for various values of θ and α. Because of the complexity of the spectrum, since each $\Delta m=0$ transition consists of five fine-structure lines, it is frequently impossible to measure all the $\Delta m=\pm 1$ transitions. Thus, in Table I, where a measurement is given for a particular value of m, this is the only value for which the corresponding doublet of lines was measurable; otherwise, the symbols $\bar{P}$ and $\bar{Q}$ denote an average over several doublets. It should be noted that $Q(m)=0$ for $\theta=0$, $\pm\frac{1}{4}\pi$, while $P(m)=0$ for $\tan\alpha \sin 4\theta=0.87$.

III. DISCUSSION

It can be seen that there is very satisfactory agreement between theory and experiment, considering the experimental difficulties of accurate alignment of the crystal and of measuring lines of low intensity. It is interesting to point out that the intensities of forbidden lines can be much increased by the presence of local deviations from cubic symmetry; this effect is spectacularly demonstrated in powder samples of MgO [15] and CaO, where the forbidden lines are an order of magnitude larger relative to the $\Delta M=1$, $\Delta m=0$ lines than in single MgO crystals. Such effects can also arise in single crystals; if we assume that the effect of random distortions can be represented by second-rank spin tensors (of the form $S_z^2-\frac{1}{3}S^2$, etc.), it can be shown that the averaged effect in a single crystal gives an additive contribution

$$I'=(F+G\cos 4\theta)[(H_1^{\perp})^2/g^2\beta^2H^2][I(I+1)-m(m\pm 1)]$$

to the forbidden line intensities of Eq. (12) (F and G are constants describing the average strength and angular distribution of the local distortions, where $F>|G|\geq 0$). However, we find that it does not appear necessary to include such terms in the analysis of our experimental results on single crystals.

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Quantum Theory of Spontaneous Parametric Scattering of Intense Light*

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A quantum theory is developed for the spontaneous optical parametric process in nonlinear, anisotropic, and dispersive media; the process is treated directly as a scattering problem rather than a problem involving equivalent coupled oscillators. Following the general procedure in quantum field theory, a general expression is obtained for the differential extinction coefficient, which contains all the information on the intensity of the spontaneous emission in the parametric scattering process. The tuning characteristics, beam divergence, and spectral properties of the spontaneous emission for colinear and noncolinear interactions are described in detail and illustrated with numerical examples on such standard nonlinear optical crystals as $LiNbO_3$, $NH_4H_2PO_4$, and KH_2PO_4 under various pumping conditions. Explicit formulas for the intensity of the spontaneous emission are also obtained. The spontaneous intensity calculated using the formulas given is shown to be in complete agreement with known experimental data on $NH_4H_2PO_4$ pumped at 3472 Å.

I. INTRODUCTION

PARAMETRIC interaction of light waves in optically transparent nonlinear media is an effective method for producing continuously tunable amplification and generation of coherent optical radiation throughout the visible and into the infrared region of the spectrum.[1] Since the initial observation[2] of the parametric effect in KH_2PO_4 (KDP), significant progress has been made in developing such amplifiers and oscillators.[3–5] The theory of optical parametric process has also been discussed extensively in the past.[6–9] The calculations dealt mainly with the gain

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[1] See, for example, the review article by S. A. Akhmanov and R. V. Khohklov, Usp. Fiz. Nauk. **88**, 437 (1966) [English transl.: Soviet Phys.—Usp. **9**, 210 (1966)]. *Note added in proof.* After our paper was submitted, a letter by D. N. Klyshko, Zh. Eksperim. i Teor. Fiz. Pis'ma v Redaktsiyu **6**, 490 (1967) [English transl.: Soviet Phys.—JETP Letters **6**, 23 (1967)], which considers the same subject, became available in English translation.

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[9] B. R. Mollow and R. Glauber, Phys. Rev. **160**, 1076 (1967)

mechanism and the behavior of the single-mode oscillators; the quantum noise of the single-mode parametric amplifier due to the zero-point fluctuations has also received considerable attention.[7,9]

The study of the quantum noise in the parametric process is important not only because the ultimate sensitivity of the optical parametric amplifier is limited by such noise but also for a more immediate reason: With all the presently available nonlinear crystals, sufficiently high parametric gain can be achieved only when pumped with the intense outputs of pulsed solid-state lasers or their harmonics. Within the limited duration of the pump light, the parametric oscillator does not always achieve steady-state oscillation[3] and the oscillator output consists primarily of amplified noise; it is proportional to the average intensity of the quantum noise. When pumped with the continuous-wave gas lasers,[10] there is hardly any gain and one can only observe the noise.

