PHYSICAL REVIEW B VOLUME 12, NUMBER 10 15 NOVEMBER 1975

Enhanced stimulated Raman scattering and general three-boson parametric instabilities*

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(Received 16 September 1974)

A recent theory of stimulated Raman scattering explained a Stokes-intensity-enhanced gain that had been observed but that was not predicted by earlier theories. It is shown that in the earlier golden-rule analyses (perturbation-theory treatment of occupation numbers n) the enhancement was lost by neglecting the increase of the vibrational amplitudes above their thermal-equilibrium values. Even though the probability of an individual ion or molecule being excited is small, the occupation number of the phonon in the Raman process is large. In the previous mode-amplitude analyses, the enhancement was lost in the method of linearizing the nonlinear differential equations. By solving these same mode-amplitude equations for the n's without using the previous linearization scheme, the enhancement is obtained and the equivalence of the mode-amplitude and golden-rule boson-occupation-number results is demonstrated explicitly. The analysis shows explicitly that the loss of phase information in using the boson occupation numbers does not cause the loss of enhancement. The results are applicable to other three-boson splitting processes that are important in ferromagnetism, phonon interactions, plasma instabilities, and device physics.

I. INTRODUCTION

Stimulated Raman scattering was first observed[1] and analyzed[2,3] in 1962. It was recently realized[4] that a parametric instability in the Raman Stokes process causes a Stokes-intensity gain enhancement that explains a number of observed anomalies including a nearly discontinuous increase, or "jump," in the Stokes intensity I_S as a function of the laser intensity I_L in the absence of self-focusing and feedback. The purpose of the present investigation is to resolve the discrepancy between early theories[2,3] that did not give the gain enhancement and the later theory[4] that did. In addition to identifying the assumptions in both types of the early theories that led to the loss of enhancement, the equivalence of the results of the occupation-number (n) rate-equation analysis and the mode-amplitude (a and $a^\dagger$) analysis is demonstrated, and it is shown that the loss of phase information in the occupation-number analysis does not affect the enhancement results. The results are of interest in the general three-boson splitting problem, which arises in a number of fields of physics, as discussed below.

The characteristic feature of a parametric instability is that as the amplitude n_0 of some mode 0 increases, the amplitude n_k of a mode k that is coupled to 0 first increases slowly, then increases rapidly to a great value as n_0 approaches a critical value. For example, in the Raman process a laser photon is annihilated, a Stokes photon is created, and a fundamental (reststrahl) phonon is created. As the laser-photon occupation number n_L approaches a critical value n_R, the occupation numbers n_f and n_S of the fundamental-phonon and Stokes-photon modes become very large. This increase in the value of n_S is the gain enhancement and jump already mentioned.

Any three-boson splitting process is potentially unstable parametrically. There are analogies between the instability in the Raman process and previously studied instabilities in ferromagnetic resonance[5-7] (premature saturation of the main resonance, subsidiary absorption below the main resonance, and parallel-pumping absorption), plasma physics,[8] and electronic devices.[9]

The physical interpretation of these instabilities is rather simple. The balance of energy put into the f phonons by the Raman process against that removed from the f phonons by relaxation is a key to the explanation. The power out by relaxation (by interaction with impurities or other phonons, for example) increases linearly with the number of phonons n_f, which is just the condition that a relaxation time exists. On the other hand, the power into the f phonons increases nonlinearly with increasing n_f since the Raman process is a three-boson process (which results in products of boson occupation numbers in the expression for the power). Thus, at a critical value of the laser intensity, the amplitude n_f becomes very large.

Previous analyses of stimulated Raman scattering and other parametric processes used either the equations of motion of the boson occupation numbers obtained from perturbation theory (the golden rule) or the equations of motion of the mode amplitudes (creation and annihilation operators or Fourier components and their complex conjugates of the electric field, for example). In the case of stimulated Raman scattering, the previous results from the mode-amplitude analysis do not give the enhancement obtained by the recent occupation-number analysis.

In the present paper the relation between the two approaches is demonstrated explicitly, and the points in the previous analyses at which the enhance-

ment was lost are identified. In the previous golden-rule-type analyses, the enhancement was lost by neglecting the deviation of the vibrational amplitude from the thermal-equilibrium value. Even though n_f becomes large, the probability of an individual ion or molecule being excited is small, roughly speaking. Specifically, $n_f/N \ll 1$ is usually satisfied, where N is the number of unit cells or molecules. It was this fact that the individual ions or molecules are not highly excited that led to the assumption that the thermal-equilibrium values were maintained in the previous analyses. There are similar results for other three-boson processes. For example, in ferromagnetism, magnon occupation numbers are large at the threshold, while the probability of an individual electron spin being in the reversed-spin state is small.

