Theory of Parametric Gain near a Lattice Resonance

C. H. Henry and C. G. B. Garrett
Bell Telephone Laboratories, Murray Hill, New Jersey
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The parametric gain is computed for the case of a cubic crystal having a single lattice resonance as the frequency of one of the generated waves, the idler, is swept through the lattice resonance. It is found that as resonance is approached, the nonlinear susceptibility and the infrared absorption at the idler frequency both become resonantly large in just such a manner as to leave the parametric gain unaltered. A simple expression is derived for the parametric gain both by solving the coupled-wave equations and by calculating the transition rate for light scattering. The parametric gain is estimated numerically for gallium phosphide.

I. INTRODUCTION

Parametric oscillation has been used by Giordmaine and Miller[1] and by others[2,3] to generate visible and near-infrared light. In principle, parametric oscillation could also be used to generate far-infrared radiation. In this case, one of the generated signals, which we will call the idler, would be close in energy to the infrared lattice-absorption resonance of the crystal. As the idler approaches the lattice resonance, the absorption coefficient at the idler frequency becomes resonantly large, but so does the nonlinear susceptibility describing the parametric amplification process.[4] It is not clear what happens to the parametric gain in this situation. In this paper, we compute the parametric gain as the idler frequency is swept through the lattice resonance.

We restrict our analysis to the simplest case, a cubic crystal with a single lattice resonance such as gallium phosphide. In the process of parametric amplification, a pump wave is used to generate a signal wave and an idler wave.[5] We shall assume that the crystal is transparent to both the pump wave and the signal wave, but that the absorption coefficient for the idler wave is large compared with the parametric gain. In practical situations, the parametric gain will be of order 1 cm^{-1}. For idler energies 160–780 cm^{-1}, the infrared absorption coefficient in gallium phosphide is greater than 10 cm^{-1} at 300°K.[6] It rises to a peak value of about 4×10^4 cm^{-1} at the lattice resonance, which occurs at an energy of 366 cm^{-1}. Our treatment will apply to gallium phosphide over this energy range. In this near-resonance region, the idler quanta are both photonlike and phononlike in character and are called polaritons.[7,8]

[1] J. A. Giordmaine and R. C. Miller, Phys. Rev. Letters **14**, 973 (1965).
[2] S. A. Akhmanov, A. I. Korrygin, A. S. Piskaraltas, V. V. Fadeev, and R. V. Khoklov, Zh. Eksperim. i Teor. Fiz. Pis'ma v Redaktsiyu **3**, 372 (1966) [English transl.: Soviet Phys.—JETP Letters **3**, 241 (1966)].
[3] L. B. Kreuzer, Appl. Phys. Letters **10**, 336 (1967).
[4] W. L. Faust and C. H. Henry, Phys. Rev. Letters **17**, 1265 (1966). Our notation is the same as in this reference, except that we have written the nonlinear constant d_Q as Nd_Q.
[5] M. Bloembergen, *Nonlinear Optics* (W. A. Benjamin, Inc., New York, 1965), Sec. 4.4.
[6] A. S. Barker, Phys. Rev. **165**, 917 (1968).
[7] J. J. Hopfield, Phys. Rev. **112**, 1555 (1958).
[8] C. H. Henry and J. J. Hopfield, Phys. Rev. Letters **15**, 964 (1965).

The problem of calculating the parametric gain near a lattice resonance has been previously treated by Loudon,[9] by Butcher, Loudon and McLean,[10] and by Shen.[11] Each of these papers presented a similar but somewhat different expression for the parametric gain. The purpose of our paper is to clear up the discrepancies between these papers by deriving a simple analytical formula for the parametric gain first by Shen's method of solving the coupled-wave equations and second by using Loudon's approach of calculating the transition rate for the light-scattering process.

Our expression for the gain has a number of simple features which are illustrated in Figs. 1 and 2:

(a) The maximum gain occurs for the case of wave-vector matching $\mathbf{k}_3=\mathbf{k}_1+\mathbf{k}_2$, where $\mathbf{k}_3$ is the wave vector of the linear pump wave, $\mathbf{k}_2$ is the wave vector of the linear idler wave, and $\mathbf{k}_1$ is the wave vector of the linear idler wave calculated in the absence of damping. (This was pointed out by Shen.) The undamped $\mathbf{k}_1$ (polariton) dispersion curve is shown in Fig. 1.

(b) The peak gain $(g_2)_{\max}$ is slowly varying with idler frequency and is essentially unchanged as the idler frequency is swept from several linewidths below resonance onto the lattice resonance (see Fig. 2).

(c) The variation of parametric gain with the idler frequency results mainly from the interference of the two terms which contribute to the nonlinear susceptibility. For gallium phosphide, this interference causes the parametric gain to go to nearly zero at an idler energy $\hbar\omega_1$ of 250 cm^{-1} (see Fig. 2).[4]

(d) The ratio of the idler power to the signal power is inversely proportional to the infrared absorption coefficient at the idler frequency. Because of this, the amount of infrared light that can be generated becomes negligible at the idler frequency approaches the lattice resonance and absorption becomes large (see Fig. 2).

We use Shen's method to calculate the parametric gain in Sec. II. Shen's[11] treatment is more general than ours. When spatial dispersion and nonlattice infrared absorption effects, included by Shen, are neglected, his gain formula reduces to ours. (An algebraic error in his

[9] R. Loudon, Proc. Phys. Soc. (London) **82**, 393 (1963).
[10] P. N. Butcher, R. Loudon, and T. P. McLean, Proc. Phys. Soc. (London) **85**, 565 (1965).
[11] Y. R. Shen, Phys. Rev. **138**, A1741 (1965).

paper must also be corrected.[12]) The generality of his treatment tends to obscure several of the results stressed in this paper.

