$+ \eta(t) \left(\frac{\partial}{\partial t} + v \frac{\partial}{\partial x}\right) E_{\delta}(\mathbf{x}, t).$$

Since $E_{\delta}(x, t)$ satisfies Eq. (11.1) and since $\eta(t)$ vanishes for t < 0, we find that

$$\left(\frac{\partial}{\partial t} + v \frac{\partial}{\partial t}\right) \mathcal{G}(x, t) = \delta(t) E_{\delta}(x, t)$$
$$+ \kappa^2 \int_0^t dt' \mathcal{G}(x, t') e^{-\beta(t-t')}.$$

In order to simplify the first term on the righthand side of this equation, we make use of Eq. (10.9) for $E_6(x, t)$. Since

= 0,

the delayed response contributes nothing to the first term, and we find

$$\delta(t) E_{\delta}(x, t) = \delta(t) \delta(x - vt)$$
$$= \delta(t) \delta(x).$$

Thus, $\Im(x, t)$ as defined by Eq. (11.2) satisfies

$$\left(\frac{\partial}{\partial t} + v \frac{\partial}{\partial x}\right) \Im(x, t) = \delta(x) \delta(t)$$
$$+ \kappa^2 \int_0^t dt' \Im(x, t') e^{-\beta(t-t')}, \quad (11.3)$$

and can serve as a Green's function for Eq. (11.1).

The inhomogeneous counterpart of the equation of motion Eq. (11.1) takes the general form

$$\left(\frac{\partial}{\partial t} + v \frac{\partial}{\partial x}\right) E(x, t) = F(x, t)\eta(t) + \kappa^2 \int_0^t dt' E(x, t') e^{-\beta(t-t')}, \quad (11.4)$$

where we have included a factor of $\eta(t)$ in the inhomogeneous term to indicate explicitly that the interaction is only turned on at t = 0. In particular, the inhomogeneous term of Eq. (7.11) is

$$F(x, t) = U^{\dagger}(x)e^{-\beta t} + \int_0^t dt' V^{\dagger}(x, t')e^{-\beta(t-t')}.$$
 (11.5)

To solve the inhomogeneous equation it is convenient to find a particular solution of Eq. (11.4) which vanishes at t=0. We can do that by making

use of the Green's function given by Eq. (11.2). The particular solution is

$$E_{F}(x, t) = \int dx' \int dt' F(x', t') \eta(t') S(x - x', t - t')$$

= $\int dx' \int_{0}^{t} dt' F(x - x', t - t') E_{\delta}(x', t').$ (11.6)

Since $E_F(x, 0)$ vanishes, the initial conditions on the field can be met by adding to $E_F(x, t)$ a solution of the homogeneous equation which satisfies the initial conditions. The general solution of Eq. (11.4) for which

$$E(x, 0) = E_0(x)$$

thus is

$$E(x, t) = \int dx' E_0(x - x') E_0(x', t) + \int dx' \int_0^t dt' F(x - x', t - t') E_0(x', t'), \quad (11.7)$$

where $E_{\delta}(x, t)$ is given by Eq. (10.9).

Equation (11.7) can be simplified somewhat after the full expressions for $E_6(x, t)$ and F(x, t) are inserted by making use of the fact that

$$I_{0}(2\kappa(t_{1}t_{2})^{1/2}) = 1 + \kappa^{2} \int_{0}^{t_{1}} dt'_{1} \int_{0}^{t_{2}} dt'_{2} I_{0}(2\kappa(t'_{1}t'_{2})^{1/2})$$
$$= 1 + \kappa \int_{0}^{t_{1}} dt'_{1}(t_{2}/t'_{1})^{1/2} I_{1}(2\kappa(t'_{1}t_{2})^{1/2})$$
(11.8)

which can readily be proved by using the integral representation of Eq. (10.8) for the modified Bessel function. After inserting the expressions for $E_{\delta}(x, t)$ and F(x, t) from Eqs. (10.9) and (11.7), using Eq. (11.8), and simplifying, we obtain

$$E(x, t) = E_0(x - vt) + \int_0^t dt_1 F(x - vt + vt_1, t_1) + \kappa^2 \int_0^t dt' \int_0^{t-t'} dt'' e^{-\beta t''} I_0(2\kappa(t't'')^{1/2}) \\ \times [E_0(x - vt + vt'') + \int_0^{t-t'-t''} dt_1 F(x - vt + vt'' + vt_1, t_1)] \\ = E_0(x - vt) + \kappa^2 \int_0^t dt' \int_0^{t-t'} dt'' E_0(x - vt + vt'') I_0(2\kappa(t't'')^{1/2}) e^{-\beta t''} + \int_0^t dt' U^{\dagger}(x - vt') \\ \times I_0(2\kappa[t'(t - t')]^{1/2}) e^{-\beta(t-t')} + \int_0^t dt' \int_0^{t-t'} dt'' V^{\dagger}(x - vt', t - t' - t'') I_0(2\kappa(t't'')^{1/2}) e^{-\beta t''}.$$
(11.9)

This is the correct quantum field in an infinite noisy medium; it exhibits no steady-state behavior since the amplification process continues indefinitely as the field propagates further through the medium.

XII. SOLUTIONS FOR SEMI-INFINITE AND FINITE MEDIA

The infinite-medium solutions we have discussed above exhibit no steady-state behavior. Any particular point within the medium receives, as time goes on, the amplified field from more and more distant points, and the medium is stimulated to radiate ever increasing amounts. Even if the signal has a finite extent initially, the noise sources associated with the medium extend to arbitrarily large distances, and could thus provide for arbitrarily large fields at the point of interest. It is only when the medium has boundaries that a steady-state amplification process becomes possible.

The simplest such situation for forward-propagating waves occurs for a semi-infinite medium occupying the region x > 0. [The function $\lambda(x)$ of Eq. (3.4) thus becomes a Heaviside function $\eta(x)$]. The steady state arises in the following way. Let us suppose a plane wave of Stokes light illuminates the plane surface of the medium beginning at time t = 0. The wave need not be normally incident – we are still dealing with the field $\vec{E}(\vec{K}; x, t)$, defined by Eq. (3.5), which has a well-defined transverse wave vector \vec{K} – but the front of the wave must be parallel to the surface of the medium. For times t > 0



FIG. 7. Propagation of a δ -function pulse through the amplifying medium. The amplitude of the delayed response field, f(x, t), is given as a function of the dimensionless parameters $\tau = \kappa t$, $\xi = \kappa x/v$, and $\alpha = \gamma/\kappa$; solid curves $\alpha = 0$, dotted curves $\alpha = 2$, dashed curves $\alpha = 20$.

the wave penetrates the medium. That portion of the wave which has penetrated the medium most deeply, i.e., that nearest the front of the wave, will have undergone the greatest amplification; but as time increases that portion recedes to infinity. At finite distances within the medium, the field will have undergone only finite amplification. We shall show below that the field amplitude at any finite position is not only bounded but approaches a steady value if the initial field is steady.

When the amplifying medium takes the form of a slab of finite thickness extending from x = 0 to x = l, say, the field ceases to be amplified once it has left the medium. Within the medium the same arguments as we have used for the semi-infinite medium indicate that the field amplitude is bounded and may achieve a steady state. Since we are only dealing with forward-propagating waves [see Eqs. (5.12) and (5.13)], the boundary at x = l can have no effect on the field at positions with x < l. Beyond the boundary, the field is just what is radiated from the boundary and propagates freely with the speed v, so that for x > l

$$E(x, t) = E(l, t - (x - l)/v) .$$
(12.1)

For positions x < l, the field in the finite medium is precisely the same as the field at the position xwithin the semi-infinite medium. It will therefore suffice in the following to consider the results for the semi-infinite medium; the results for the finite medium can always be generated from them and Eq. (12.1).