In the previous mode-amplitude analyses, the enhancement was lost in the method of linearizing and decoupling the nonlinear differential equations for the mode amplitudes a_L, a_S, a_f, and their complex conjugates (or Hermitian conjugates in the quantum-mechanical solution). It is shown specifically that reducing the nonlinear equations to parametric linear equations (that is, linear equations with time-dependent coefficients) by assuming that the laser-field amplitude $a_L = b_L e^{-i\omega_L t}$, where b_L *is a constant*, results in the loss of the enhancement. The same linearization scheme applied to well-known magnon or phonon parametric instabilities results in the loss of the steady-state solution, even though damping is included and a steady-state solution is expected on the basis of simple physical arguments. By solving *the same mode-amplitude equations* for the n's without using this linearization scheme, the difficulties are removed and the equivalence of the mode-amplitude and golden-rule results is demonstrated explicitly for the stimulated-Raman-scattering and magnon problems.

The central features of enhanced stimulated Raman scattering are classical. In the classical analyses, care has been exercised to keep the order of the a and $a^\dagger$ correct so that the equations can easily be converted to quantum equations in Sec. IV. The present analysis is concerned only with the steady-state solution.

In Sec. IV several points that are not apposite to the central results, but are of general interest, are discussed. These include a ferromagnetic instability, purely quantum-mechanical effects, resolution of a difficulty in the quantum treatment when dissipation is included, and the problem of phases mentioned above. Important results are indicated by underscored equation numbers.

II. MODE-AMPLITUDE ANALYSIS OF ENHANCED STIMULATED RAMAN SCATTERING

In this section the same mode-amplitude equations for stimulated Raman scattering that were previously solved by a common linearization approximation to obtain gain without enhancement are solved without making this linearization approximation. The present solution gives the gain enhancement and agrees with the results of the golden-rule analysis, as discussed in Sec. III.

The mode-amplitude equations have been obtained classically from Maxwell's equations with terms added to account for the coupling of the electromagnetic and elastic waves.[3,10,11] Specifically, an interaction Lagrangian was added to the sum of the electromagnetic and elastic Lagrangians and the field-amplitude equations were obtained from the Lagrangian. The resulting second-order partial differential equations were reduced by standard methods to the following first-order partial differential equations[10,11]:

$$\frac{\partial a_f}{\partial t} = -i\omega_f a_f - V a_L a_S^\dagger - \tfrac{1}{2}\Gamma a_f \,, \tag{2.1}$$

$$\frac{\partial a_S}{\partial t} = -i\omega_S a_S - V a_L a_f^\dagger - c_S \frac{\partial a_S}{\partial x} \,, \tag{2.2}$$

$$\frac{\partial a_L}{\partial t} = -i\omega_L a_L + V^* a_f a_S - c_L \frac{\partial a_L}{\partial x} \,, \tag{2.3}$$

where the a's are the Fourier components of the fundamental phonon field f, the laser field L, and the Stokes field S, the $a^\dagger$'s are the complex conjugates of the a's, V is the coupling coefficient for the coupling of the three fields, and V^* is the complex conjugate of V. Equations (2.1)–(2.3) and the complete treatment of Secs. II and III are classical. If (2.1)–(2.3) are to be interpreted as quantum-mechanical operator equations, a noise source must be added to preserve the commutation relations since damping has been added in these equations. The quantum treatment will be considered in Sec. IV.

The previous solutions were obtained by linearizing these equations by assuming that $a_L = a_{L0} e^{-i\omega t}$, where a_{L0} is a constant, as discussed in Sec. I. There are several possible ways of solving these equations without using this linearization method, as discussed in Sec. IV. The simplest method, which also best illustrates the relation to the golden-rule results, is to convert Eqs. (2.1)–(2.3) to rate equations for $n_f \equiv a_f^\dagger a_f$, $n_L = a_L^\dagger a_L$, and $n_S = a_S^\dagger a_S$, which will be solved for the steady-state solution. This is easily accomplished by using

$$\frac{\partial n_f}{\partial t} = \frac{a_f^\dagger \partial a_f}{dt} + \text{c.c.}$$

and similar equations for n_S and n_L, which give

$$\frac{\partial n_f}{\partial t} = F - \Gamma n_f \,, \tag{2.4}$$

$$\frac{\partial n_S}{\partial t} = F - c_S \frac{\partial n_S}{\partial x} \,, \tag{2.5}$$

$$\frac{\partial n_L}{\partial t} = -F - c_L \frac{\partial n_L}{\partial x} \ , \tag{2.6}$$

$$\frac{\partial F}{\partial t} = 2|V|^2[n_L(n_f+n_S) - n_S n_f] - \tfrac{1}{2}\Gamma F \ , \tag{2.7}$$

$$F \equiv -V a_L a_f^\dagger a_S^\dagger + \text{c.c.} \ . \tag{2.8}$$

Equation (2.7) was obtained by substituting (2.1)–(2.3) into

$$\frac{-\partial F}{\partial t} = V\frac{\partial a_L}{\partial t} a_f^\dagger a_S^\dagger + V a_L \frac{\partial a_f^\dagger}{\partial t} a_S^\dagger + V a_L a_f^\dagger \frac{\partial a_S^\dagger}{\partial t} + \text{c.c.} \ .$$