In Sec. III, we calculate the parametric gain using Loudon's[9] method. We take into account the frequency dependence of the idler damping constant, which Loudon neglected. This leads to a simpler expression for the parametric gain.

The method used by Butcher *et al.*[10] will not be discussed. These authors neglected certain second derivatives of the field amplitudes at the beginning of their calculation. They failed to realize that their approximation is invalid very close to the lattice resonance, where the infrared absorption coefficient is very large. This mistake resulted in their deriving an erroneous expression for the parametric gain near the lattice resonance. The correct expression is in fact a good deal simpler than that found by Butcher *et al.*

In Sec. IV, we relate the parametric gain to the spontaneous Raman-scattering cross section, discuss the efficiency of producing infrared light, and numerically estimate the parametric gain for gallium phosphide.

II. PARAMETRIC GAIN FROM COUPLED-WAVE EQUATIONS

We will assume that the pump, signal, and lattice fields are plane waves. The pump amplitude is fixed, while the other fields may grow (or decay) exponentially. Accompanying the electric field $E(\omega_1)$ at the idler frequency ω_1 is a transverse optical lattice wave $Q(\omega_1)$. The quantity $Q(\omega_1)/E(\omega_1)$ becomes resonantly large as ω_1 approaches ω_0, the resonant frequency of the lattice. We write these fields as

$$\begin{aligned} E(\text{pump}) &= E(\omega_3)+\text{c.c.} = A_3 e^{i(\mathbf{k}_3\cdot\mathbf{r}-\omega_3 t)}+\text{c.c.}, \\ E(\text{signal}) &= E(\omega_2)+\text{c.c.} = A_2 e^{i(\mathbf{k}_2\cdot\mathbf{r}-\omega_2 t)+\gamma_2\cdot\mathbf{r}}+\text{c.c.}, \\ E(\text{idler}) &= E(\omega_1)+\text{c.c.} = A_1 e^{i(\mathbf{k}_1\cdot\mathbf{r}-\omega_1 t)+\gamma_1\cdot\mathbf{r}}+\text{c.c.}, \\ Q(\text{lattice}) &= Q(\omega_1)+\text{c.c.} = Q_1 e^{i(\mathbf{k}_1\cdot\mathbf{r}-\omega_1 t)+\gamma_1\cdot\mathbf{r}}+\text{c.c.} \end{aligned} \tag{1}$$

The coupled-wave equations and the force equation for the lattice oscillator $Q(\omega_1)$ are given by

$$\begin{aligned} [\nabla^2+(\omega_2^2/c^2)\epsilon_2]E(\omega_2) &= -(4\pi\omega_2^2/c^2) \\ &\quad \times P^{\text{NL}}(\omega_2=\omega_3-\omega_1), \\ [\nabla^2+(\omega_1^2/c^2)\epsilon_\infty]E(\omega_1) &= -(4\pi\omega_1^2/c^2) \\ &\quad \times P^{\text{NL}}(\omega_1=\omega_3-\omega_2), \\ \mu D(\omega_1)Q(\omega_1) &= eE(\omega_1)+F^{\text{NL}}(\omega_1=\omega_3-\omega_2), \end{aligned} \tag{2}$$

where $D(\omega_1)=\omega_0^2-\omega_1^2-i\omega_1\Gamma$. $Q(\omega_1)$ is the lattice displacement within the primitive cell, μ is the reduced mass, and e is the charge associated with the transverse optical lattice mode.

Following the approach of Kleinman[13] and of Bloembergen,[14] we assume that there exists a phenomenological energy-density function $U^{\text{NL}}[E(\omega_3),E(\omega_2),E(\omega_1),Q(\omega_1)]$ from which the nonlinear polarizations P^{NL} and the nonlinear force F^{NL} can be derived using

$$\begin{aligned} P^{\text{NL}}(\omega_i) &= -\partial U/\partial E(\omega_i)^*, \quad i=1,2,3 \\ F^{\text{NL}}(\omega_1) &= -\partial U/\partial Q(\omega_1)^*. \end{aligned} \tag{3}$$

In this case,

$$\begin{aligned} U = -[d_E E(\omega_3)E(\omega_2)^*E(\omega_1)^* \\ + d_Q N E(\omega_3)E(\omega_2)^*Q(\omega_1)^*]+\text{c.c.}, \end{aligned} \tag{4}$$

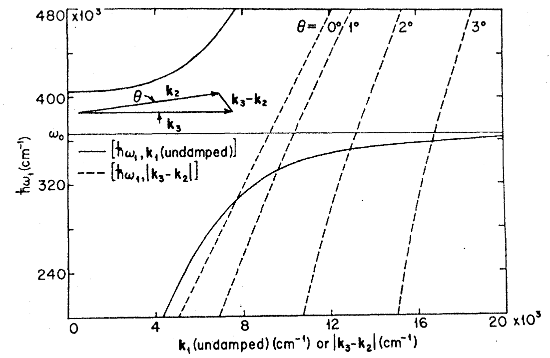

FIG. 1. Solid curve shows the undamped polariton dispersion curve (ω_1 versus k_1) for gallium phosphide. The peak parametric gain occurs when $k_1=|\mathbf{k}_3-\mathbf{k}_2|$. Dashed curves are $|\mathbf{k}_3-\mathbf{k}_2|$ versus $\omega_1=\omega_3-\omega_2$ for various scattering angles θ. These curves were calculated assuming the pump is the 6943 Å line of a ruby laser. The peak parametric gains occurs where the solid curves and the dashed curves cross.