In Ref. 3, the average intensity of the quantum noise produced in $LiNbO_3$ pumped at 5300 Å was inferred from the measured oscillator output by Miller and Nordland. Madge and Mahr[5] have made extensive direct measurement of the average intensity of the quantum noise in $NH_4H_2PO_4$ (ADP) pumped at 3472 Å. It is known that the quantum noise intensity per mode[7] is equal to the usual zero-point energy of harmonic oscillators. One can then make a rough estimate of the expected total intensity of the quantum noise from this single-mode result and the estimated flux of modes that can be parametrically amplified. Indeed it was found[5] that the measured result in ADP agrees reasonably well with such an estimate. On the other hand, there are indications[3] that the measured result in $LiNbO_3$ was far less than that estimated on this basis.

The comparison of theory and experiment is complicated by the fact that the extension of the single-mode results on quantum fluctuations, which are quite adequate for the cavity-type microwave amplifiers, to the optical parametric process is far from trivial. In this case, the nonlinear crystal is anisotropic and dispersive and the pumping laser beam always has a finite spectral width and is rarely diffraction limited; there are no clearly isolated modes. Whole regions, rather than isolated points, in the wave-vector space of the electromagnetic fields must be considered in the interaction process. The optical parametric process should then be treated directly as a scattering problem rather than a problem involving equivalent coupled oscillators. The quantum noise is the spontaneous emission in the parametric scattering of the incident pumping light in the nonlinear crystal. We have carried out the calculations this way.

Following the general procedure in quantum field theory,[11] we obtain in the next section an expression for the differential extinction coefficient for the parametric scattering process. This coefficient is defined as the ratio of the number of photons spontaneously emitted into a given differential solid angle and frequency interval due to the parametric scattering process per unit time per unit volume to the incident-pump photon-flux density; it contains all the information on the intensity of the spontaneous emission. In Sec. III, explicit formulas are obtained and the properties of the spontaneous emission described. Finally, detailed numerical results are given for various representative experimental situations and comparisons are made with known experimental results.

II. DERIVATION OF THE DIFFERENTIAL EXTINCTION COEFFICIENT

A. Field Quantization and the Interaction Lagrangian

The total Lagrangian density $L_{\rm tot}$ of the fields in the nonlinear medium can be split up,

$$L_{\rm tot}=L_0+L_1, \tag{1}$$

into a part L_0 for the free fields in the medium in the absence of the nonlinearity and a part L_1 that describes the interaction due to the nonlinearity.

Consider first the part independent of the nonlinearity[12]:

$$L_0=(1/8\pi)\,[\mathbf{D}\cdot\mathbf{E}-(1/\mu_o)\mathbf{B}\cdot\mathbf{B}], \tag{2}$$

and the corresponding Hamiltonian

$$H_0=\frac{1}{8\pi}\int(\mathbf{D}\cdot\mathbf{E}+\mu_o^{-1}\mathbf{B}\cdot\mathbf{B})d\mathbf{r}, \tag{3}$$

where μ_o is the permeability of the medium and the **D** vector is related to the **E** vector by a linear dielectric tensor.

For anisotropic crystals, the formal problem is slightly complicated by the fact that for extraordinary waves the wave vector **k** and the Poynting vector **s** are not always in the same direction since **D** and **E** are not always colinear. However, for all the crystals of interest (e.g., $LiNbO_3$, ADP, KDP, etc.) this difference is extremely small. As a simplification, we assume in what follows that the $(\hat{k}\cdot\mathbf{E})^2$ term is negligible compared with the $\mathbf{E}\cdot\mathbf{E}$ term in electric energy density[13] for the extraordinary waves. In a dispersive medium,[13] the electric energy density for a spectral component of frequency ω would contain an additional term of the form $(\omega/2)[d\epsilon(\omega)/d\omega]\mathbf{E}\cdot\mathbf{E}$, where $\epsilon(\omega)$ refers to the dielectric constant at this frequency. Again, in all the crystals and spectral ranges of interest,

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[11] N. N. Bogoliubov and D. V. Shirkov, *Introduction to the Theory of Quantized Fields* (Interscience Publishers, Inc. New York, 1959).

[12] cgs electrostatic units are used throughout this paper.

[13] See, for example, L. Landau and E. Lifshitz, *Electrodynamics of Continuous Media* (Addison-Wesley Publishing Co., Reading, Mass., 1960), p. 316 and Sec. 61.

this term is generally small compared with the remaining terms in the energy density. As an additional simplification, in what follows, we also neglect this term. The main effects of the anisotropy and dispersiveness of the medium on the optical parametric process are not dependent on these neglected terms and these approximations would have no effect on our final results.