The term

$$V c_L \frac{\partial a_L}{\partial x} a_f^\dagger a_S^\dagger + V c_S a_L a_f^\dagger \frac{\partial a_S^\dagger}{\partial x} + \text{c.c.}$$

vanishes since a Stokes photon is created for every laser photon annihilated and the propagation of the two photons is the same for $c_S \cong c_L$.

Setting the time derivatives of n_f, n_S, and F equal to zero, as discussed in Sec. IV, gives the steady-state solution

$$c_S \frac{\partial n_S}{\partial x} = 4|V|^2\Gamma^{-1}[n_L(n_f+n_S) - n_S n_f] \tag{2.9}$$

and

$$n_R^{-1}[n_L(n_f+n_S) - n_S n_f] - n_f = 0 \ , \tag{2.10}$$

where $n_R \equiv \Gamma^2/4|V|^2$. Neglecting the saturation term $n_S n_f$ in (2.10), which has been considered elsewhere[12] and not apposite to the argument, and solving for n_f gives

$$n_f = \frac{(n_L/n_R)n_S}{1 - n_L/n_R} \ . \tag{2.11}$$

The results (2.11) and (2.9) with $n_S n_f$ neglected give the enhanced stimulated-Raman-scattering result

$$n_S = +n_S(0)e^{\beta_{g\,\text{new}} x} \ , \tag{2.12}$$

where

$$\beta_{g\,\text{new}} = \frac{(\Gamma/c_S)n_L}{n_R(1-n_L/n_R)^{-1}} \ .$$

The results (2.9)–(2.12) are identical to the results derived previously[4] using the golden rule. In passing, notice that n_L can be considered as a constant in (2.7).

III. LOSS ENHANCEMENT IN PREVIOUS ANALYSES

Previous treatments[2,3] of stimulated Raman scattering did not yield the enhancement obtained in Sec. II. These analyses either specifically assumed no increase in the vibrational energy above the thermal-equilibrium value or solved Eqs. (2.1) and (2.2) or their equivalents by a method equivalent to that described below. In the former case, (2.11) [or (4.19) in Sec. IV] is replaced by

$$n_f = \bar{n}_f \ .$$

Substituting this expression into (2.9), neglecting the saturation term $n_S n_f$, and solving for n_S gives

$$n_S = \bar{n}_f(e^{\beta_{\text{old}} x} - 1) + n_S(0)e^{\beta_{\text{old}} x} \ , \tag{3.1}$$

where $\beta_{\text{old}} = 4|V|^2 n_L/\Gamma c_S = (\Gamma/c_S)(n_L/n_R)$. This is just the previous Raman gain factor with no enhancement.

In the latter of these two previous type of analyses, the nonlinear Eqs. (2.1) and (2.2), which were treated as *classical* equations, were linearized and decoupled from (2.3) by assuming that

$$a_L \equiv b_L e^{-i\omega_L t} \ , \tag{3.2}$$

where b_L is independent of time. Then substituting $a_f = b_f e^{-i\omega_f t}$ and $a_S^\dagger = b_S^\dagger e^{i\omega_S t}$ into (2.1) and the Hermitian conjugate of (3.2) gives

$$\frac{\partial b_f}{\partial t} = V b_L b_S^\dagger - \tfrac{1}{2}\Gamma b_f \ , \tag{3.3a}$$

$$\frac{\partial b_S^\dagger}{\partial t} = V^* b_L^\dagger b_f - c_S \frac{\partial b_S^\dagger}{\partial x} \tag{3.3b}$$

for the case of resonance, that is, $\omega_L = \omega_S + \omega_f$. Setting the time derivatives equal to zero and eliminating b_f from the two equations gives

$$c_S \frac{\partial b_S}{\partial x} = 2|V|^2 n_L b_S/\Gamma \ ,$$

which has the solution

$$b_S(x) = b_S(0)e^{\beta x} \ , \tag{3.4}$$

where $\beta = 2|V|^2 n_L/c_S\Gamma$, which shows no gain enhancement. A nonzero steady-state solution (3.4) was obtained in this previous analysis because the pump b_L acts as a source. This is particularly clear in the analogous problems of a parametrically pumped pendulum or moving-plate capacitor, where the energy is supplied by the mechanism that changes the length of the pendulum or moves the capacitor plates.