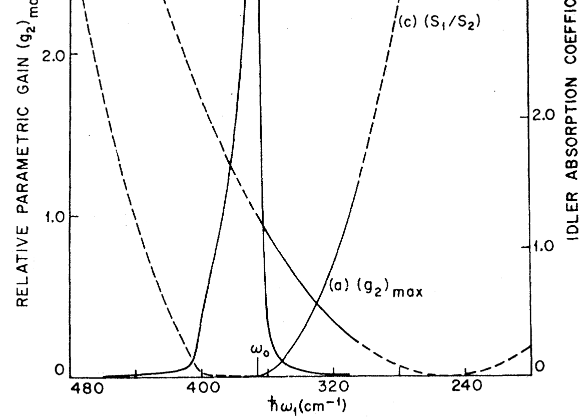

FIG. 2. (a) Relative parametric gain $(g_2)_{\max}$ versus the idler frequency ω_1 for gallium phosphide. Solid portion of the curve indicates the frequency range of the idler over which phase matching is possible. (b) Infrared absorption coefficient for gallium phosphide using $\Gamma=4$ cm^{-1}. (c) Ratio of the idler flux density S_1 to the signal flux density S_2.

[12] A_{123} listed by Shen after Eq. (6) is too large by a factor of 2.
[13] D. A. Kleinman, Phys. Rev. **126**, 1977 (1962).
[14] Reference 5, Sec. 1.3.

where N is the number of primitive cells per cm^3. Garrett[15] has shown, using an explicit anharmonic oscillator model which reproduces the optical nonlinearities of the crystal, that d_E and d_Q are slowly varying and may be taken as real and constant for the experimental frequency ranges we have in mind, i.e., $160 \leq \omega_1 \leq 780$ cm^{-1} for gallium phosphide. These assumptions have been verified experimentally by Faust and Henry,[4] who were able to fit their measured resonant frequency dependence of the nonlinear susceptibility $P^{NL}(\omega_2)/E(\omega_1)E(\omega_3)$ assuming d_E and d_Q to be real and constant. The nonlinear susceptibility constants d_Q and d_E are actually third-order tensors. For a cubic material like gallium phosphide, these tensors are determined by one parameter. The tensor notation adds nothing important to the problem and will not be included. The relative magnitude of the nonlinear polarization for various polarizations of $E(\omega_1)$, $E(\omega_2)$, and $E(\omega_3)$ has been discussed by Loudon.[16]

Equation (2) can be solved only by equating the exponents on each side of the equation. This gives

$$\mathbf{k}_1+\mathbf{k}_2=\mathbf{k}_3\,,\qquad \gamma_1=\gamma_2\equiv\gamma\,. \tag{5}$$

We may simplify notation by defining several new symbols:

$$\begin{aligned} d_Q' &= d_Q/e\,,\\ d_E' &= 4\pi d_E\,,\\ \omega_P^2 &= 4\pi Ne^2/\mu\,,\\ Q_1' &= 4\pi eNQ_1\,. \end{aligned} \tag{6}$$

The coupled amplitude equations then become

$$\begin{aligned} d_E'A_3A_1^*+d_Q'A_3Q_1'^* &+[\epsilon_2-(c^2/\omega_1^2)(\mathbf{k}_2-i\gamma)^2]A_2=0\,,\\ [\epsilon_\infty-(c^2/\omega_1^2)(\mathbf{k}_1+i\gamma)^2]A_1^*&+Q_1'^*+d_E'A_3^*A_2=0\,,\\ -\omega_P^2A_1^*+D(\omega_1)^*Q_1'^*&-\omega_P^2d_Q'A_3^*A_2=0\,. \end{aligned} \tag{7}$$

This leads to the secular equation

$$\begin{vmatrix} d_E'A_3 & d_Q'A_3 & \epsilon_2-(\omega_2^2/c^2)(\mathbf{k}_2-i\gamma)^2\\ \epsilon_\infty-(c^2/\omega_1^2)(\mathbf{k}_1+i\gamma)^2 & 1 & d_E'A_3^*\\ -1 & D(\omega_1)^*/\omega_P^2 & -d_QA_3^* \end{vmatrix}=0\,. \tag{8}$$

We define the parametric gains g_2 and g_1 of the signal and the idler waves as twice the projection of γ along the propagation directions $\hat{k}_2$ and $\hat{k}_1$:

$$g_2\equiv 2\hat{k}_2\cdot\gamma\,,\qquad g_1\equiv 2\hat{k}_1\cdot\gamma\,. \tag{9}$$

Neglecting terms of order γ^2, we may write

$$\begin{aligned}(\mathbf{k}_2-i\gamma)^2&=k_2^2-ik_2g_2\,,\\ (\mathbf{k}_1+i\gamma)^2&=k_1^2+ik_1g_1\,.\end{aligned} \tag{10}$$

The secular equation (8) thus establishes a relation between g_1 and g_2. Expanding the secular equation, we have

$$\begin{aligned}\left[\left(k_2^2-\frac{\omega_2^2}{c^2}\epsilon_2\right)-ik_2g_2\right]&\left[\left(k_1^2-\frac{\omega_1^2}{c^2}\epsilon_1'\right)+i\left(\frac{\omega_1^2}{c^2}\epsilon_1''+k_1g_1\right)\right]\\ &=\frac{\omega_1^2\omega_2^2}{c^2}|A_3|^2\left[d_E'^2+\frac{2d_E'd_Q'\omega_P^2}{D(\omega_1)^*}+\frac{\omega_P^2}{D(\omega_1)^*}\left(\frac{c^2k_1^2}{\omega_1^2}-\epsilon_\infty+\frac{ic^2k_1g_1}{\omega_1^2}\right)d_Q'^2\right]\,,\end{aligned} \tag{11}$$

where

$$\epsilon_1=\epsilon_1'+i\epsilon_1''=\epsilon_\infty+\omega_P^2/D(\omega_1) \tag{12}$$

is the linear dielectric constant at ω_1. We now assume that the damping of a undriven (linear) wave at the idler frequency is large, so that k_1g_1 can be neglected compared to $(\omega_1^2/c^2)\epsilon_1''$. We also neglect the k_1g_1 term on the right side of Eq. (11) since this term is small compared to

$$|k_1^2-(\omega_1^2/c^2)\epsilon_\infty|$$

in the near-resonance region which we are considering. Having neglected g_1, we can solve Eq. (11) for g_2.