We now expand the electric field in the medium in plane extraordinary and ordinary waves normalized in the continuous spectrum:

$$\mathbf{E}(\mathbf{r},t)=\frac{i}{2\pi}\int[n_o(\mathbf{k})]^{-1}(\hbar\omega_o)^{1/2}\times[a_o(\mathbf{k})\exp(i\mathbf{k}\cdot\mathbf{r}-i\omega_o t)-a_o^\dagger(\mathbf{k})\times\exp(-i\mathbf{k}\cdot\mathbf{r}+i\omega_o t)]\hat{o}(\hat{k})d\mathbf{k}+\frac{i}{2\pi}\int[n_e(\mathbf{k})]^{-1}(\hbar\omega_e)^{1/2}[a_e(\mathbf{k})\exp(i\mathbf{k}\cdot\mathbf{r}-i\omega_e t)-a_e^\dagger(\mathbf{k})\exp(-i\mathbf{k}\cdot\mathbf{r}+i\omega_e t)]\hat{e}(\hat{k})d\mathbf{k},\quad(4)$$

where $a_e^\dagger(\mathbf{k})$ and $a_e(\mathbf{k})$ are the creation and annihilation operators for a quantum of extraordinary wave with the wave vector $\mathbf{k}$ and polarization $\hat{e}(\hat{k})$; similarly, $a_o^\dagger(\mathbf{k})$, $a_o(\mathbf{k})$, and $\hat{o}(\hat{k})$ refer to the ordinary wave. According to the usual rules of field quantization,[11] the creation and annihilation operators satisfy the commutation rules:

$$[a(\mathbf{k}),a(\mathbf{k}')]=[a^\dagger(\mathbf{k}),a^\dagger(\mathbf{k}')]=0\quad\text{for any}\quad a,\quad(5)$$

$$[a_o(\mathbf{k}),a_o^\dagger(\mathbf{k}')]=\delta(\mathbf{k}-\mathbf{k}'),\quad(6)$$

$$[a_e(\mathbf{k}),a_e^\dagger(\mathbf{k}')]=\delta(\mathbf{k}-\mathbf{k}'),\quad(7)$$

$$[a_e(\mathbf{k}),a_o^\dagger(\mathbf{k}')]=0.\quad(8)$$

The angular frequencies ω_e and ω_o are related to the magnitude of the corresponding $\mathbf{k}$ vector and the indices of refraction, $n_e(\mathbf{k})$ and $n_o(\mathbf{k})$, for the extraordinary and ordinary waves, respectively, as follows:

$$\omega_e n_e(\mathbf{k})-|\mathbf{k}|C=0,\quad(9)$$

$$\omega_o n_o(\mathbf{k})-|\mathbf{k}|C=0,\quad(10)$$

where C is the speed of light in free space. Using this representation, Eq. (4), the interaction-free part of the Hamiltonian, Eq. (3), becomes simply:

$$H_0=\frac{1}{2}\int\hbar\omega_o[a_o^\dagger(\mathbf{k})a_o(\mathbf{k})+a_o(\mathbf{k})a_o^\dagger(\mathbf{k})]d\mathbf{k}+\frac{1}{2}\int\hbar\omega_e[a_e^\dagger(\mathbf{k})a_e(\mathbf{k})+a_e(\mathbf{k})a_e^\dagger(\mathbf{k})]d\mathbf{k}.\quad(11)$$

The interaction Lagrangian L_1 is determined by the following considerations: In a nonlinear optical crystal,[14] a component of the electric displacement vector at a given frequency may contain an additional term $D_i^{\mathrm{NL}}(\omega_1)$ corresponding to the nonlinear polarization $P_i^{\mathrm{NL}}(\omega_1)$ which is related to the electric field components at two other frequencies, $E_j(\omega_2)$ and $E_k(\omega_3)$, through a nonlinear susceptibility coefficient[14]

$$D_i^{\mathrm{NL}}(\omega_1)=4\pi P_i^{\mathrm{NL}}(\omega_1)=4\pi\chi_{ijk}^{\mathrm{NL}}(\omega_1\omega_2\omega_3)E_j(\omega_2)E_k(\omega_3).\quad(12)$$

The interaction Lagrangian would have to be consistent with this, since in general[15]

$$D_i=(4\pi)^{-1}(\partial L/\partial E_i)\quad(13)$$

and within the context of the present problem we have

$$D_i^{\mathrm{NL}}(\omega_1)=(4\pi)^{-1}[\partial L_1/\partial E_i(\omega_1)],\text{ etc.}\quad(14)$$