In order to further show how the assumption (3.2) causes the loss of enhancement, (3.3) with b_L independent of time will be solved by another method, in direct analogy with the solution of Sec. II where the time dependence of b_L was retained. By the same method used in Sec. II, (3.3a) and (3.3b) give

$$\frac{\partial n_f}{\partial t} = F - \Gamma n_f \ , \tag{3.5}$$

$$\frac{\partial n_S}{\partial t} = F - c_S \frac{\partial n_S}{\partial x} \ , \tag{3.6}$$

$$\frac{\partial F}{\partial t} = 2|V|^2 n_L(n_f+n_S) - \tfrac{1}{2}\Gamma F - \left(c_S V b_L b_f \frac{\partial b_S}{\partial x} + \text{c.c.}\right) \ . \tag{3.7}$$

By neglecting the time dependence of b_L, the nonlinear term $-2|V|^2 n_f n_S$ in (2.7) is lost and the last

term in (3.7), which did not appear in (2.7), is gained. The former makes the solution incorrect in the saturation region and the latter eliminates the enhancement. Neglecting the time dependence of b_L is equivalent to neglecting the last two terms in (2.3) for da_L/dt. By so neglecting the last term $c_L \partial a_L/\partial x$ in (2.3), the cancellation of the similar term $c_S \partial a_S/\partial x$ from (2.2) does not occur in the equation for $\partial F/\partial t$; thus, the last term in (3.7) is present. Neglecting the other term $V^* a_f a_S$ in (2.3) corresponds directly to the absence of the term $2|V|^2 n_S n_f$ in (3.7). This discussion indicates that the physical significance of the linearization by using (3.2) is that the effect of increases in the amplitudes a_f and a_S on the amplitude a_L is neglected and the spatial rate of change of a_L is neglected while a comparable term of a_S is retained.

Stated differently, it is tempting to neglect the time derivative of b_L when b_L is large so that the fractional change in b_L is small. However, db_L/dt must be retained since it is of comparable magnitude to other terms such as db_S/dt that are retained. It makes no difference that the fraction change in b_S is large while that in b_L is small.

Finally, the loss of enhancement in (3.5)–(3.7) can be seen by setting the time derivatives equal to zero, $\partial n_S/\partial x = \beta n_S$, and $\partial b_S/\partial x = \frac{1}{2}\beta b_S$ in (3.5)–(3.7) and eliminating F. This gives

$$2|V|^2 n_L(n_f + n_S) - \tfrac{1}{2}(\Gamma + c_S\beta)c_S\beta n_S = 0 ,$$

$$c_S\beta n_S = \Gamma n_f .$$

Eliminating n_f gives

$$2|V|^2\Gamma^{-1} n_L(c_S\beta + \Gamma) - \tfrac{1}{2}(c_S\beta + \Gamma)c_S\beta = 0 .$$

Dividing by $\frac{1}{2}c_S(c_S\beta + \Gamma)$ gives

$$\beta = \beta_{\text{old}} \equiv 4|V|^2/c_S\Gamma ,$$

in agreement with (3.4).

IV. FERROMAGNETIC INSTABILITIES, QUANTUM-MECHANICAL EFFECTS, AND PHASES

The considerations of this section are not essential to the explanation of differences in the three types of treatment of enhanced Raman scattering, but are of general interest. First consider the parametric instability in the simplest three-boson process, illustrated in Fig. 1, where one boson, 0, is annihilated and two bosons having equal frequencies and damping are created. The propagation of all three bosons is negligible. As specific examples, in the case of ferromagnetic subsidiary-resonance absorption,[5–7] boson 0 is a uniform precession (wave vector $\vec{k} = 0$) magnon, and in parallel pumping, boson 0 is a photon in the microwave cavity. In both cases, the output bosons are magnons having wave vectors $\vec{k}$ and $-\vec{k}$. Propagation effects are negligible since the magnons cannot propagate out of the sample and the sample is small with respect to the electromagnetic wavelength. The process also represents phonon processes[12] and other boson processes.

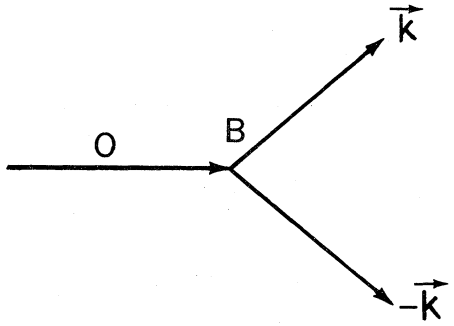

FIG. 1. Three-boson splitting process that exhibits a parametric instability.

It will be demonstrated that the equations of motion of the mode amplitudes can be solved to give the golden-rule results directly. The same mode-amplitude equations will be solved by an approximate method of converting nonlinear differential equations into linear differential equations with time-dependent coefficients, or so called parametric equations. This approximate method, which is the same method used in the early treatments of the stimulated Raman scattering, gives incorrect results in the present magnon problem as it did in the stimulated-Raman-scattering problem.