Let us then consider the initial-value problem for a semi-infinite medium. We can construct a solution for the semi-infinite medium from that for the infinite medium since the introduction of the boundary involves essentially no new dynamical considerations. As we showed, in Sec. X, Eq. (10.10), the general initial value problem is solved once the behavior of an initial δ -function pulse is known. Since we shall need to consider initial conditions which specify the field inside as well as outside the medium, we must consider the behavior of δ -function pulses that originate in both regions. Within the medium, of course, the amplification of the field is unaffected by the boundary at x = 0; for x > 0 the field propagates as in infinite medium.

Let us consider first a δ -function pulse that is outside the medium at, say, x' < 0, at t = 0. It propagates freely until it reaches the boundary of the medium at t = -x'/v. From there it propagates precisely as a pulse that was at x = 0 at the "initial" time t = -x'/v in an infinite medium. A pulse which starts within the medium (x' > 0) is amplified as in an infinite medium from the instant it appears. Thus, the field due to an initial δ -function pulse at the position x' is

$$E_{\delta}(x'; x, t) = \eta(-x') \eta(-t - x'/v) \delta(x - x' - vt) + \eta(-x') \eta(t + x'/v) E_{\delta}(x, t + x'/v) + \eta(x') E_{\delta}(x - x', t) .$$
(12.2)

The first term of Eq. (12.2) represents the free propagation of the pulse outside the medium; the second term, the amplification within the medium of a pulse initially outside the medium; and the last term, the amplification of a pulse that starts within the medium.

Using Eq. (12.2), we can now write the solution of the homogeneous equations of motion Eqs. (8.1)-(8.3) for the semi-infinite medium as

$$E(x, t) = \int dx' E_0(x') E_{\delta}(x'; x, t) , \qquad (12.3)$$

which is the analog of Eq. (10.10) for the infinite medium. When we introduce the explicit expression Eq. (12.2) for E_b , rearrange the arguments of the Heaviside functions, and make a change of variables of integration from x' to t'' = t - (x - x')/v, Eq. (12.3) becomes

$$E(x, t) = E_0(x - vt) + \kappa^2 \int_0^t dt' \int_0^{t-t'} dt'' \eta(x - vt')$$

× $e^{-\beta t''} I_0 (\kappa(t't'')^{1/2}) E_0(x - vt + vt'')$. (12.4)

The factor of $\eta(x - vt')$ in Eq. (12.4) guarantees that $E(x, t) = E_0(x - vt)$ for x < 0. We thus have a solution for the initial-value problem for the homogeneous equations analogous to that of Eq. (10.11) for the infinite medium. It remains to find a particular solution to the inhomogeneous equations analogous to that of Eq. (11.6).

Since the only noise sources are associated with the amplifying medium, the particular solution of the inhomogeneous equations given by Eq. (11.6)

needs no change in structure. The fact that F(x, t)vanishes for x < 0 takes complete account of the fact that no amplification takes place for x < 0. Since we have

$$\eta(x) \overline{E}_{\mathfrak{s}}(x'; x, t) = \eta(x) E_{\mathfrak{s}}(x - x', t) ,$$

we can write the analog of Eq. (11.6) for the semiinfinite medium as

$$E_{F}(x, t) = \int dx' \int_{0}^{t} dt' F(x', t') \overline{E}_{6}(x'; x, t-t') .$$

The complete solution for the electric field analogous to Eq. (11.7) thus becomes

$$\begin{split} E(x, t) &= \int dx' E_0(x') \overline{E}_6(x'; x, t) \\ &+ \int dx' \int_0^t dt' F(x', t') \overline{E}_6(x'; x, t-t') \; . \end{split}$$

Using Eqs. (12.2) and (10.9) for \overline{E}_5 , we obtain

$$E(x, t) = E_0(x - vt) + \int_0^t dt_1 F(x - vt + vt_1, t_1) + \kappa^2 \int_0^t dt' \int_0^{t-t'} dt'' \eta(x - vt') e^{-\beta t''} \times I_0(2\kappa(t't'')^{1/2}) [E_0(x - vt + vt'') + \int_0^{t-t'-t''} dt_1 F(x - vt + vt'' + vt_1, t_1)] (12.5)$$

as the complete solution for the inhomogeneous equations of motion for the semi-infinite medium.

It may be noted that other methods of solving Eq. (7.11) for a semi-infinite medium, viz., by using Laplace-transform techniques on Eq. (7.11) directly or by transforming Eq. (7.11) to a pure integral equation and finding a power-series solution by iteration, lead directly to Eq. (12.5). The powerseries solution, can, in fact, be summed in closed form for the finite medium as well as for the semiinfinite medium.¹⁸

The fact that the semi-infinite medium has a boundary makes it possible to consider problems in which time varying fields are incident from the outside. Let us consider, for example, a semi-infinite medium illuminated by a steady monochromatic beam of light which is switched on at t = 0. We assume that the frequency of the beam is not far from the Stokes frequency. As we have mentioned earlier, we are dealing with beams having a transverse wave vector \vec{K} , so that the beam is a plane wave which need not be propagating normally to the surface of the medium. The assumption that the field is switched on at t = 0, however, corresponds to the assumption that the wave front is parallel to the surface of the medium. The illumination can be achieved by filling the half-space outside the medium with an electric field such that

$$E_0(x) = E_0 \eta(-x) e^{ikx};$$

the frequency associated with the longitudinal propa-

gation of the light is $\omega = vk$; the total frequency of the electric field is $\omega + \omega_T$. The field E(x, t) then propagates in the +x direction and penetrates the medium for t > 0. An alternative way of generating the incident field is to place a plane source at x = 0which oscillates steadily with a frequency $\omega = kv$ beginning at t = 0, so that the inhomogeneous term of Eq. (12.5) becomes

$$F(x, t) = E_0 \delta(x) e^{-ikvt} \eta(t) .$$

Both methods of generating the incident field lead to identical responses within the medium. If we neglect the contribution from the noise sources in Eq. (12.5), the time-dependent field becomes

$$E(x, t) = E_0 \eta(vt - x) e^{ik(x - vt)}$$

$$\times [1 + \kappa^2 \int_0^{x/v} dt' \int_0^{t - x/v} dt'' I_0 (2\kappa (t't'')^{1/2}) e^{-(\beta - ikv)t''}]$$
(12. 6)

within the medium, i.e., for x > 0. The Bessel function can be integrated over t', as in Eq. (11.8), so that

$$E(x, t) = E_0 \eta(vt - x) e^{ik(x-vt)} \left[1 + \kappa \int_0^{t-x/v} dt'' \times (x/vt'')^{1/2} I_1(2\kappa(t''x/v)^{1/2}) e^{-(\beta - ikv)t''}\right]. \quad (12.7)$$

This field shows a complex transient behavior, but assumes a simple steady-state value for large times. As $t \rightarrow \infty$ and for finite x, the integral of Eq. (12.6) becomes a familiar Laplace transform,¹⁹ and we have • / 、

$$E(x, t) \cong E_0 \exp\left(ik(x-vt) + \frac{\kappa^2 x/v}{\beta - ikv}\right).$$
(12.8)

As is to be expected, once the transient effects of the passage of the initial wave front have died out, the field assumes the same form as in the infinite medium, i.e., the exponential solution given by Eq. (8.4) with the complex propagation constant k given by Eq. (8.5).