Furthermore, in considering the parametric process which corresponds to the simultaneous annihilation of a pump photon with wave vector $\mathbf{k}_p$ and creation of two other photons and the reverse process, one needs to consider only those terms in the interaction Lagrangian that are proportional to $a(\mathbf{k}_p)a^\dagger(\mathbf{k}_1)a^\dagger(\mathbf{k}_2)$ and $a^\dagger(\mathbf{k}_p)a(\mathbf{k}_1)a(\mathbf{k}_2)$. Finally, one can assume, for definitness, that the pump wave is an extraordinary wave while the other two are ordinary waves; this corresponds to the usual experimental arrangement in $LiNbO_3$, ADP, KDP, etc. The final results can be easily modified to apply to the other situations when required. Thus, one finds that for the present purpose the interaction Lagrangian density L_1 should have the following form:

$$L_1=-\frac{i}{(2\pi)^3}\iiint\sum_{ijk}\frac{\chi_{ijk}^{\mathrm{NL}}e_i(\hat{k}_p)o_j(\hat{k}_1)o_k(\hat{k}_2)}{n_e(\mathbf{k}_p)n_o(\mathbf{k}_1)n_o(\mathbf{k}_2)}\times(\hbar^3\omega_{ep}\omega_{o1}\omega_{o2})^{1/2}\{a_e(\mathbf{k}_p)a_o^\dagger(\mathbf{k}_1)a_o^\dagger(\mathbf{k}_2)\exp[i(\mathbf{k}_p-\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{r}-i(\omega_{ep}-\omega_{o1}-\omega_{o2})t]-a_e^\dagger(\mathbf{k}_p)a_o(\mathbf{k}_1)a_o(\mathbf{k}_2)\exp[-i(\mathbf{k}_p-\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{r}+i(\omega_{ep}-\omega_{o1}-\omega_{o2})t]\}d\mathbf{k}_p d\mathbf{k}_1 d\mathbf{k}_2\quad(15)$$

in terms of the representation given in Eq. (4). $e_i(\hat{k}_p)$, $o_j(\hat{k}_1)$, and $o_k(\hat{k}_2)$ are the projections of the unit vectors $\hat{e}(\hat{k}_p)$, $\hat{o}(\hat{k}_1)$, and $\hat{o}(\hat{k}_2)$ on the unit vectors $\hat{\imath}$, $\hat{\jmath}$, $\hat{k}$, respectively, referred to in the subscripts of the nonlinear susceptibility coefficient.

With the interaction Lagrangian given, the parametric process in the nonlinear optical medium can now be described in terms of an appropriate scattering matrix and studied by means of the usual perturbation theory.[11]

[14] P. A. Franken and J. F. Ward, Rev. Mod. Phys. **35,** 23 (1963); N. Bloembergen, *Nonlinear Optics* (W. A. Benjamin, Inc., New York, 1965); D. A. Kleinman, Phys. Rev. **126,** 1977 (1962).

[15] See, for example, A. I. Akhiezer and V. Berestetsky, *Quantum Electrodynamics* (Consultants Bureau Enterprises, Inc., New York, 1953), Part II, Sec. 47, p. 491.

B. S Matrix and the Differential Extinction Coefficient

In considering the spontaneous emission in the parametric scattering in the nonlinear optical medium, only the lowest-order term $S^{(1)}$ in the perturbation expansion of the S matrix[16] is needed:

$$S^{(1)}=\frac{i}{\hbar}\iint L_1 d\mathbf{r}dt$$

$$=\hbar^{-1}\iiint\frac{\Delta\epsilon_1(\hbar^3\omega_{ep}\omega_{o1}\omega_{o2})^{1/2}}{2n_e(\mathbf{k}_p)n_o(\mathbf{k}_1)n_o(\mathbf{k}_2)}$$

$$\times[a_e(\mathbf{k}_p)a_o^\dagger(\mathbf{k}_1)a_o^\dagger(\mathbf{k}_2)-a_e^\dagger(\mathbf{k}_p)a_o(\mathbf{k}_1)a_o(\mathbf{k}_2)]$$

$$\times\delta(\mathbf{k}_p-\mathbf{k}_1-\mathbf{k}_2)\delta(\omega_{ep}-\omega_{o1}-\omega_{o2})d\mathbf{k}_pd\mathbf{k}_1d\mathbf{k}_2, \quad (16)$$

which is obtained from Eq. (15) and after carrying out the integrations in $\mathbf{r}$ and t, where

$$\Delta\epsilon_1\equiv 4\pi\sum_{ijk}\chi_{ijk}^{\mathrm{NL}}e_i(\hat{k}_p)o_j(\hat{k}_1)o_k(\hat{k}_2). \quad (17)$$

We must next specify the types of initial states that are of interest when calculating the spontaneous emission. Physically, in the absence of the nonlinear interaction, now characterized by $S^{(1)}$, the only field present should be that of the pump beam. In order to study the influence of the coherence property of the pump wave, two representative situations are, however, of interest: a coherent pump wave represented by a Glauber state[17] or an incoherent pump wave represented by a fixed-number state.