Neglecting any imaginary component of ϵ_2, the linear wave vector for the signal wave k_{2L} is defined by

$$k_{2L}^2\equiv(\omega_2^2/c^2)\epsilon_2\,. \tag{13}$$

For $k_2\approx k_{2L}$ we may write

$$k_2^2-(\omega_2^2/c^2)\epsilon_2=(k_2-k_{2L})(k_2+k_{2L})\approx 2\Delta k_2k_2\,, \tag{14}$$

where Δk_2 is the nonlinear change in wave vector of the signal wave. Using these approximations, we show in Appendix A that Eq. (11) can be rewritten to an excellent approximation as

$$(2\Delta k_2-ig_2)=\frac{\omega_2^2\omega_P^2}{k_2c^2}|A_3|^2\times\left(\frac{\omega_0^2-\omega_1^2}{\omega_P^2}d_E'+d_Q'\right)^2\Big/D(\omega_1)_{\text{PM}}^*\,. \tag{15}$$

The resonant denominator $D(\omega_1)_{\text{PM}}$ is given:

$$D(\omega_1)_{\text{PM}}=\left(\omega_0^2-\frac{\omega_P^2}{c^2k_1^2/\omega_1^2-\epsilon_\infty}-\omega_1^2\right)-i\omega_1\Gamma\,. \tag{16}$$

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[15] C. G. B. Garrett, IEEE J. Quantum Electron. **QE-4**, 70 (1968).

[16] R. Loudon, Advan. Phys. **13**, 423 (1964); **14**, (E)621 (1965).

The real part of $D(\omega_1)$ is zero when

$$k_1^2=\frac{\omega_1^2}{c^2}\left(\epsilon_\infty+\frac{\omega_P^2}{\omega_0^2-\omega_1^2}\right), \quad (17)$$

that is, when (ω_1,k_1) lie on the undamped polariton dispersion curve plotted in Fig. 1. This is the condition for the maximum gain, if the resonance is narrow. In this case, the maximum gain is given by

$$(g_2)_{\max}=\frac{\omega_2^2\omega_P^2|A_3|^2}{k_2c^2\omega_1\Gamma}\left(\frac{\omega_0^2-\omega_1^2}{\omega_P^2}d_E'+d_Q'\right)^2. \quad (18)$$

If the resonance is broad, the maximum gain is found by computing the imaginary part of Eq. (15). According to Eq. (15), $\Delta k_2=0$ when g_2 is a maximum, for a narrow resonance.

III. PARAMETRIC GAIN FROM TRANSITION-RATE THEORY

In conventional treatments of parametric amplification away from a resonance, the parametric gain is written as a function of the idler damping coefficient and of the wave-vector mismatch. We cast our solution into such a form in Appendix B. There we also derive an expression for the gain which holds both near and away from the lattice resonance.

Consider a volume V with linear dimensions small compared with $1/g_2$. Let $a_{k_3}{}^\dagger$ and a_{k}. be the creation and annihilation operators for the pump wave with wave vector k_3. Similarly, let $a_{k_2}{}^\dagger$, a_{k_2}, $a_{k_1}{}^\dagger$, and a_{k_1} be the creation and annihilation operators of the signal wave and the idler wave. These operators satisfy the commutation relations[17]

$$[a_{k_i},a_{k_j}{}^\dagger]=\delta_{k_ik_j}, \quad i, j=1, 2, 3. \quad (19)$$

The fields may be written as

$$\begin{aligned} E(\text{pump})&=E_3(\mathbf{r})=\sum_{k_3} E_1(a_{k_3}e^{i\mathbf{k}_3\cdot\mathbf{r}}+a_{k_3}{}^\dagger e^{-i\mathbf{k}_3\cdot\mathbf{r}}),\\ E(\text{signal})&=E_2(\mathbf{r})=\sum_{k_2} E_2(a_{k_2}e^{i\mathbf{k}_2\cdot\mathbf{r}}+a_{k_2}{}^\dagger e^{-i\mathbf{k}_2\cdot\mathbf{r}}),\\ E(\text{idler})&=E_1(\mathbf{r})=\sum_{k_1} E_1(a_{k_1}e^{i\mathbf{k}_1\cdot\mathbf{r}}+a_{k_1}{}^\dagger e^{-i\mathbf{k}_1\cdot\mathbf{r}}),\\ Q'(\text{lattice})&=Q'(\mathbf{r})=\sum_{k_1} Q_1'(a_{k_1}e^{i\mathbf{k}_1\cdot\mathbf{r}}+a_{k_1}{}^\dagger e^{-i\mathbf{k}_1\cdot\mathbf{r}}). \end{aligned} \quad (20)$$

Let n_{k_1}, n_{k_2}, and n_{k_3} be the mode occupation numbers where the nonzero matrix elements of the operators are[17]

$$\begin{aligned} \langle n_{k_i}-1|a_{k_i}|n_{k_i}\rangle&=n_{k_i}{}^{1/2},\\ \langle n_{k_i}+1|a_{k_i}{}^\dagger|n_{k_i}\rangle&=(n_{k_i}+1)^{1/2}. \end{aligned} \quad (21)$$