In order to illustrate the transient behavior of the solution given in Eq. (12.7) we have evaluated the required integral numerically for several cases. In Fig. 8 we have plotted the behavior of the normalized field amplitude $|E(x, t)|/E_0$ as a function of the distance parameter $\xi = \kappa x/v$ for several values of the parameters $\tau = \kappa t$, $\alpha = \gamma/\kappa$, and $\delta = (\omega_s - kv)/\kappa$.

In spite of the formal similarity, however, the physical contexts of Eq. (12.8) and Eqs. (8.4) and (8.5) are quite different.²⁰ The latter, in the classical context of Sec. VIII, represent the normal modes of the infinite amplifying medium; for real values of the frequency these are spatially growing. Equations (8.4) and (8.5) may also, of course, be interpreted as representing the growth in time of the amplitude of a spatially homogeneous plane wave; it is this approach we followed in our sub-

iku) t''

sequent analysis. This latter approach is the more natural one in a quantum-mechanical context where dynamical problems must usually be regarded as initial-value problems (real k) rather than time-dependent boundary-value problems (real ω). Although the solution to the boundaryvalue problem for a steady incident wave achieves a steady state for all finite x even in the infinite medium, the solution of the initial-value problem for a spatially homogeneous plane wave does so only in a finite or semi-infinite medium.

Sommerfeld and Brillouin have treated the response of a dispersive medium to a semi-infinite plane wave in classic papers.²¹ Since their model involves a rather more complex form of dispersion than our Eq. (8.8), their results differ in several



FIG. 8. Propagation of a semi-infinite plane wave through the amplifying medium. The normalized amplitude $|E|/E_0$ is given as a function of the dimensionless parameters $\tau = \kappa t$, $\xi = \kappa x/v$, $\alpha = \gamma/\kappa$, $\delta = (\omega_s - \kappa v)/\kappa$. Graph (a) shows the envelope of the field amplitude medium; the curves are drawn for $\delta = 0$, for weak damping, $\alpha = 1$ (solid curves), and for strong damping, $\alpha = 10$ (dashed curves). The vertical steps at $\xi = \tau$, which are common to both sets of curves, represent the onset of the field. The field is seen to reach its asymptotic value more rapidly for strong damping. In (b) the field amplitude is shown for $\alpha = 1$ and several values of the frequency mismatch δ at the fixed time $\tau = 5$.



FIG. 9. Region of integration for the electric field of Eq. (13.1), t > x/v.

details from those presented here. However, Sommerfeld and Brillouin also find that an initial wave propagates through the medium with a speed unaffected by the interaction of the field with the medium. The initial wave is followed by a transition field, also given in terms of integrals of Bessel functions. Ultimately the field assumes the steady-state behavior determined by the index of refraction in a manner analogous to our Eq. (12.8).

XIII. ASYMPTOTIC BEHAVIOR OF THE AMPLIFIED FIELD

Let us now consider the behavior of the homogeneous terms of the amplified field, given by Eq. (12.4), for more general values of the initial field. In the operation of the medium as an amplifier, these terms represent the amplified signal, while the inhomogeneous terms $E_F(x, t)$ represent noise.

For times sufficiently large that the field that was initially within the medium has propagated past the point of observation, i.e., for t > x/v, the amplified signal at the point x is, from Eq. (12.4), for arbitrary initial fields

$$E(x, t) = E_0(x - vt) + \kappa^2 \int_0^{x/v} dt' \int_0^{t-t'} dt'' e^{-\beta t''} \times I_0(2\kappa(t't'')^{1/2}) E_0(x - vt + vt'') .$$
(13.1)

The region of integration is shown in Fig. 9.

In many cases the initial field $E_0(x)$ has a reasonably well-defined wavelength, and can thus be expressed as a slowly varying amplitude multiplying a rapidly oscillating factor e^{ikx} . Now, the dominant time dependence of the kernel of the integral in Eq. (13.1) is given by the exponential factor

$$e^{-\beta t''} = e^{-\gamma t''} - i(\omega_s - \omega_T)t''$$

Since, in general, the phonon damping ν is large compared to the coupling κ , the major contributions to the double integral come for $t'' < 1/\gamma$. If the amplitude does not vary appreciably over that time, it may be factored out of the integral. The remaining integrals are precisely the same as in Eq. (12.6) and lead to the asymptotic result of Eq.

(12.8). Thus, if the signal amplitude varies slowly over distances of the order of v/γ , the signal is amplified as if it were monochromatic; asymptot-ically it approaches a form, similar to that of Eq. (12.8), but with the factor

$$\exp\left(ik(x-vt)+\frac{\kappa^2 x/v}{\beta-ikv}\right)$$

multiplied by the slowly varying amplitude.

For more general values of the initial field, the fact that $\gamma \gg \kappa$ still ensures that the major contributions to the integral come from small values of t''. Thus, for large times the upper limit of integration may be taken to be infinite, and the amplified signal approaches the asymptotic form

$$E_{\infty}(x, t) = E_{0}(x - vt) + \kappa^{2} \int_{0}^{x/v} dt' \int_{0}^{\infty} dt'' e^{-\beta t''} \times I_{0}(2\kappa(t't'')^{1/2}) E_{0}(x - vt + vt'') . \quad (13.2)$$

In order to examine this asymptotic form in somewhat more detail, let us expand the initial field in terms of normal modes, as in Eq. (4.1). To simplify the notation, let

$$A_{k} = \sum_{\lambda} i \left[\hbar \omega(\vec{k}) A / 2L \epsilon \right]^{1/2} e_{j}^{(\lambda)}(k) a_{k\lambda}(0) , \qquad (13.3)$$

and let $\sum_{k}^{\prime\prime}$ denote sums over positive values of k only. The initial field then becomes

$$E_0(x) = \sum_{k}^{\prime\prime} A_k e^{ikx} . (13.4)$$

Thus, from Eqs. (13.1) and (13.2), we obtain

$$E(x, t) = \sum_{k}^{\prime\prime} A_{k} e^{ik(x-vt)} [1 + \kappa^{2} \int_{0}^{x/v} dt' \\ \times \int_{0}^{t-t'} dt'' e^{-(\beta - ikv)t''} I_{0}(2\kappa(t't'')^{1/2})], \quad (13.5)$$

$$E_{\infty}(x,t) = \sum_{k}' A_{k} \exp[ik(x-vt)] \exp \frac{\kappa^{2} x/v}{\beta - ikv} , \quad (13.6)$$

where we have again used a known Laplace transform¹⁹ to simplify the asymptotic result.

For large times, it can be shown (see the Appendix) that a negligible contribution is added to the double integral of Eq. (13.5) by increasing the limit of the t'' integration to t. Thus, for $\gamma t \gg 1$, we have

$$E(x, t) \cong \sum_{k}^{\prime\prime} A_{k} e^{ik(x-vt)} \left[1 + \kappa^{2} \int_{0}^{x/v} dt' \right] \\ \times \int_{0}^{t} dt'' e^{-(\beta - ikx)t''} I_{0}(2\kappa(t't'')^{1/2}) \left].$$

The fields E(x, t) and $E_{\infty}(x, t)$ can then easily be written as similar inverse Laplace transforms:

$$E(x,t) \cong \sum_{k} {'' A_{k} e^{ikx}} \int_{c-i\infty}^{c+i\infty} \frac{ds}{2\pi i} \frac{e^{st}}{s+ikv} \exp\left(\frac{\kappa^{2} x/v}{\beta+s}\right) , \qquad (13.7)$$

$$E_{\infty}(x, t) = \sum_{k}'' A_{k} e^{ikx} \int_{c-i\infty}^{c+i\infty} \frac{ds}{2\pi i} \frac{e^{st}}{s+ikv} \exp\left(\frac{\kappa^{2} x/v}{\beta - ikv}\right) .$$
(13.8)

Since the major contribution to the contour integrals comes in the vicinity of the pole at s = -ikv, it is clear that there is only a small difference between the coefficients of A_k in Eqs. (13.7) and (13.8). The expression for $E_{\infty}(x, t)$ thus furnishes a good description of measurements involving the strength or amplitude of the field at large times. It should be noted, however, that the asymptotic approximation does make some sacrifice with respect to the commutation properties of the field. The correct value for the field commutator can only be obtained from the full expression for the field, Eq. (12.5). The contributions added to the correct expression for the homogeneous part of the field, Eq. (13.1), in order to obtain the asymptotic expression Eq. (13.8) lead to a divergence of the field commutator; they do not, however, lead to significant errors in the description of properties of the amplified field that have classical or semiclassical analogs.