The initial state representing an incoherent pump wave with precisely N_p pump photons per unit volume in the medium is

$$\phi_{\mathrm{incoh}}=[(2\pi)^{3/2}/(N_p!)^{1/2}][a_e^\dagger(\mathbf{k}_p)]^{N_p}\phi_0, \quad (18)$$

where ϕ_0 is the ground state. The coherent state is constructed from a superposition of fixed-number states with a Poisson distribution:

$$\phi_{\mathrm{coh}}=(2\pi)^{3/2}\sum_{N=0,1\ldots}^{\infty}\frac{\alpha_p^N\exp(-\frac{1}{2}|\alpha_p|^2)}{(N!)^{1/2}}[a_e^\dagger(\mathbf{k}_p)]^N\phi_0, \quad (19)$$

where $|\alpha_p|^2$ is equal to the *average number* of pump photons per unit volume in the nonlinear medium $\bar{N}_p$.

In the presence of the nonlinearity, the parametric process characterized by $S^{(1)}$ will lead to the creation of photons at lower frequencies from the pump photons. The probability for such transitions depends upon the matrix elements

$$\phi_{\mathrm{final}}^\dagger S^{(1)}\phi_{\mathrm{incoh}} \quad (20a)$$

or

$$\phi_{\mathrm{final}}'^\dagger S^{(1)}\phi_{\mathrm{coh}}, \quad (20b)$$

depending on whether an incoherent or a coherent pump is used, where ϕ_{final} and ϕ_{final}' are the corresponding final states.

When an incoherent pump is used, the final state will contain one less pump photon, but there will be one photon each in some suitable pair of momentum states $\mathbf{k}_1$ and $\mathbf{k}_2$, or

$$\phi^\dagger_{\mathrm{final}}=\phi^\dagger_0 a_o(\mathbf{k}_1)a_o(\mathbf{k}_2)\{[a_e(\mathbf{k}_p)]^{N_p-1}/[(N_p-1)!]^{1/2}\}, \quad (21)$$

which is normalized to unity: $|\phi_{\mathrm{final}}|^2=1$. The matrix element of interest, Eq. (20a), then becomes

$$\phi_{\mathrm{final}}^\dagger S^{(1)}\phi_{\mathrm{incoh}}=\phi_0^\dagger a_o(\mathbf{k}_1)a_o(\mathbf{k}_2)\frac{[a_e(\mathbf{k}_p)]^{N_p-1}}{[(N_p-1)!]^{1/2}}\Bigg(\hbar^{-1}\iiint\frac{\Delta\epsilon_1(\hbar^3\omega_{ep}'\omega_{o1}'\omega_{o2}')^{1/2}}{2n_e(\mathbf{k}_p')n_o(\mathbf{k}_1')n_o(\mathbf{k}_2')}$$

$$\times[a_e(\mathbf{k}_p')a_o^\dagger(\mathbf{k}_1')a_o^\dagger(\mathbf{k}_2')-a_e^\dagger(\mathbf{k}_p')a_o(\mathbf{k}_1')a_o(\mathbf{k}_2')]\delta(k_p'-k_1'-k_2')\delta(\omega_{ep}'-\omega_{o1}'-\omega_{o2}')dk_p'dk_1'dk_2'\Bigg)$$

$$\times\frac{(2\pi)^{3/2}}{(N_p!)^{1/2}}[a_e^\dagger(\mathbf{k}_p)]^{N_p}\phi_0=\frac{(2\pi)^{3/2}\Delta\epsilon_1(\hbar\omega_{ep}\omega_{o1}\omega_{o2})^{1/2}(N_p)^{1/2}}{n_e(\mathbf{k}_p)n_o(\mathbf{k}_1)n_o(\mathbf{k}_2)}\delta(\mathbf{k}_p-\mathbf{k}_1-\mathbf{k}_2)\delta(\omega_{ep}-\omega_{o1}-\omega_{o2}). \quad (22)$$