Just as in Sec. II, the three fields are assumed to be coupled by a phenomenological interaction Hamiltonian given by

$$\begin{aligned} \mathcal{H}&=-\int_v dx\left[\frac{d_E'}{4\pi}E_3(\mathbf{r})E_2(\mathbf{r})E_1(\mathbf{r})+\frac{d_Q'}{4\pi}E_3(\mathbf{r})E_2(\mathbf{r})Q'(\mathbf{r})\right]\\ &=-(1/4\pi)E_2E_3(d_Q'Q_1'+d_E'E_1)\\ &\quad\times V(a_{k_1}{}^\dagger a_{k_2}{}^\dagger a_{k_3}+a_{k_1}a_{k_2}a_{k_3}{}^\dagger), \end{aligned} \quad (22)$$

where $\mathbf{k}_1+\mathbf{k}_2=\mathbf{k}_3$. Following Loudon,[9] we can determine the constants E_i, $i=1, 2, 3$, from the requirement that for large values of n_{k_i} the expectation value of the Poynting vector S is given by

$$\begin{aligned}\langle n_i|S|n_i\rangle&=(c^2/4\pi v_{pi})\langle n_{ki}|E_i(x)E_i(x)|n_{ki}\rangle\\ &=\hbar\omega n_{ki}v_{gi}/V, \end{aligned} \quad (23)$$

where v_{pi} and v_{gi} are the phase and group velocities for the undamped ith wave.

We find

$$E_i^2=(2\pi\hbar\omega_1/V)(v_{gi}v_{pi}/c^2). \quad (24)$$

We want to find the net probability of a Stokes scattering in which a signal quantum and an idler quantum are created. We will assume that the system is initially in state $|n_{k_1}n_{k_2}n_{k_3}\rangle$. The rate is given by the Golden rule[18]

$$\begin{aligned} W=\frac{2\pi}{\hbar^2}[&|\langle n_{k_1}+1, n_{k_2}+1, n_{k_3}-1|\mathcal{H}|n_{k_1}, n_{k_2}, n_{k_3}\rangle|^2\\ &-|\langle n_{k_1}-1, n_{k_2}-1, n_{k_3}+1|\mathcal{H}|n_{k_1}, n_{k_2}, n_{k_3}\rangle|^2]\\ &\times\delta(\omega_2+\omega_1-\omega_3). \end{aligned} \quad (25)$$

If this transition rate becomes large compared with the lattice damping rate, the system will develop a coherent state composed of linear combinations of the states $|n_{k_1},n_{k_2},n_{k_3}\rangle$, $|n_{k_1}+1, n_{k_2}+1, n_{k_3}-1\rangle$, and $|n_{k_1}-1, n_{k_2}-1, n_{k_3}+1\rangle$. In this case, the initial state can no longer be described as $|n_{k_1},n_{k_2},n_{k_3}\rangle$ and the Raman gain cannot be computed using the Golden rule. We are restricting ourselves to the case where the lattice damping rate is large compared to W. Using Eq. (22), W becomes:

$$\begin{aligned} W=&(2\pi/\hbar^2)(E_{k_2}E_{k_3}V/4\pi)^2(d_q'Q_1'+d_EE_1)^2\\ &\times[(n_{k_1}+1)(n_{k_2}+1)n_{k_3}-n_{k_1}n_{k_2}(n_{k_3}+1)]\\ &\times\delta(\omega_3-\omega_2-\omega_1). \end{aligned} \quad (26)$$

For large values of n_{k_3}, this reduces to

$$\begin{aligned} W=&(2\pi/\hbar^2)(E_2V/4\pi)^2(d_q'Q_1'+d_E'E_1)^2\\ &\times\delta(\omega_3-\omega_1-\omega_2)(n_{k_1}+n_{k_2}+1). \end{aligned} \quad (27)$$

The mode-occupation number density

$$\rho_{k_2}=n_{k_2}/V \quad (28)$$

[17] W. Heitler, *Quantum Theory of Radiation* (Oxford University Press, London, 1954), Sec. 7.

[18] Reference 17, Sec. 14.

will satisfy the continuity equation

$$\frac{d\rho_{k_1}}{dt}=\frac{\partial\rho_{k_2}}{\partial t}+v_{g2}\frac{\partial\rho_{k_3}}{\partial x}=\frac{W}{V}. \tag{29}$$

In the steady state $\partial\rho_{k_2}/\partial t=0$,

$$\frac{\partial\rho_{k_2}}{\partial x}=\frac{W}{v_{g2}V}=\frac{1}{v_{g2}}\left(\frac{W}{n_{k_2}}\right)\rho_{k_2}\equiv g_2\rho_{k_2}. \tag{30}$$

Thus the parametric gain g_2 is given by $W/n_{k_2}v_{g2}$. For large n_{k_2}, this term is independent of n_{k_2} and is given by

$$g_2=\frac{2\pi}{\hbar^2}\left(\frac{E_2V}{4\pi}\right)^2\left(d_E'\frac{E_1}{Q_1'}+d_Q'\right)^2 Q_1'^2E_3^2n_{k_3}\times\delta(\omega_3-\omega_2-\omega_1)\frac{1}{v_{g2}}. \tag{31}$$

The final state damps out with rate Γ_1. In this case we must replace $\delta(\omega_3-\omega_2-\omega_1)$ by a normalized Lorenztian of width Γ_1 at half-maximum[19]:

$$\delta(\omega_3-\omega_2-\omega_1)\rightarrow\frac{\frac{1}{2}\Gamma_1/\pi}{(\omega_3-\omega_2-\omega_1)^2+(\frac{1}{2}\Gamma_1)^2}. \tag{32}$$

Making use of Eq. (24) to evaluate E_2^2 and using Eq. (31), we find the maximum value of the Raman gain to be

$$(g_2)_{\max}=\frac{\omega_2V}{2\pi}E_3^2n_{k_3}\frac{v_{P2}}{c^2}\left|d_E'\frac{E_1}{Q_1}+d_Q'\right|^2\frac{Q_1^2}{\Gamma_1}. \tag{33}$$