The intensity of the field is a particular case of the first-order correlation function²²

$$G^{(1)}(x_1, t_1; x_2 t_2) = \langle E^{\dagger}(x_1, t_1) E(x_2, t_2) \rangle ; \qquad (13.9)$$

it is just the particular value $G^{(1)}(x, t; x, t)$. For stationary fields the power spectrum is, from the Wiener-Khinchine theorem, the Fourier transform of $G^{(1)}$. For the steady-state amplified signal, the correlation function becomes asymptotically

$$G_{\infty}^{(1)}(x_1, t_1; x_2, t_2) = \langle E_{\infty}^{\dagger}(x_1, t_1) E_{\infty}(x_2, t_2) \rangle . \quad (13.10)$$

We shall, for the moment, consider only the contributions to these correlation functions from the homogeneous parts of the field, Eqs. (13.1) and (13.2), and shall defer to Sec. XIV consideration of the contributions to $G^{(1)}$ and $G^{(1)}_{\infty}$ due to the noise sources.

If the field is initially, i.e., at t=0, in a stationary state for which

$$\langle a_{k\lambda}^{\dagger}(0), a_{k'\lambda'}(0) \rangle = n_{k\lambda} \delta_{kk'} \delta_{\lambda\lambda'}, \qquad (13.11)$$

then the steady-state correlation function becomes

$$G^{(1)}_{\infty}(x,t+\tau;x,t)$$

$$= \sum_{k\lambda} \frac{\hbar \omega(\bar{k})A}{2 \epsilon L} |e_j^{\lambda}(k)|^2 n_{k\lambda} \exp\left(ikv \tau + \frac{2\kappa^2 \gamma x/v}{\gamma^2 + \Delta \omega^2}\right),$$

where

....

$$\Delta \omega = \omega_s - \omega_T - kv \; .$$

Since the wave vectors are extremely dense, the sum can be well approximated by an integral. If we further let

$$N_{0}(\omega_{T}+kv) = \sum_{\lambda} \left| e_{j}^{\lambda}(k) \right|^{2} A n_{k\lambda} / 4\pi \epsilon v , \qquad (13.12)$$

so that $\hbar\omega N_0(\omega)$ is the power spectrum of the field

(13.13)

present initially, then we have

$$G_{\infty}^{(1)}(x, t+\tau; x, t) = \int d\omega \, \hbar \omega N_0(\omega) \exp\left(i\omega \tau + \frac{2\kappa^2 \gamma x/\nu}{\gamma^2 + \Delta \omega^2}\right) \,,$$

where we have written

 $\omega = \omega_T + kv, \quad \Delta \omega = \omega_s - \omega \ .$

The power spectrum of the amplified signal is of the form $\hbar\omega N(\omega)$, where

$$N(\omega) = N_0(\omega) \exp\left(\frac{2\kappa^2 \gamma x/\nu}{\gamma^2 + \Delta \omega^2}\right) .$$
(13.14)

Thus, we see that each Fourier component is amplified independently of the others; the steady-state gain constant of the amplitude is just given by the imaginary part of the index of refraction $n''(\omega)$ of Eq. (8.10).

XIV. QUANTUM NOISE

Although classical theory would allow, in principle, the possibility of constructing a noise-free amplifier, that is not true for the quantum-mechanical case. The fluctuations necessarily associated with the phonon damping always provide, in the presence of the incident laser field, a source of spontaneously emitted Stokes light; these contributions are also amplified, and appear as noise in the output of the medium.

The contribution of the noise sources is, from Eqs. (11.5) and (12.5), for the semi-infinite medium, ²³

$$E_{F}(x, t) = \int_{0}^{t} dt' \eta(x - vt') e^{-\beta(t-t')} \\ \times I_{0}(2\kappa [t'(t - t')]^{1/2}) U^{\dagger}(x - vt') \\ + \int_{0}^{t} dt \int_{0}^{t-t'} dt'' \eta(x - vt') e^{-\beta t''} \\ \times I_{0}(2\kappa (t't'')^{1/2}) V^{\dagger}(x - vt', t - t' - t'') ;$$
(14. 1)

we have used Eq. (11.8) to simplify this expression, and have explicitly included factors of $\eta(x - vt')$ to emphasize the cutoff of the integration due to the boundary of the medium at x = 0. After any excitations initially present within the medium have propagated past the point x, that is for t > x/v, the field becomes

$$E_{F}(x, t) = \int_{0}^{x'v} dt' \ e^{-\beta(t-t')} I_{0} (2\kappa [t'(t-t')]^{1/2}) U^{\dagger}(x-vt) + \int_{0}^{x'v} dt' \ \int_{0}^{t-t'} dt'' \ e^{-\beta t''} I_{0} (2\kappa (t't'')^{1/2}) \times V^{\dagger}(x-vt', t-t'-t'') \ .$$
(14.2)

This field must be added to the solution of the homogeneous (classical) equations given by, say, Eq. (13.1).

In order to compute the contribution due to noise to measured quantities such as the correlation functions, we note that the fields U, V, and E_0 all refer to different dynamical systems. Since there is in general no phase coherence between the initial states (the Heisenberg-picture states) of the noise sources and the electric field, the intensities contributed by the signal and the noise sources are additive. More generally, the corresponding correlation functions are additive as well, so that

$$G^{(1)}(x_1, t_1; x_2, t_2) = G^{(1)}_s(x_1, t_1; x_2, t_2)$$

 $+ \, G_{s}^{(1)}(x_1,t_1;x_2,t_2) \ . \label{eq:Gs}$ Here $G_{s}^{(1)}$ is the signal correlation function

$$G_{s}^{(1)}(x_{1}, t_{1}; x_{2}, t_{2}) = \langle E^{\dagger}(x_{1}, t_{1}) E(x_{2}, t_{2}) \rangle$$

with E(x, t) the homogeneous solution of, say Eq. (13. 1); and $G_N^{(1)}$ is the noise correlation function

$$G_N^{(1)}(x_1, t_1; x_2, t_2) = \langle E_F^{\dagger}(x_1, t_1) E_F(x_2, t_2) \rangle$$