The corresponding transition probability is equal, in accordance with the general rule of quantum mechanics, to

$$\frac{|\phi_{\mathrm{final}}^\dagger S^{(1)}\phi_{\mathrm{incoh}}|^2}{VT}=\frac{(\Delta\epsilon_1)^2N_p\hbar\omega_{ep}\omega_{o1}\omega_{o2}}{(2\pi)n_e^2(\mathbf{k}_p)n_o^2(\mathbf{k}_1)n_o^2(\mathbf{k}_2)}\delta(\mathbf{k}_p-\mathbf{k}_1-\mathbf{k}_2)\delta(\omega_{ep}-\omega_{o1}-\omega_{o2}) \quad (23a)$$

from Eq. (22) and making use of certain results[18] given in Ref. 11. When a coherent pump is used, the final state is

[16] Reference 11, Sec. 18.
[17] R. J. Glauber, Phys. Rev. **131**, 2766 (1963).
[18] Reference 11, Eqs. (21.35) and (21.37).

of course no longer a particular fixed-number state and the transition probability now becomes

$$\frac{|\phi_{\text{final}}'^{\dagger}S^{(1)}\phi_{\text{coh}}|^2}{VT}=(VT)^{-1}\sum_N\left|\frac{(2\pi)^{3/2}\alpha_p^N\exp(-\frac{1}{2}|\alpha_p|^2)}{[N!(N-1)!]^{1/2}}\phi_0^{\dagger}a_o(\mathbf{k}_1)a_o(\mathbf{k}_2)[a_e(\mathbf{k}_p)]^{N-1}S^{(1)}[a_e^{\dagger}(\mathbf{k}_p)]^N\phi_0\right|^2$$

$$=\frac{(\Delta\epsilon_1)^2\bar{N}_p\hbar\omega_{ep}\omega_{o1}\omega_{o2}}{(2\pi)n_e^2(\mathbf{k}_p)n_o^2(\mathbf{k}_1)n_o^2(\mathbf{k}_2)}\delta(\mathbf{k}_p-\mathbf{k}_1-\mathbf{k}_2)\delta(\omega_{ep}-\omega_{o1}-\omega_{o2}) \quad (23b)$$

from Eqs. (20b) and (19) and with the help of the results in Ref. 11. The only difference between this result and Eq. (23a) is that here the average photon number $\bar{N}_p$ appears in place of N_p. Thus, when normalized to the intensity of the pump beam, the properties of the *intensity* of the spontaneous emission in the parametric scattering process are independent of the coherence property of the pump beam. From now on, only one of these cases needs to be considered.

We are now in a position to calculate the differential extinction coefficient $d\sigma(\mathbf{k}_s)$ for the spontaneous emission in the parametric scattering process in the nonlinear medium. It is defined here as the ratio of the number of photons spontaneously emitted into the solid angle $d\Omega_s$ and the frequency interval $d\omega_{os}$ per unit time per unit volume to the incident pump photon flux $\bar{N}_pC/n_e$ regardless of the coherence property of the pump wave. The transition probability, either Eq. (23a) or Eq. (23b), is already correctly normalized. Thus, to obtain the differential extinction coefficient one simply integrates in the $\mathbf{k}$ spaces the following:

$$d\sigma(\mathbf{k}_s)=\iint|\phi_{\text{final}}^{\dagger}(\mathbf{k}_1=\mathbf{k}_s)S^{(1)}\phi_{\text{incoh}}|^2d\mathbf{k}_2n_eG(\mathbf{k}_p)(VTN_pC)^{-1}d\mathbf{k}_pd\mathbf{k}_s$$

$$=\iint|\phi_{\text{final}}'^{\dagger}(\mathbf{k}_1=\mathbf{k}_s)S^{(1)}\phi_{\text{coh}}|^2d\mathbf{k}_2n_eG(\mathbf{k}_p)(VT\bar{N}_pC)^{-1}d\mathbf{k}_pd\mathbf{k}_s$$

$$=\iint\frac{(\Delta\epsilon_1)^2\hbar\omega_{ep}\omega_{os}\omega_{oi}G(\mathbf{k}_p)}{2\pi Cn_e(\mathbf{k}_p)n_o^2(\mathbf{k}_s)n_o^2(\mathbf{k}_i)}\delta(\mathbf{k}_p-\mathbf{k}_i-\mathbf{k}_s)\delta(\omega_{ep}-\omega_{oi}-\omega_{os})d\mathbf{k}_id\mathbf{k}_pd\mathbf{k}_s, \quad (24)$$