The equations of motion of the Fourier components, a's of the $k = 0$ and the $+\vec{k}$ and $-\vec{k}$ modes, denoted 0, +, and −, and the complex conjugates are[5,7]

$$\frac{da_+}{dt} = -i\omega_+ a_+ - Ba_0 a_-^\dagger - \gamma a_+ , \tag{4.1}$$

$$\frac{da_-^\dagger}{dt} = -i\omega_- a_-^\dagger - B^* a_0^\dagger a_+ - \gamma a_-^\dagger , \tag{4.2}$$

$$\frac{da_0}{dt} = -i\omega_0 a_0 + B^* a_+ a_- , \tag{4.3}$$

where B is the coupling constant. This set of Eqs. (4.1)–(4.3) and the Hermitian-conjugate equations is a set of six nonlinear classical differential equations for the six variable a_i and $a_i^\dagger$, with $i = 0, +, -$.

A previous classical method of solution was to linearize the equations by formally assuming that $a_0 = b_0 e^{-i\omega_0 t}$ and $a_0^\dagger = b_0^\dagger e^{i\omega_0 t}$, where b_0 and $b_0^\dagger$ are constants, as in Sec. II. Then (4.1) and (4.2) are a set of two parametric differential equations for a_+ and $a_-^\dagger$. Substituting these expressions for a_0 and $a_0^\dagger$ along with $a_+ = b_+ e^{-\omega_+ t}$ and $a_-^\dagger = b_-^\dagger e^{i\omega_- t}$ into (4.2) and (4.3) and taking the derivatives gives

$$\frac{db_+}{dt} = Bb_0 b_-^\dagger - \gamma b_+ , \tag{4.4}$$

$$\frac{db_-^\dagger}{dt} = B^* b_0^\dagger b_+ - \gamma b_-^\dagger \,. \tag{4.5}$$

The time-dependent coefficients were eliminated by assuming that the resonance condition $\omega_0 = \omega_+ + \omega_-$ is satisfied. Since these two equations are linear with constant coefficients, their solution is simple. Taking the derivative of (4.4), using (4.5) to eliminate b_-, and substituting the trial solution $b_+ = b_{+0} e^{\lambda t}$ into the resulting equation gives

$$a_+ = e^{-i\omega_+ t} e^{-\gamma t} (b_+^{(+)} e^{|B b_0| t} + b_+^{(-)} e^{-|B b_0| t}) \,. \tag{4.6}$$

There is no nonzero steady-state solution except in the singular case of $|Bb_0| = \gamma$. The common expression "parametric instability" arises from the fact that a_+ becomes infinite as $t \to \infty$ when the amplitude $|b_0|$ of the zero mode is sufficiently great, that is, when

$$|b_0|^2 > \gamma^2 / |B|^2 \,. \tag{4.7}$$

The classical effect of neglecting the time dependence of b_0 is somewhat less dramatic in the present case of the magnon instability than in the Raman scattering case since the correct steady-state solution for small values of b_0 is $a_+ = 0$ even when the time dependence of b_0 is retained. In the case of large values of b_0, the difference between the zero steady-state solution obtained for the case of b_0 independent of time and the nonzero steady-state solutions (4.13) and (4.14) for the case of time dependence included is of course more significant. In passing it is mentioned that in parametric equations the terms such as $Bb_0 b_-^\dagger$ and its complex conjugate in (4.4) and (4.5) are source terms, which give rise to the energy flow from the $\vec{k} = 0$ to the $\pm\vec{k}$ modes. Thus, the steady-state solution is not $b_+ = b_- = 0$, in general.

To resolve the difficulty of no classical steady-state solution, a method of solution other than the parametric linearization used above is needed. One approach would be to linearize (4.1) simply by considering $a_0 a_-^\dagger$ as a single variable. Since $a_0 a_-^\dagger$ is coupled to a_+ according to (4.1), the standard procedure is to consider the equation of motion for $a_0 a_-^\dagger$. If this equation contains only a_+ and $a_0 a_-^\dagger$ (and no product $a_+ a_0 a_-^\dagger$), then the two linear differential equations could be easily solved. Unfortunately the equations do not uncouple at this step. Furthermore, taking the derivatives of the additional variables that appear in the $a_0 a_-^\dagger$ equation couples in still more variables, and the chain of equations becomes large. Nevertheless, this method, along with several other powerful and elegant methods, afford useful tools for attacking the problem. As already noted in Sec. II, a simpler method is to start with the operator $a_+^\dagger a_+ \equiv n_+$, rather than a_+. Just as in Sec. II, it is found that