The intensity of the noise is the particular value of the correlation function

$$\mathcal{J}_{N}(x, t) = G_{N}^{(1)}(x, t; x, t) \quad . \tag{14.3}$$

For t > x/v, the intensity becomes

$$\begin{aligned} \mathfrak{S}_{N}(x,t) &= e^{-2\gamma t} \int_{0}^{x/\nu} dt_{1}' \int_{0}^{x/\nu} dt_{2}' \exp[\gamma(t_{1}'+t_{2}')-i(\omega_{s}-\omega_{T})(t_{1}'-t_{2}')] I_{0} \left(2\kappa[t_{1}'(t-t_{1}')]^{1/2}\right) \\ &\times I_{0} \left(2\kappa[t_{2}'(t-t_{2}')]^{1/2}\right) \langle U(x-\nu t_{1}') U^{\dagger}(x-\nu t_{2}') \rangle \\ &+ \int_{0}^{x/\nu} dt_{1}' \int_{0}^{x/\nu} dt_{2}' \int_{0}^{t-t_{1}'} dt_{1}'' \int_{0}^{t-t_{2}'} dt_{2}'' \exp[\gamma(t_{1}''+t_{2}')-i(\omega_{s}-\omega_{T})(t_{1}''-t_{2}'')] I_{0} \left(2\kappa(t_{1}'t_{1}'')^{1/2}\right) \\ &\times I_{0} \left(2\kappa(t_{2}'t_{2}'')^{1/2}\right) \langle V(x-\nu t_{1}',t-t_{1}'-t_{1}'') V^{\dagger}(x-\nu t_{2}',t-t_{2}'-t_{2}'') \rangle \end{aligned}$$
(14.4)

Like the amplified signal, the noise exhibits a complicated transient behavior after the laser beam is turned on at t = 0. In actual practice, however, the laser beam is presumably turned on some time before the signal to be amplified reaches the medium. The noise output would then have already reached its steady-state value when its contribution is of interest.

Even for fairly small times t, the contribution to the noise due to the initial phonon excitation becomes negligible since the phonons initially present decay rapidly. In the Appendix, we present a more detailed argument that the first term of Eq. (14.4) is dominated by the factor $e^{-2\gamma t}$ and can be neglected for large times. Thus, only the radiation transferred from the thermal reservoir through the phonon modes contributes to the steady-state noise.

In order to determine the contribution of the thermal reservoir to $\mathscr{I}_{N}(x, t)$, it is necessary to compute the expectation value

 $\langle V(x_1, t_1) V^{\dagger}(x_2, t_2) \rangle$.

In case the reservoir is at a temperature of absolute zero, so that

$$\langle V^{\dagger}(x_1, t_1) V(x_2, t_2) \rangle = 0$$

we have

$$\langle V(x_1, t_1) V^{\dagger}(x_2, t_2) \rangle = \langle [V(x_1, t_1), V^{\dagger}(x_2, t_2)] \rangle$$
.

Using the definition of V in terms of $v^{(*)}$ [Eq. (7.9)] and the commutator given by Eq. (6.2), we have

$$\langle V(x_1, t_1) V^{\dagger}(x_2, t_2) \rangle = \hbar \omega_s \kappa^2 v N \delta(x_1 - x_2) \delta(t_1 - t_2) ,$$
(14.5)

where

$$\kappa^2 v N = (\omega_s / 4 \epsilon^2 A) \Gamma |\mathcal{E}_L|^2 \sum_i \overline{\chi}_{yji} \overline{\chi}_{yji} ;$$

using the definition of κ^2 [Eq. (7.5)] we see that N has a magnitude given by

 $N \simeq \rho A \Gamma \omega_0 / \epsilon v$,

and represents an effective volume density of phonons in the reservoir. In case the system is not at a temperature of absolute zero, each of the Fourier components of the δ function, $\delta(t_1 - t_2)$, of Eq. (14.5) must be multiplied by the appropriate factor for the thermal excitation of that mode, ²⁴ 1 + $n_{\rm th}(\omega)$. Since, however, we are chiefly interested in the frequency components of the correlation function near the Stokes frequency, the thermal correction can be assumed constant. Thus, the antinormal correlation function $\langle V(x_1 t_1)V^{\dagger}(x_2 t_2) \rangle$ assumes, to a good approximation, the form of Eq. (14.5), but with

$$N \cong (\rho \Gamma \omega_0 A / \epsilon v) [1 + n_{\rm th}(\omega_*)]$$

For times sufficiently large that the contribution of the initial phonon excitation can be neglected, the noise intensity becomes

$$\mathfrak{s}_{N}(x, t) \cong \hbar \omega_{s} \kappa^{2} N \int_{0}^{x/v} dt' \int_{0}^{t-t'} dt'' e^{-2\gamma t''} I_{0}^{2} (2\kappa (t't'')^{1/2})$$
(14.6)

For very large times, the noise intensity thus approaches the steady-state value²⁵

$$\begin{split} \mathfrak{s}_{N}(x, \ \infty) &= \hbar \omega_{s} \kappa^{2} N \int_{0}^{x/\nu} dt' \int_{0}^{\infty} dt'' \ e^{-2\gamma t''} I_{0}^{2} \left(2\kappa (t't'')^{1/2} \right) \\ &= \hbar \omega_{s} \frac{\kappa^{2}}{2\gamma} N \int_{0}^{x/\nu} dt' \ e^{\kappa^{2} t' / \gamma} I_{0}(\kappa^{2} t' / \gamma) \\ &= \frac{1}{2} \hbar \omega_{s} N \frac{\kappa^{2} x}{\gamma v} \ e^{\kappa^{2} x / \gamma v} \left[I_{0} \left(\kappa^{2} x / \gamma v \right) - I_{1}(\kappa^{2} x / \gamma v) \right] . \end{split}$$

$$(14.7)$$

This result represents the intensity radiated from the medium due to the presence within it of a uniformly distributed source of broad-band incoherent noise.

When the medium is functioning as an amplifier, the noise intensity σ_N must be added to the signal intensity given by the homogeneous contribution to $G^{(1)}(x, t; x, t)$. If the signal varies sufficiently slowly that the output is always in a steady state, then the intensity of the amplified signal is given by Eq. (13.13) to be

$$\boldsymbol{g}_{\boldsymbol{s}}(\boldsymbol{x}, \ \infty) = G_{\boldsymbol{\infty}}^{(1)} \ (\boldsymbol{x}, \ t; \ \boldsymbol{x}, \ t)$$

$$= \int d\omega \, \hbar \omega N_0(\omega) \exp\left(\frac{2\kappa^2 \gamma x/\nu}{\gamma^2 + \Delta \omega^2}\right) \,. \tag{14.8}$$

For a signal at the Stokes frequency, with

$$N_0(\omega) = N_s \delta(\omega - \omega_s)$$

the amplified signal intensity is

$$\boldsymbol{s}_{\boldsymbol{s}}(\boldsymbol{x}, \ \boldsymbol{\infty}) = \hbar \boldsymbol{\omega}_{\boldsymbol{s}} N_{\boldsymbol{s}} e^{2\kappa^2 \boldsymbol{x} / \boldsymbol{\gamma} \boldsymbol{v}} \ . \tag{14.9}$$

The signal-to-noise ratio for the amplifier thus is

$$\frac{\mathbf{g}_{\mathbf{g}}(x,\infty)}{\mathbf{g}_{N}(x,\infty)} = 2\frac{N_{\mathbf{g}}}{N} \frac{\gamma v}{\kappa^{2} x} e^{\kappa^{2} x/\gamma v} \left[I_{0}\left(\frac{\kappa^{2} x}{\gamma v}\right) - I_{1}\left(\frac{\kappa^{2} x}{\gamma v}\right) \right]^{-1}.$$
(14.10)

For small values of the gain parameter $\kappa^2 x/\gamma v$, this ratio is a decreasing function of the gain. However, for $\kappa^2 x/\gamma v$ greater than roughly 0.77, the signal-to-noise ratio increases with increasing gain (see Fig. 10).