In Appendix C we show that

$$Q_1^2/\Gamma_1=2\pi\hbar\omega_P^2/\omega_1\Gamma V, \tag{34}$$

and in Appendix D we show that

$$E_1/Q_1=(\omega_0^2-\omega_1^2)/\omega_P^2. \tag{35}$$

Using the relations $v_{P2}=\omega_2/k_2$ and $E_3^2n_{k_3}=A_3^2$ (A_3 was defined in Sec. II), we find exactly the same expression for the parametric gain as was derived in Sec. II.

$$(g_2)_{\max}=\frac{A_3^2\omega_2\omega_P^2}{k_2c^2\omega_1\Gamma}\left(\frac{\omega_0^2-\omega_1^2}{\omega_P^2}d_E'+d_Q'\right)^2. \tag{18}$$

For a real solid, the Lorentz oscillator model description of the infrared resonance holds only near the resonance frequency ω_0. In order to take the infrared absorption properly into account, we must allow the damping constant and the resonance frequency ω_0 to be frequency-dependent.[20] It is this frequency-dependent value of Γ which must be substituted into Eq. (35). Generally, Γ will only vary by a factor of less than 10 over the near-resonance region of interest to us and the frequency dependence of ω_0 is negligible.

[19] Reference 17, Sec. 18.

[20] A. S. Barker (private communication).

IV. DISCUSSION

A. Numerical Computation of Parametric Gain

The spontaneous Stokes Raman-scattering cross section is given by W/NV, where N is the number of primitive cells per cm^3, multiplied by the density of states for photons with wave-vector $\mathbf{k}_2$ and divided by the incident-laser photon flux $S_3/\hbar\omega_3$. According to Eqs. (27) and (30)

$$\frac{d\sigma}{d\omega d\Omega}=\frac{g_2(n_{k_1}+1)k_2^2\hbar\omega_3}{NS_3(2\pi)^3}. \tag{36}$$

Equation (36), together with Eqs. (5) and (15), could be used to compute the Raman-scattering cross section for various scattering angles θ (defined in Fig. 1). Alternatively, we can measure the Raman-scattering cross section and use Eq. (36) to estimate g_2. At large angles, the Raman-scattering peaks at the lattice resonance frequency ω_0 and has a linewidth Γ. Therefore

$$(d\sigma/d\Omega d\omega)_{\max}=(d\sigma/d\Omega)\omega_0 2/\pi\Gamma. \tag{37}$$

Barker[20] has measured $N(d\sigma/d\Omega)\omega_0$ to be $(0.2\pm0.1)\times10^{-6}$ cm^{-1} sr^{-1} for GaP using a helium-neon 6328 Å laser. Evaluating Eq. (35) and using $\Gamma=4$ cm^{-1}, we find $g_2(\omega_0)_{\max}=1.24\times10^{-9}$ $cm^{-1}\times S_3$ cm^2, where S_3 is measured in W/cm^2. The frequency dependence of $g_2(\omega_1)_{\max}$ according to Eq. (17) is given by

$$g_2(\omega_1)_{\max}=g_2(\omega_0)_{\max}\left(\frac{\omega_0^2-\omega_1^2}{\omega_P^2}\frac{d_E'}{d_Q'}+1\right)^2\frac{\omega_0\Gamma}{\omega_1\Gamma(\omega_1)}. \tag{38}$$

The quantity $g_2(\omega_1)_{\max}/g_2(\omega_0)_{\max}$ is plotted in Fig. 2 for gallium phosphide assuming $\Gamma(\omega_1)=\Gamma$, using

$$(d_E'/d_Q')\omega_0^2/\omega_P^2=-1.89$$

as determined by Faust and Henry.[4] As shown in Fig. 2, the parametric gain $(g_2)_{\max}$ is slowly varying and exhibits virtually no variation very near resonance where α_1, the infrared absorption coefficient also plotted in Fig. 2, becomes resonantly large. The variation of $(g_2)_{\max}$ near resonance results primarily from the constructive and destructive interference of the two terms, proportional to d_E' and d_Q', which contribute to the nonlinear susceptibility. This interference causes $g_2(\omega_1)_{\max}$ to go to nearly zero at $\omega_1=250$ cm^{-1}. [The term neglected at the end of Appendix A keeps $(g_2)_{\max}$ from going exactly to zero.] As shown in Fig. 1, phase matching in gallium phosphide is possible only for ω_1 between 305 and 366 cm^{-1}. The relative gain curve $g_2(\omega_1)_{\max}$ in Fig. 2 is drawn as a solid line over this energy range.

B. Ratio of Infrared Light to Visible Light Generated

While the resonant behavior of the infrared absorption does not affect the parametric gain $(g_2)_{\max}$, it greatly influences the production of infrared light. We can easily calculate the ratio of infrared and visible light produced in parametric amplification. In each scattering event, an idler quantum as well as a signal quantum is produced, but the idler quanta are simultaneously damped out at rate $\Gamma_1=\alpha_1/v_{g1}$. The net rate of production of idler quanta will be

$$\dot{n}_1=W-\Gamma_1 n_1\,, \qquad (39)$$

where W is the transition rate which according to Eq. (30) is given by $g_2 v_{g2} n_2$. Equation (39) may be rewritten as

$$\dot{n}_1=g_2 v_{g2} n_2-\alpha_1 v_{g1} n_1\,. \qquad (40)$$

But we may also write $\dot{n}_1$ as

$$\dot{n}_1=g_1 v_{g1} n_1\,. \qquad (41)$$

Combining Eqs. (40) and (41) gives us the ratio of $v_{g1}n_1$ and $v_{g2}n_2$. Using this ratio, we find that the ratio of the ratio of Poynting vectors of the idler wave and signal waves is

$$\frac{S_1}{S_2}=\frac{n_1 v_{g1}\omega_1}{n_2 v_{g2}\omega_2}=\frac{g_2}{\alpha_1+g_1}\frac{\omega_1}{\omega_2}\approx\frac{g_2\omega_1}{\alpha_1\omega_2}\,. \qquad (42)$$

This result could have been derived starting with Eq. (7), but with much more labor.[21] As ω_1 approaches the lattice resonance frequency, ω_0, g_2 changes slowly, so that the signal flux that can be generated will remain roughly constant. On the other hand, α increases enormously near resonance, so that S_1/S_2 and consequently the amount of light generated at the idler frequency will be greatly reduced near resonance.