$$\frac{dn_+}{dt} = F_+ - \Gamma(n_+ - \bar{n}_+) \,, \tag{4.8}$$

$$\frac{dn_-}{dt} = F_+ - \Gamma(n_- + \bar{n}_-) \,, \tag{4.9}$$

$$\frac{dn_0}{dt} = -F_+ \,, \tag{4.10}$$

$$\frac{dF_+}{dt} = 2|B|^2 [n_0(n_+ + n_-) - n_+ n_-] - \Gamma F_+ \,, \tag{4.11}$$

where $F_+ = -Ba_0 a_+^\dagger a_-^\dagger + \text{c.c.}$ and $\Gamma \equiv 2\gamma$, the factor of 2 arising as usual from the fact that $a \sim e^{-\gamma t}$ implies that $|a|^2 \sim e^{-2\gamma t}$. The terms $\Gamma\bar{n}_+$ and $\Gamma\bar{n}_-$ were added formally to make n_+ and n_- relax to their thermal-equilibrium values $\bar{n}_+$ and $\bar{n}_-$. As in the previous cases, the treatment is classical. If the usual commutation relations for the $a^\dagger$ and a are used formally, then $n_+ + n_-$ in (4.11) is replaced by $n_+ + n_- + 1$. The considerations of the commutation relations and of adding damping, noise sources, and thermal-equilibrium values that are addressed below for the case of Raman scattering are not considered here since this would carry us too far afield.

There are several physical situations for which the solutions to (4.8)–(4.11) are of interest. The first is that to which the golden rule is commonly applied. That is, at time $t = 0$ the system is in the state in which n_L is very large and all other modes are in thermal equilibrium, roughly speaking. The perturbation $-i\hbar B a_0 a_+^\dagger a_-^\dagger + \text{c.c.}$ is then applied for a time short with respect to the time for n_0 to change substantially, but sufficiently long for energy conservation to be well satisfied. The case in which n_0 is maintained at a constant value by the microwave field in the cavity also is of interest. In both of these cases, n_0 is constant, or approximately constant. However, dn_0/dt in (4.10) is not zero because (4.10) is only the contribution to the rate of change of n_0 from the coupling to the $\pm k$ modes. Stated differently, $\hbar\omega_0 dn_0/dt$ from (4.10) gives the power from the zero mode to the pair $\pm\vec{k}$, which is not zero in the steady state. The contribution to dn_0/dt from the coupling of the zero mode to the microwave field could be added, by replacing (4.10) with $dn_0/dt = \text{const} n_0 - F_+$, for example, but this would carry us too far from the issue at hand. In passing notice the physical significance of F_+ as the energy flow from the zero mode to the pair $\pm\vec{k}$, in units of quanta per second.

The steady-state solution to (4.8) and (4.9) is obtained by setting $dF_+/dt = 0$ (i.e., constant energy flow) and $dn_+/dt = 0$ and solving for n_+ and n_-. This gives

$$n_- = n_+ = 2|B|^2 \Gamma^{-2} (2n_0 n_+ - n_+^2) \,, \tag{4.12}$$

which are the standard results obtained from the golden rule. For $n_0 < n_c - \epsilon$, where ϵ is very small,

the solution to (4.12) is

$$n_- = n_+ = \frac{\bar{n}_+}{1 - n_0/n_c} , \quad n_c \equiv \frac{\Gamma^2}{4|B|^2} , \tag{4.13}$$

where $\bar{n}_- = \bar{n}_+$, for $n_0 > n_c + \epsilon$ the solution is

$$n_- = n_+ = 2(n_0 - n_c) , \tag{4.14}$$

and for $n_0 = n_c$, the solution is $n_- = n_+ = (2n_0\bar{n}_+)^{1/2}$.

These results are obtained simply from the golden rule as follows: The standard expression $(2\pi/\hbar^2) \times |\langle f|\mathcal{H}|i\rangle|^2 \delta(\omega)$ for the transition rate between states $|i\rangle$ and $|f\rangle$ gives

$$\frac{dn_+}{dt} = \frac{2\pi}{\hbar^2}(|\mathcal{H}|_+^2 - |\mathcal{H}|_-^2)\rho(\omega) - \Gamma(n_+ - \bar{n}_+) ,$$

where $|\mathcal{H}|_+^2$ is the matrix element for increasing n_+ by 1, $|\mathcal{H}|_-^2$ is that for decreasing n_+ by 1, $\rho(\omega)$ is the density of states, and the relaxation term $\Gamma(n_+ - \bar{n}_+)$ is added formally. For a single transition on resonance ($\omega_0 = \omega_+ + \omega_-$) the appropriate value of $\rho(\omega)$ is[7,6,4] $\rho(\omega) = 1/\pi\Gamma$. Using the usual expressions for the matrix elements of the a's and $a^\dagger$'s gives

$$\frac{dn_+}{dt} = 2|B|^2\Gamma^{-1}[(n_+ + 1)(n_- + 1)n_0 - n_+n_-(n_0 + 1)] - \Gamma(n_+ - \bar{n}_+). \tag{4.15}$$

Since the bracket factors in (4.15) and (4.11) (with the replacement $n_+ + n_- \to n_+ + n_- + 1$) are equal, the steady-state solution to (4.15) is given by (4.13) and (4.14) as already mentioned. Notice that when the factor of 1 is added to (4.11), $\bar{n}_+$ in (4.13) is replaced by $\bar{n}_+ + n_0/2n_c$.