For signals that are not precisely at the Stokes frequency, the amplification drops off rapidly, and the signal-to-noise ratio decreases appreciably. If the unamplified signal is monochromatic with frequency $\omega_s + \Delta \omega$, so that its spectrum is given by

$$N_{0}(\omega) = N_{s} \delta(\omega - \omega_{s} - \Delta \omega) ,$$

then the signal-to-noise ratio is

$$\frac{g_{s}(x,\infty)}{g_{N}(x,\infty)} = 2\frac{N_{s}}{N}\frac{\gamma v}{\kappa^{2}x}\exp\left[\frac{\kappa^{2}x}{\gamma v}\left(\frac{\gamma^{2}-\Delta\omega^{2}}{\gamma^{2}+\Delta\omega^{2}}\right)\right] \times \left[I_{0}\left(\frac{\kappa^{2}x}{\gamma v}\right) - I_{1}\left(\frac{\kappa^{2}x}{\gamma v}\right)\right]^{-1}.$$
 (14.11)

Clearly, for $|\Delta\omega| > \gamma$, the signal-to-noise ratio is a decreasing function of the gain parameter $\kappa^2 x/\gamma v$ for all values of the parameter. Thus, although the medium amplifies signals of all frequencies to some extent, it does not usefully amplify signals outside the bandwidth of the spontaneous Stokes radiation.

The power spectrum of the noise output of the amplifier is the Fourier transform of the steadystate correlation function

$$G_N^{(1)}(x,\infty,\tau) \equiv \lim G_N^{(1)}(x,t+\tau;x,t) \quad \text{as } t \to \infty;$$

with Eq. (14.5) this correlation function becomes



FIG. 10. Signal intensity *S*, noise intensity *N*, and signal-to-noise ratio S/N for a signal precisely at the Stokes frequency $\omega = \omega_s$ as a function of the dimensionless gain parameter $y = \kappa^2 x/\gamma v$. Vertical scale is arbitrary.

$$G_{N}^{(1)}(x, \infty, \tau) = \hbar \omega_{s} \kappa^{2} N \exp[-\gamma |\tau| + i(\omega_{s} - \omega_{T})\tau]$$

$$\times \int_{0}^{x/\nu} dt' \int_{0}^{\infty} dt'' e^{-2\gamma t''}$$

$$\times I_{0}(2\kappa (t't'')^{1/2}) I_{0}(2\kappa [t'(t'' + |\tau|)]^{1/2}) .$$
(14.12)

The observed correlation function also includes a factor of $e^{i\omega_T\tau}$ due to the transverse propagation of the field. For negligible amplification $(\kappa^2 x/\gamma v \simeq 0)$, the observed noise correlation function is

$$G_N^{(1)}(x,\infty,\tau)e^{i\omega_T\tau} = \frac{1}{2}\hbar\omega_s N(\kappa^2 x/\gamma v)e^{-\gamma|\tau|+i\omega_s\tau}.$$

The corresponding spectrum is Lorentzian in character with center at ω_s and half-width γ . For appreciable amplifications, the double integral of Eq. (14.12) is an increasing function of $|\tau|$. The line is thus no longer precisely Lorentzian in shape, and its half-width may be shown to be less than γ . The line narrowing arises from the fact that noise at the Stokes frequency is more strongly amplified as it propagates through the medium than noise at other frequencies.

XV. QUASIPROBABILITY DISTRIBUTION FOR ELECTRIC FIELD

In order to compute the expectation values of more general functions of the electric field than those we have discussed, it is convenient to introduce a quasiprobability distribution for the electric field strength so that the quantum-mechanical averages can be represented as integrals over the distribution of field strengths.²⁶

The expectation value of a normally ordered operator function²⁷ – or functional – of the electric field, $\{\Im(E(x, t))\}_N$, can be expressed as

$$\langle \{ \mathfrak{F}(E(x,t)) \}_N \rangle$$

= $\int d^2 \mathcal{S} \mathfrak{F}_N(\mathcal{S}) \langle \{ \delta^{(2)}(E(x,t) - \mathcal{S}) \}_N \rangle .$

Here $d^2 \mathcal{S} = d(\operatorname{Re} \mathcal{S})d(\operatorname{Im} \mathcal{S})$ is the real element of area in the complex \mathcal{S} plane; $\delta^{(2)}(z) = \delta(\operatorname{Re} z)\delta(\operatorname{Im} z)$ is the two-dimensional δ function, and $\{\delta^{(2)}(E)\}_N$ is the normally ordered δ function. The function $\mathfrak{F}_N(\mathcal{S})$ is obtained by writing the *c* numbers \mathcal{S}^* and \mathcal{S} for $E^{\dagger}(x, t)$ and E(x, t), respectively, in the normally ordered expression $\{\mathfrak{F}(E(x, t))\}_N$. If we define a quasiprobability distribution

$$W(\mathcal{E}; x, t) = \langle \{ \delta^{(2)}(E(x, t) - \mathcal{E}) \}_N \rangle$$
(15.1)

for normally ordered operators, then

$$\langle \{ \mathfrak{F}(E(x,t)) \}_N \rangle = \int d^2 \mathcal{E} \mathfrak{F}_N(\mathcal{E}) W(\mathcal{E};x,t) . \qquad (15.2)$$

The operator $\{\delta^{(2)}(E(x,t)-\mathcal{E})\}_N$ is, of course, highly singular, ²⁸ so that its expectation value $W(\mathcal{E}; x, t)$ may be difficult to define for some states of the field; we shall see below, however, that it is a well-behaved and familiar function for the usual states of the amplified field.

The Fourier transform of the quasiprobability distribution of Eq. (15.1) is called the normally ordered characteristic function. We shall write it, using complex notation, as

$$\chi_N(\Lambda; x, t) = \int d^2 \mathcal{E} \, e^{\Lambda \mathcal{E}^* - \Lambda^* \mathcal{E}} \, W(\mathcal{E}; x, t) \, . \tag{15.3}$$

With this notation, the Fourier representation of the $\boldsymbol{\delta}$ function is

$$\pi^2 \delta^{(2)}(\Lambda) = \int d^2 \mathcal{S} e^{\Lambda \mathcal{S}^* - \Lambda^* \mathcal{S}} . \qquad (15.4)$$

The operator analog of Eq. (15.4) for the normally ordered δ function is

$$\{\delta^{(2)}(E(x,t)-\mathcal{E})\}_{N} = (1/\pi 2) \int d^{2}\Lambda \exp\{[E^{\dagger}(x,t)-\mathcal{E}^{*}]\Lambda\}$$
$$\times \exp\{[E(x,t)-\mathcal{E}]\Lambda^{*}\}, \quad (15,5)$$

so that the characteristic function may also be written as

$$\chi_N(\Lambda; x, t) = \langle e^{E^{\dagger}(x, t)\Lambda} e^{-E(x, t)\Lambda^*} \rangle .$$
 (15.6)

To compute a distribution function for \mathcal{E} , let us suppose, for example, that initially the phonon system and the heat bath are at a temperature of absolute zero, i.e., totally unexcited, and that the field is in a coherent state:

$$U(x) | \mathcal{E}_{0}, 0, 0 \rangle = 0 ,$$

$$V(x) | \mathcal{E}_{0}, 0, 0 \rangle = 0 ,$$

$$E_{0}(x) | \mathcal{E}_{0}, 0, 0 \rangle = \mathcal{E}_{0}(x) | \mathcal{E}_{0}, 0, 0 \rangle ,$$

(15.7)

where $|\mathcal{E}_0, 0, 0\rangle$ is the state of the entire system.

If we let

be the amplified classical signal, then we find

$$\chi_{N}(\Lambda; x, t) = \exp[\mathcal{E}^{*}(x, t)\Lambda - \mathcal{E}(x, t)\Lambda^{*}] \\ \times \langle \exp[E^{*}_{F}(x, t)\Lambda] \exp[-E_{F}(x, t)\Lambda^{*}] \rangle ,$$
(15.9)

where $E_F(x, t)$ is defined by Eq. (14.1).