where $G(\mathbf{k}_p)$ is the distribution function of the pump intensity in the $\mathbf{k}_p$ space; it is, as usual, normalized to unity. The δ functions of course ensure that the energy and momentum of the photons are conserved. The integration in $\mathbf{k}_i$ space can be carried out immediately by virtue of the wave-vector, or momentum, matching condition embodied in one of the δ functions:

$$\mathbf{k}_p-\mathbf{k}_i-\mathbf{k}_s=0. \quad (25)$$

By further reexpressing $d\mathbf{k}_s$ in terms of the corresponding spectral width $d\omega_{os}$ and solid angle $d\Omega_s$ *in the medium*, we obtain the final result:

$$d\sigma(\mathbf{k}_s)=\int\frac{(\Delta\epsilon_1)^2\hbar\omega_{ep}\omega_{os}^3\omega_{oi}n_o(\mathbf{k}_s)G(\mathbf{k}_p)}{2\pi C^4n_e(\mathbf{k}_p)n_o^2(\mathbf{k}_p-\mathbf{k}_s)}\delta(F)d\mathbf{k}_pd\omega_{os}d\Omega_s, \quad (26)$$

where in this equation and what follows

$$\omega_{oi}=\omega_{ep}-\omega_{os} \quad (27)$$

and

$$F=\omega_{ep}-\omega_{os}-[n_o(\mathbf{k}_p-\mathbf{k}_s)]^{-1}[n_e^2(\mathbf{k}_p)\omega_{ep}^2+n_o^2(\mathbf{k}_s)\omega_{os}^2-2n_o(\mathbf{k}_s)n_e(\mathbf{k}_p)\omega_{ep}\omega_{os}\hat{k}_s\cdot\hat{k}_p]^{1/2}, \quad (28)$$

on account of the wave-vector matching condition, Eq. (25), and the dispersion relations, Eqs. (9) and (10). These final results, Eqs. (26)–(28), contain all the information on the intensity of the spontaneous emission: For a given pump beam and nonlinear crystal, the tuning characteristics, the spectral properties, and the beam divergence are completely determined through the single remaining δ function, or equivalently the equation

$$F=0, \quad (29)$$

which combines the wave-vector and frequency matching conditions. Equation (26) itself gives the intensity distribution function in the frequency and the direction of the spontaneous emission. In the following section we shall examine in detail the physical consequences of these results.

In crystals such as ADP, the wave-vector and frequency matching conditions can also be satisfied simultaneously when one of the scattered beams is also an extraordinary wave. All the results obtained so far can be simply modified to take care of such situations by using the appropriate subscripts for ω and n.

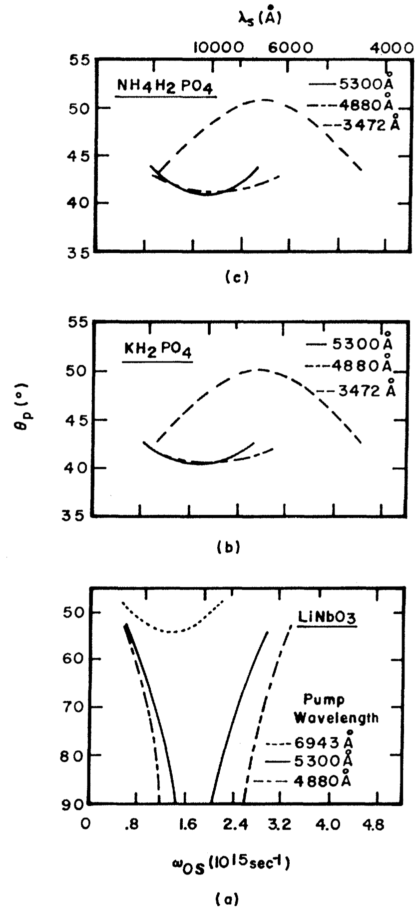

FIG. 1. Colinear tuning characteristics of $LiNbO_3$, KH_2PO_4, and $NH_4H_2PO_4$. λ_p is the pump wavelength and θ_p is the angle between the pump ray and the optic axis of the crystals.

III. SPONTANEOUS EMISSION

We now make quantitative estimates and describe the characteristics of the spontaneous emission in the parametric scattering of intense light in nonlinear optical crystals.