Even though only the steady-state case is considered here, it should be mentioned that the transient solutions of (4.15) and of (4.8)–(4.10) are different in general. The simplest case of n_+n_- negligible, $n_+(0) = \text{const}$, and $n_0 = \text{const}$ can be solved trivially to illustrate this point.

In the quantum-mechanical treatment it is well known[9(b),9(c),13,14] that formally adding the term $-\frac{1}{2}\Gamma a_f$ to Eq. (2.1) for da_f/dt is inconsistent with the commutation relation $[a_f, a_f^\dagger] = 1$. The difficulties with the commutation relations and with relaxation to zero have been the subjects of numerous previous investigations.[5,9,13,14] In the magnon problem, relaxation to thermal equilibrium has been treated *classically* by formally adding a noise source.[5] Both the commutation-relation problem and that of relaxing to zero can be treated by enlarging the system to include the modes that are responsible for the damping, that is, by including a damping reservoir explicitly.[9(c),13,14] Since noise problems are not addressed in the present study, it is not surprising that the phenomenological damping used above can be justified within the usual enlarged-system approach as follows.

The simplest damping mechanism is that of two-boson damping induced by a localized imperfection such as a void or impurity ion. (For nonlocalized imperfections, wave vector is conserved and the sum on j below is reduced to a single term.[7]) Even though this damping mechanism is not often the actual dominant mechanism, we follow the standard practice of using it in order to illustrate how including the damping reservoir resolves the commutator difficulty and gives relaxation to thermal equilibrium. The reservoir is taken as a set of bosons, say phonons to be concrete, with creation and annihilation operators $a_j^\dagger$ and a_j. Then the Hamiltonian for the interaction with the reservoir is

$$\mathcal{H}_i = \sum_j \hbar\Omega_j a_f a_j^\dagger + \text{H. c.} ,$$

where H. c. denotes Hermitian conjugate. The Heisenberg equation of motion of a_f, with this interaction included, is (2.1) with $-\frac{1}{2}\Gamma a_f$ replaced by

$$G'_{af} \equiv -i\sum_j \Omega_j a_j .$$

By writing the Heisenberg equations of motion of a_j and $a_j^\dagger$ as integral equations which are substituted into G'_{af}, taking the Laplace transform of G'_{af}, using the approximation

$$[s - i(\omega_f - \omega_j)]^{-1} \cong \pi\delta(\omega_f - \omega_j) + i\mathcal{P}(\omega_f - \omega_j)^{-1}$$

(since the pole of interest is near $s = 0$ in the perturbation-theory limit[15]) with $\mathcal{P}$ denoting the principal part and s the transform variable, and inverting the transform gives

$$G'_{af} \equiv -i\sum_j \Omega_j a_j \cong -\tfrac{1}{2}\Gamma a_f + G_{a_f} , \tag{4.16}$$

where

$$G_{a_f} = -i\sum_j \Omega_j a_j(0) e^{-i\omega_j t}$$

is the noise source and

$$\Gamma = 2\pi\sum_j |\Omega_j|^2 \delta(\omega_j - \omega_f)$$

is the golden-rule result for the relaxation frequency. The real parts of the frequency shifts arising from $\mathcal{P}$ are ignored for simplicity.

The equations of motion for the n's and F can be obtained directly from the Heisenberg equations or from the equations for the a's, with $-\frac{1}{2}\Gamma a_f$ replaced by the right-hand side of (4.16). In either case, using

$$\langle G_{n_f}(t)\rangle \equiv \left\langle -\sum_j \Omega_j a_j(0) a_j^\dagger(t) e^{-i\omega_j t} + \text{H. c.}\right\rangle \cong \Gamma\bar{n}_f ,$$

which is not difficult to derive,[9(c)] gives

$$\frac{dn_f}{dt} = F - \Gamma(n_f - \bar{n}_f) + g_{n_f}(t) , \tag{4.17}$$

where

$$g_{n_f}(t) = G_{n_f}(t) - \langle G_{n_f}(t)\rangle$$

and similar equations for n_S, n_L, and F. Taking the averages over the reservoir (the phonon-bath mode) of all terms in (4.22) and using $\langle g_{n_f}\rangle = \langle G_{nf}\rangle - \langle G_{n_f}\rangle = 0$ gives