To simplify Eq. (15.9), we note that the commutators

 $[U^{\dagger}(x), U(x')],$

given by Eqs. (4.8) and (7.8), and

 $[V^{\dagger}(x), V(x')]$

given by Eqs. (6.2) and (7.9) are both c numbers. Thus, the commutator of $E_F(x, t)$ with its adjoint is also a c number, and we may use the identity

$$e^{A}e^{B} = e^{B}e^{A}e^{[A,B]}$$

if

$$[[A, B], A] = [[A, B], B] = 0,$$

to write

$$\exp[\Lambda E_F^{\dagger}(x,t)]\exp[-\Lambda^* E_F(x,t)]$$

$$= \exp[-\Lambda^* E_F(x,t)] \exp[\Lambda E_F^{\dagger}(x,t)]$$

$$\times \exp\left\{-\left|\Lambda\right|^{2}\left[E_{F}^{\dagger}(x,t),E_{F}(x,t)\right]\right\}.$$

Furthermore, since

 $E_F(x,t) | \mathcal{E}_0, 0, 0 \rangle = 0$,

the expectation values of antinormally ordered functions of $E_F(x, t)$ vanish. Thus, we may write the characteristic function as

 $\langle \exp[\Lambda E_F^{\dagger}(x,t)] \exp[-\Lambda^* E_F(x,t)] \rangle$

$$= \exp\{-|\Lambda|^{2}[E_{F}^{\dagger}(x,t), E_{F}(x,t)]\}, \qquad (15.10)$$

and the commutator as

$$\begin{bmatrix} E_F^{\dagger}(x,t), E_F(x,t) \end{bmatrix} = \langle E_F^{\dagger}(x,t), E_F(x,t) \rangle$$
$$= \mathfrak{g}_N(x,t) . \tag{15.11}$$

$$\chi_{N}(\Lambda; x, t) = \exp[\mathcal{E}^{*}(x, t)\Lambda - \mathcal{E}(x, t)\Lambda^{*} - \mathcal{I}_{N}(x, t)|\Lambda|^{2}],$$
(15.12)

whose Fourier transform is, from Eq. (15.4),

$$W(\mathcal{S}; x, t) = (1/\pi^2) \int d^2 \Lambda \chi_N(\Lambda; x, t) e^{\Lambda^* \mathcal{S} - \Lambda \mathcal{S}^*}$$
$$= \frac{1}{\pi \mathfrak{s}_N(x, t)} \exp\left(-\frac{|\mathcal{S} - \mathcal{S}(x, t)|^2}{\mathfrak{s}_N(x, t)}\right) \cdot (15.13)$$

Thus, for purposes of computing normally ordered expectation values, the (positive-frequency) electric field at x and t has a Gaussian distribution about the "classical" amplified signal, with a variance just equal to the noise intensity.

Of course, the quasiprobability distribution of field values at any point changes with time. For large times, the distribution approaches the steadystate value

$$W_{\infty}(\mathcal{E}; x, t) = \frac{1}{\pi g_N(x, \infty)} \exp\left(-\frac{|\mathcal{E} - \mathcal{E}_{\infty}(x, t)|^2}{g_N(x, \infty)}\right),$$
(15.14)

where $g_N(x, \infty)$ is given by Eq. (14.9), and $\mathcal{E}_{\infty}(x, t)$ is given by the *c*-number equivalent of Eqs. (13.2) or (13.6):

$$\mathcal{S}_{\infty}(x,t) = \mathcal{S}_{0}(x-vt) + \kappa^{2} \int_{0}^{x/b} dt' \int_{0}^{\infty} dt'' e^{-\beta t''}$$
$$\times I_{0}(2\kappa(t't'')^{1/2}) \mathcal{S}_{0}(x-vt+vt'')$$
$$= \sum_{k}' a_{k} \exp\left(ik(x-vt) + \frac{\kappa^{2}x/v}{\beta-ikv}\right). (15.15)$$

Thus, the distribution $W_{\infty}(\mathcal{E}; x, t)$ is, for fixed x, a Gaussian distribution with a constant standard deviation whose center follows the oscillatory behavior of $\mathcal{E}_{\infty}(x, t)$.

In particular, if the signal has a well-defined frequency, $\mathscr{E}_{\infty}(x, t)$ varies only in phase as t increases, not in amplitude, and the Gaussian hump of $W_{\infty}(\mathscr{E};$ x, t) circles the origin of the \mathscr{E} plane at a constant radius of $|\mathscr{E}_{\infty}(x, t)|$ and with an angular frequency equal to the signal frequency.

In case the Stokes frequency field is initially not in a coherent state, it is still often true that the density operator for the field can be written as a superposition of coherent states in terms of a Prepresentation, ²⁶ and that the density operator for the system can be written in terms of a superposition of the states defined by Eqs. (15.7). The distribution function for the field is then given by the corresponding superposition of the distribution functions for the coherent field, Eq. (15.13) or (15.14).

The *P* representation is generally given in terms of the eigenvalues $\alpha_{k\lambda}$ of the mode operators $a_{k\lambda}(0)$. The initial density operator of the system is thus written as

$$\rho = \int P(\{\alpha\}) | \mathcal{E}_0(x), 0, 0\rangle \langle \mathcal{E}_0(x), 0, 0 | \prod_{k} d^2 \alpha_{k\lambda},$$

where the states are functions of all $\alpha_{_{k\lambda}}$ via

$$\mathcal{E}_0(x) = \sum_{k\lambda} \left[\hbar \omega(\mathbf{k}) A / 2 \epsilon L \right]^{1/2} e_j^{\lambda}(k) \alpha_{k\lambda} e^{ikx}.$$

The quasiprobability distribution

$$\overline{W}(\mathcal{E}; x, t) = \operatorname{tr}[\rho\{\delta^2(E(x, t) - \mathcal{E})\}_N]$$

is then

$$\overline{W}(\mathcal{E}; x, t) = \int P(\{\alpha\}) W(\mathcal{E}; x, t) \prod_{k} d^2 \alpha_{k}, \quad (15.16)$$

where $W(\mathcal{E}; x, t)$ is given by Eq. (15.13) and is a function of all the $\alpha_{k\lambda}$ through the amplified signal of Eq. (15.8).

APPENDIX

We wish to evaluate two integrals for large times. The first, from Eq. (13.5), is

$$K_1 = \kappa^2 \int_0^{x/\nu} dt' \int_{t-t}^t e^{-(\beta - ik\nu)t''} I_0(2\kappa(t't'')^{1/2});$$

the region of integration is the small triangular region shown in Fig. 8. The second, from Eq. (14.4), is

$$\begin{split} K_2 &= \left(\hbar \, \omega_s \, A/2\epsilon \right) \kappa^2 (1+n_{\rm th}) \, \int_0^{\chi/\nu} \, dt' \, e^{-2\gamma (t-t')} \\ &\times I_0^2 \left(2\kappa [t'(t-t')]^{1/2} \right) \, , \end{split}$$

where we have made use of the fact that, from Eqs. (4.8) and (7.8),

$$\langle U(x)U^{\dagger}(x')\rangle \cong (\hbar \omega_{s} A/2\epsilon)\kappa^{2}(1+n_{th})\delta(x-x'),$$

where $n_{\rm th}$ is the number of thermal phonons in each mode.

Using the mean-value theorem to evaluate the integrals, we have

$$K_1 = \kappa^2 \frac{1}{2} (x/v)^2 e^{-\beta(t-\zeta x/v)} I_0(2\kappa [(\mu x/v)(t-\zeta x/v)]^{1/2}) ,$$

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 1 W. H. Louisell, Coupled Mode and Parametric Electronics (Wiley, New York, 1961).