APPENDIX A: DERIVATION OF EQ. (15)

Neglecting g_1 and using Eqs. (12) and (14), we may rewrite the secular equation (11) as

$$(2\Delta k-ig_2)=\frac{\omega_1^2\omega_2^2|A_3|^2}{c^4k_2}\times\left[d_E'^2+\frac{2d_E'd_Q'\omega_P^2}{D(\omega_1)^*}+\frac{\omega_P^2}{D(\omega_1)^*}\left(\frac{c^2k_1^2}{\omega_1^2}-\epsilon_\infty\right)d_Q'^2\right]\Bigg/\left(k_1^2-\frac{\omega_1^2}{c^2}\epsilon_\infty-\frac{\omega_1^2}{c^2}\frac{\omega_P^2}{D(\omega_1)^*}\right)^{-1}. \qquad (A1)$$

The first bracketed term in the denominator on the right side of Eq. (A1) can be rewritten as

$$k_1^2-\frac{\omega_1^2}{c^2}\epsilon_1^*=\frac{(k_1^2-(\omega_1^2/c^2)\epsilon_\infty)}{D(\omega_1)^*}D(\omega_1)_{\rm PM}^*\,, \qquad (A2)$$

where the resonant denominator $D(\omega_1)_{\rm PM}$ is defined by

$$D(\omega_1)_{\rm PM}=\left[\omega_0^2-\frac{\omega_P^2}{((c^2/\omega_1^2)k_1^2-\epsilon_\infty)}-\omega_1^2\right]-i\omega_1\Gamma\,. \qquad (A3)$$

$[D(\omega_1)_{\rm PM}]^{-1}$ is resonant when (ω_1,k_1) lie on the undamped polariton dispersion curve shown in Fig. 1. Using Eq. (A2), Eq. (A1) becomes

$$(2\Delta k-ig_2)=\frac{\omega_2^2|A_3|^2\omega_P^2}{k_2c^2D(\omega_1)_{\rm PM}^*}\left[\frac{D(\omega_1)^*d_E'^2}{((c^2/\omega_1^2)k_1^2-\epsilon_\infty)\omega_P^2}+\frac{2d_E'd_Q'}{((c^2/\omega_1^2)k_1^2-\epsilon_\infty)}+d_Q'^2\right]. \qquad (A4)$$

We expect d_Q' and d_E' to be of the same order of magnitude. For example, Faust and Henry[4] found

$$d_Q'/d_E'=-0.28 \qquad (A5)$$

for gallium phosphide. As long as d_Q' is not negligible compared with d_E', the square-bracketed term on the right side of (A4) will be slowly varying. The resonant behavior of g_2 will be determined by the resonant denominator $D(\omega_1)_{\rm PM}^*$. The maximum value of g_2 occurs at resonance when (ω_1,k_1) lie on the undamped polariton dispersion curve and

$$c^2k_1^2/\omega_1^2-\epsilon_\infty=\omega_P^2/(\omega_0^2-\omega_1^2)\,. \qquad (A6)$$

Rewriting the square-bracketed term on the right side of (A4) and using Eq. (A6) [which is a good approximation for (ω_1,k_1) near the undamped polariton dispersion curve], we get

$$\left\{\frac{D(\omega_1)^*d_E'^2}{\omega_P^2[(c^2/\omega_1^2)k_1^2-\epsilon_\infty]}+\frac{2d_E'd_Q'}{[(c^2/\omega_1^2)k_1^2-\epsilon_\infty]}+d_Q'^2\right\}\approx\left(d_E'\frac{\omega_0^2-\omega_1^2}{\omega_P^2}+d_Q'\right)^2-\frac{i\omega_1\Gamma}{\omega_P^2}\left(\frac{\omega_0^2-\omega_1^2}{\omega_P^2}\right)d_E'^2. \qquad (A7)$$

The contribution of the second term on the right side of Eq. (A7) to g_2 is negligible except when the first term goes to zero as a result of cancellation of the two terms within the braces. Even in this case, the contribution of the second term will be small. We will neglect this term. Substituting Eq. (A7) into Eq. (A4) gives Eq. (15).

APPENDIX B: PARAMETRIC GAIN AWAY FROM THE LATTICE RESONANCE

The argument given in Sec. II and in Appendix A is adequate where one of the interacting fields is close

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[21] By starting with the last two equations of Eq. (7), eliminating $Q_1'^*$, neglecting γ, and solving for A_1^*/A_2, one can show that

$$S_1/S_2=(\omega_1/\omega_2)g_2/2k_1^{\dagger\prime\prime},$$

where $k_1^{\dagger\prime\prime}$ is defined in Appendix B. This is, for all practical purposes, the same result as Eq. (42).

to the lattice resonance and the other is at a frequency at which the optical loss is negligible. It is easy, however, to generalize the result [given above as Eq. (14)] to the case where both signal and idler are far from resonance. The work should then reproduce standard results on nonresonant parametric amplification given, for example, by Bloembergen.[22]