$$\frac{d\langle n_f\rangle}{dt} = \langle F\rangle - \Gamma(\langle n_f\rangle - \bar{n}_f) \tag{4.18}$$

and similar equations for $\langle n_S\rangle$, $\langle n_L\rangle$, and $\langle F\rangle$. Thus, (2.4)–(2.7) with the $\bar{n}_f$ term added as above are valid when the operators n_f, etc., are interpreted as averages over the *reservoir*. Recall that $\bar{n}_f$ is the thermal-equilibrium value of n_f, that is, the average over the complete ensemble, while $\langle n_f\rangle$ is the average over the reservoir only (*phonons* in thermal equilibrium). Notice that in the absence of the source, $\langle a_f\rangle$ relaxes to zero while $\langle a_f^\dagger a_f\rangle$ relaxes to the thermal-equilibrium value $\bar{n}_f$.

The following physical description of the damping and noise term is useful in visualizing these effects even though the explanation is oversimplified. The energy flow from the f phonons to the phonons in the bath is not a smooth function of time. For example, if the energy transfer is visualized as the interchange of quanta of energy between the f phonons and the bath, then in the time interval 0 to t_1, there may be 10 net quanta of energy entering the bath. In the interval t_1 to $2t_1$ there may be 8, in the interval $2t_1$ to $3t_1$ there may be 12, and so forth. The average value of, say, 10 quanta per time t_1 corresponds to Γ, and the fluctuations of, say, 2 quanta per time t_1 correspond to the noise term that averages to zero. There are also fluctuations in the energy flow F from the laser photons L to the Stokes photons S and phonons f since F depends on the fluctuating amplitude of the f modes.

This visualization explains why it is not rigorously correct to set the various time derivatives equal to zero in the "steady state." That is, the fluctuations are still present when average values have reached constant values. Since the interest here is in the average values rather than the fluctuations, the time derivatives can be set equal to zero, or, mathematically, the reservoir averages can be used in order to eliminate the fluctuation terms.

Consider the purely quantum-mechanical effects in the Raman scattering problem. When the mode amplitudes in (2.1)–(2.3) are operators with the usual commutation relation $[a_i, a_i^\dagger] = 1$, the factors $n_f + n_S$ in (2.7), (2.9), and (2.10) are replaced by $n_f + n_S + 1$. Replacing Γn_f by $\Gamma(n_f - \bar{n}_f)$ formally in the relaxation terms in (2.4) and (2.10) gives a relaxation of n_f to its thermal-equilibrium value $\bar{n}_f$. The complete expression $\Gamma(n_f - \bar{n}_f)$ is of course obtained in standard quantum-mechanical calculations of relaxation.[9,7] With these two additions, (2.11) and (2.12) become

$$n_f = \frac{\bar{n}_f + (n_L/n_R)(n_S + 1)}{1 - n_L/n_R}\ , \tag{4.19}$$

$$n_S = (\bar{n}_f + 1)(e^{\beta g_{\text{new}} x} - 1) + n_S(0)e^{\beta g_{\text{new}} x}\ . \tag{4.20}$$

Thus, comparison of (4.20) with (2.12) shows that the quantum-mechanical effect is that the zero-point oscillation is amplified ($e^{\beta g_{\text{new}} x} - 1$ term), and the effect of including $\bar{n}_f$ is that the term $\bar{n}_f(e^{\beta g_{\text{new}} x} - 1)$ accounts for "amplification of the thermal-equilibrium value of n_f."

Finally, consider the physical explanation of the result that the loss of phase information in using the golden rule does not affect the final result in view of the fact that the phases are important in some sense. Specifically, a parametric process which increases the amplitude of a given phase will decrease the amplitude of a mode that is π rad out of phase with the increasing mode. The reason that the loss of phase information in using occupation numbers is not important in the final result is that the modes in the original thermal distribution that have the correct phase are the ones that are amplified. A similar situation exists in the simple case of a classical harmonic oscillator responding to an applied harmonic force. The phase of the oscillator is important since it determines whether energy is extracted from or delivered to the driving force. This does not imply that energy-conservation arguments, which suffer from an analogous loss of phase information, are not valid. It should be mentioned that wave-vector and frequency phase matching *are* included in the occupation-number approach. Wave-vector phase matching arises from Kronecker δ's in sums over wave vectors, and frequency phase matching arises from the energy-conserving δ function. The present calculation settles the question of the importance of phases in obtaining the enhancement by showing explicitly that the phases are unimportant in the result.

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*Research supported by the Advanced Research Projects Agency of the Department of Defense and monitored by the Defense Supply Service, Washington, D. C.

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[15]The reader with explicit interest in this reservoir approach should transform the complete differential equation [Ref. 9(c)]. The results are unchanged.