²J. A. Giordmaine, in *Quantum Optics*, edited by R. J. Glauber (Academic, New York, 1969), p. 493 ff.

 3 For a general review of the stimulated Raman effect and an extensive bibliography, see N. Bloembergen, Am. J. Phys. <u>35</u>, 989 (1967).

 4 A discussion of the parametric Brillouin effect for classical fields with results for transient response of a finite medium has been given by N. M. Kroll, J. Appl. Phys. <u>36</u>, 34 (1965). Kroll's results are consistent with ours in the classical limit.

⁵A Yariv and W. H. Louisell, IEEE J. Quant. Electron. <u>QE2</u>, 418 (1966); B. R. Mollow and R. J. Glauber, Phys. Rev. <u>160</u>, 1076 (1967); <u>160</u>, 1097 (1967); B. R. Mollow, *ibid.* <u>162</u>, 1256 (1967).

⁶W. H. Louisell, Radiation and Noise in Quantum Electronics (McGraw-Hill, New York, 1964), p. 274 ff. ⁷J. A. Armstrong *et al.*, Phys. Rev. <u>127</u>, 1918 (1962); P. A. Silberg, *ibid.* <u>153</u>, 342 (1967).

 8 J. Tucker and D. Walls, Phys. Rev. <u>178</u>, 2036 (1969), describe in quantum-mechanical terms the propagation of wave packets in an infinite medium functioning as a frequency converter; their techniques may also be used for a semiclassical amplifying medium.

 3 R. Graham and H. Haken, Z. Physik <u>213</u>, 420 (1968) have obtained a similar equation of motion for the propagation of light through a medium of pumped twolevel atoms. Their treatment, however, concentrates on the nonlinear aspects introduced by the saturation of the atomic transition; only their results for the linear

$$K_2 = (\hbar \omega_s A/2\epsilon) \kappa^2 (1 + n_{\rm th}) (x/v) e^{-2\gamma (t - \zeta x/v)}$$

$$\times I_0^2 (2\kappa [(\zeta x/v)(t-\zeta x/v)]^{1/2}),$$

where ζ and μ are dimensionless numbers between 0 and 1 and are, of course, functions of x and t. We obtain upper bounds for these integrals by taking $\zeta = \mu = 1$, so that

$$\begin{split} |K_1| &\leq \frac{1}{2} \kappa^2 (x/v)^2 \, e^{-\gamma (t-x/v)} I_0 (2\kappa \, [x(vt-x)/v^2]^{1/2}), \\ K_2 &\leq (\hbar \, \omega_s \, A/2\epsilon) \kappa^2 (1+n_{\rm th}) (x/v) e^{-2\gamma (t-x/v)} \\ &\times I_0^2 (2\kappa \, [x(vt-x)/v^2]^{1/2}). \end{split}$$

For very large times, we make use of the asymptotic expression for the modified Bessel function

$$I_0(z) \sim [1/(2\pi z)^{1/2}] e^z$$
,

and obtain, with
$$vt - x \approx vt$$
,
 $|K_1|^2 \lesssim \frac{1}{4} (\kappa x/v)^4 e^{-2\gamma t} (v/4\pi\kappa) (xt)^{-1/2} e^{2\kappa (xt/v)^{1/2}}$
 $K_2 \lesssim (\hbar \omega_s A/2\epsilon) \kappa^2 (1+n_{\rm th}) e^{-2\gamma t}$
 $\times (x/4\pi\kappa) (xt)^{-1/2} e^{4\kappa (xt/v)^{1/2}}$.

Thus, as $t \to \infty$ for fixed x, both $|K_1|^2$ and K_2 are bounded by

 $(\operatorname{const}/\sqrt{t})\exp\left[-2\gamma t+4\kappa(xt/v)^{1/2}\right],$

which becomes negligibly small for large times.

¹¹The subscripts F and B do not refer to forward- and backward-propagating phonons; each of the phonon fields \tilde{u}_F and \tilde{u}_B is still a superposition of standing waves.

¹²More detailed discussions of the damped oscillator can be found, for example, in I. R. Senitzky, Phys. Rev. <u>119</u>, 670 (1960); R. Graham and H. Haken, Z. Physik <u>210</u>, 276 (1968); W. H. Louisell and J. H. Marburger, IEEE J. Quantum Electron. <u>QE3</u>, 348 (1967); W. H. Louisell, Ref. 6, p. 255 ff; R. J. Glauber, in Ref. 2, p. 32 ff.

¹³L. D. Landau and E. M. Lifshitz, *Electrodynamics* of Continuous Media (Pergamon, Oxford, 1960), p. 381.
 ¹⁴The approximation also has the effect of suppressing a

spurious pole at $\omega = 0$; the pole is in fact a result of our earlier approximation $\omega(\vec{k}) \cong \omega_s$ in Eq. (4.16).

¹⁵The Lorentzian shape of the anti-Stokes line has a physical basis similar to that of the Stokes line (see, e. g., J. Ducuing, in Ref. 2, p. 455 ff.) and has been observed by P. D. Maker and R. W. Terhune, Phys. Rev. <u>137</u>, A801 (1965).

¹⁶N. Bloembergen, Ref. 10, p. 151, and Ref. 3, p. 1007.

¹⁷W. Magnus, F. Oberhettinger, and R. P. Soni, Formulas and Theorems for the Special Functions of Mathematical Physics, 3rd ed. (Springer, New York, 1966), p. 83.

¹⁸T. von Foerster, thesis, Harvard University, 1968 (unpublished).

regime may be compared with ours.

¹⁰G. Placzeck, in *Handbuch der Radiologie*, Vol. VI/2, (Akademische Verlagsgesellschaft, Leipzig, 1934), p. 205 ff; N. Bloembergen, *Nonlinear Optics*, (Benjamin, New York, 1965), p. 102 ff.

¹⁹Handbook of Mathematical Functions, edited by M. Abramowitz and I. A. Stegun (Dover, New York, 1965), p. 1026, formula 29.3.81 or p. 487, formula 11.4.31.

²⁰C. Y. She, Phys. Rev. <u>176</u>, 461 (1968) has considered traveling wave problems by means of a symmetrical treatment of space and time variables.

²¹A. Sommerfeld, Optics, (Academic, New York, 1964), pp. 97 ff., 114 ff.; L. Brillouin, Wave Propagation and Group Velocity (Academic, New York, 1964).

²²R. J. Glauber, Phys. Rev. <u>130</u>, 2529 (1963).

²³The subscript F here serves to distinguish the inhomogeneous contribution to the field, and should not be confused with that used to distinguish the forward-propagating part of the field in Sec. VI.

²⁴See, for example, M. Lax, Phys. Rev. <u>145</u>, 110 (1960).

²⁵The integrations can be performed with formulas 11.4.7. (p. 485), 29.3.81 (p. 1026), 9.6.16 (p. 376), and 11.3.12 (p. 483) of Ref. 19.

²⁶R. J. Glauber, in Quantum Optics and Electronics, edited by C. De Witt et al. (Gordon and Breach, New York, 1965), p. 151 ff; R. J. Glauber, in Fundamental Problems in Statistical Mechanics, Vol. II, edited by E. G. D. Cohen (North-Holland, Amsterdam, 1968); K. E. Cahill and R. J. Glauber, Phys. Rev. 177, 1882 (1969).

²⁷That is, an operator function in which all creation operators E^{\dagger} are written to the left of any destruction operators E.

²⁸K. E. Cahill and R. J. Glauber, Phys. Rev. <u>177</u>, 1857 (1969).