To show this, we restrict ourselves to the case of forward scattering, where the pump, signal, and idler waves are all propagating in the same direction. In this case,

$$g_1 = g_2 = g. \tag{B1}$$

We allow ϵ_2 to be complex. Then the complex linear wave vector of the signal wave is given by

$$(k_{20}' + ik_{20}'') = (\omega_2^2/c^2)\epsilon_2. \tag{B2}$$

Starting with the secular equation [Eq. (11)] and proceeding as in Appendix A, it may be shown after some algebra that to an excellent approximation the secular equation can be rewritten as

$$[\Delta k_2 - i(\tfrac{1}{2}g + k_{20}'')][\Delta k_1 + i(\tfrac{1}{2}g + k_1^{\dagger\prime\prime})] - \alpha_P^2 = 0, \tag{B3}$$

where

$$\Delta k_1 = k_1 - k_1^{\dagger\prime}, \tag{B4}$$

$$\Delta k_2 = k_2 - k_{20}, \tag{B5}$$

$$k_1^{\dagger\prime} \equiv \frac{\omega_1^2}{c^2}\left(\epsilon_\infty + \frac{\omega_P^2}{\omega_0^2 - \omega_1^2}\right), \tag{B6}$$

$$k_1^{\dagger\prime} k_1^{\dagger\prime\prime} = +\frac{\omega_1\Gamma}{2}\frac{\omega_P^2}{(\omega_0^2 - \omega_1^2)^2}\frac{\omega_1^2}{c^2}, \tag{B7}$$

$$\alpha_P^2 \equiv \frac{\omega_1^2\omega_2^2|A_3|^2}{4c^4 k_1^{\dagger\prime} k_{20}'}\left(d_E' + \frac{\omega_P^2}{\omega_0^2 - \omega_1^2} d_Q'\right)^2. \tag{B8}$$

Here $k_1^{\dagger\prime}$ is the *real* part of the propagation constant for the *undamped* polariton curve and $k_1^{\dagger\prime\prime}$ has the frequency dependence of the imaginary part of the undamped polariton curve in the limit $\omega_0\Gamma \to 0$. The quantities Δk_1 and Δk_2 are phase-mismatch parameters. $2\alpha_P$ is the parametric gain in the absence of losses or wave-vector mismatch. Since g is real, we have, from the imaginary part of Eq. (B8),

$$\Delta k_1(\tfrac{1}{2}g + k_{20}'') = \Delta k_2(\tfrac{1}{2}g + k_1^{\dagger\prime\prime}), \tag{B9}$$

which, together with

$$\Delta k = \Delta k_1 + \Delta k_2 = k_3 - k_1^{\dagger\prime} - k_{20}', \tag{B10}$$

determines both Δk_1 and Δk_2.

We now quote the consequences of Eq. (B3):

1. Threshold: Setting $g = 0$ and using Eq. (B9), we get from Eq. (B3)

$$\alpha_P^2 = \{[(\Delta k_1)^2 + (k_1^{\dagger\prime\prime})^2]/k_1^{\dagger\prime\prime}\}k_{20}'', \tag{B11}$$

which is completely general, independent of whether or not ω_1 is close to ω_0.

2. Gain near resonance: Setting $k_1^{\dagger\prime\prime} \gg g$, k_{20}'', we get

$$g = -2k_{20}'' + 2\alpha_P^2 k_1^{\dagger\prime\prime}/[(\Delta k_1)^2 + (k_1^{\dagger\prime\prime})^2]. \tag{B12}$$

The second term on the right side of Eq. (B12) can be shown to be the same as our previous expression for the parametric gain near resonance given by the imaginary part of Eq. (15).

3. Gain far from resonance: If α_P, $k_1^{\dagger\prime\prime}$, and k_{20}'' are all comparable, Eq. (B3) gives

$$g = -(k_1^{\dagger\prime\prime} + k_{20}'') \pm \mathrm{Re}[(k_1^{\dagger\prime\prime} - k_2'' - i\Delta k)^2 + 4\alpha_P^2]^{1/2}, \tag{B13}$$

which has the same form as the conventional expression for the parametric gain discussed by Bloembergen.[22]

APPENDIX C: EVALUATION OF $Q_1'^2/\Gamma_1$

An energy density $\mathcal{E}$ for the idler wave will damp out at rate Γ_1. That is,

$$\dot{\mathcal{E}} = -\Gamma_1\mathcal{E}. \tag{C1}$$

This damping is produced by the dissipative force of the oscillator and is given by

$$\dot{\mathcal{E}} = -N\mu\Gamma\langle\dot{Q}^2\rangle_{\mathrm{av}} = -\frac{\mu\Gamma\omega_1^2\langle Q'^2\rangle_{\mathrm{av}}}{(4\pi)^2 Ne^2} = -\frac{\Gamma\omega_1^2}{2\pi\omega_P^2}Q_1'^2(n_{k_1} + \tfrac{1}{2}). \tag{C2}$$

Combining (C1) and (C2), and using $\epsilon = \hbar\omega_1/v(n_{k_1} + \frac{1}{2})$, we find

$$\frac{Q_1'^2}{\Gamma_1} = \frac{2\pi\omega_P^2}{\Gamma\omega_1^2}\frac{\mathcal{E}}{(n_{k_1} + \frac{1}{2})} = \frac{2\pi\hbar\omega_P^2}{\Gamma\omega_1 V}. \tag{C3}$$

APPENDIX D: EVALUATION OF Q_1'/E_1

In our notation, the linear force equation for the lattice wave is given by

$$\ddot{Q}_1' + \Gamma\dot{Q}_1' + \omega_0^2 Q_1' = \omega_P^2 E_1. \tag{D1}$$

For a wave at angular frequency ω_1, according to Eq. (D1),

$$Q_1'/E_1 = \omega_P^2/(\omega_0^2 - \omega_1^2 - i\omega_1\Gamma). \tag{D2}$$

In evaluating $[d_E'(E_1/Q_1') + d_Q']$, damping may be neglected unless d_Q' is very small compared to d_E'. Neglecting the damping, we find

$$d_E'\frac{E_1}{Q_1'} + d_Q' = \left[d_E'\frac{\omega_P^2}{\omega_0^2 - \omega_1^2} + d_Q'\right]. \tag{D3}$$

[22] Reference 5, p. 99, Eq. (4–58).