A number of simplifications are possible at the outset, however. The crystals of interest here, such as $LiNbO_3$, ADP, or KDP used in most experimental studies, are all uniaxial crystals; our considerations can, therefore, be limited to crystals with such a symmetry. The spectral width of the pump light is generally much smaller than the spectral width of the spontaneous emission due to, for example, the finite beam divergence of the pump light in most cases. In addition, none of the relevant parameters vary much at all over the pump frequency band. The spectral distribution function of the pump light can, therefore, be assumed to be a δ function centered at the pump frequency ω_p. Finally, we can assume without loss of generality that the pump light is uniformly distributed within its beam width $\Delta\Omega_p$ inside the medium. Thus specialized, Eq. (26) now becomes

$$d\sigma(\mathbf{k}_s)=\int_{\Delta\Omega_p}\frac{(\Delta\epsilon_1)^2\hbar\omega_p\omega_{os}^3\omega_{oi}n_o(\omega_{os})}{2\pi C^4 n_e(\omega_p\theta_p)n_o^2(\omega_{oi})\Delta\Omega_p}\delta(F)\,d\Omega_p d\omega_{os}d\Omega_s, \qquad (30)$$

where θ_p is the direction of a pump ray relative to the optical axis of the crystal and $d\Omega_p$ refers to the corresponding differential solid angle in the medium.

The differential extinction coefficient will vanish unless

$$F=0; \qquad (29)$$

for each set of ω_p, θ_p, and $\hat{k}_s\cdot\hat{k}_p$ values, each real solution of this equation implies a possible frequency for the spontaneous emission. Let us consider first those characteristics of the spontaneous emission that are primarily determined by this equation alone. This will be followed by a detailed study of the physical consequences of Eq. (30).

A. Colinear Tuning Characteristics

First of all, as in the case of the parametric oscillator, Eq. (29) determines the basic tuning characteristics of the spontaneous emission. Let us start with the colinear situation $\hat{k}_s\cdot\hat{k}_p=1$; this case has already been discussed extensively in the past in connection with the parametric oscillators.[1–5,8] Figure 1 gives some additional numerical results covering the entire room-temperature colinear tuning range of ADP, KDP, and $LiNbO_3$ with various pump sources. These computer solutions are based upon the room-temperature

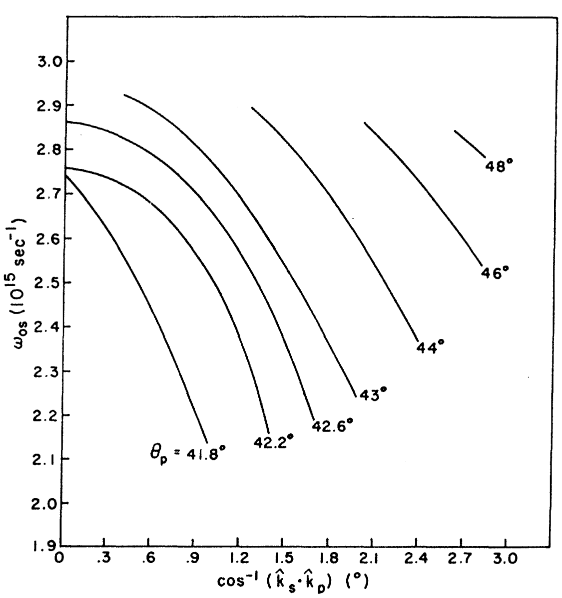

FIG. 2. Noncolinear tuning characteristics of $NH_4H_2PO_4$ pumped at $\lambda_p=4880$ Å.

index of refraction data given in Ref. 19 for ADP and KDP and Ref. 20 for $LiNbO_3$. There are some small differences between the numerical results given in Fig. 1 and the measured results given in Refs. 2, 3, and 5; these small discrepancies are due to the fact that the index of refraction of the crystals vary[3] from boule to boule and the tuning characteristics are very sensitive to such small variations. A precise prescription for determining the temperature tuning characteristics of the oscillators at a fixed θ_p along with extensive additional data on $LiNbO_3$ are given in Ref. 21.

B. Noncolinear Interaction, Beam Width, and Spectral Characteristics

The spectral characteristics and the beam width of the spontaneous emission are primarily determined by the solutions of Eq. (29) for noncolinear interaction, $\hat{k}_s \cdot \hat{k}_p < 1$. As an example, Fig. 2 gives ω_{os} as a function of the noncolinear angle $\cos^{-1}(k_s \cdot k_p)$ for various values of θ_p covering the entire colinear tuning range of ADP pumped at 4880 Å at room temperature. Figure 3 shows the similar results for $LiNbO_3$ pumped at 6943 Å. We have also obtained numerical results for all the other cases shown in Fig. 1. These results on noncolinear interaction point up a number of interesting properties of the spontaneous emission; for example:

(1) The range for noncolinear interaction could be quite large. Thus, even for a very well collimated pump beam, the beam width of the spontaneous emission could be quite broad; the exact width depends, of course, on the crystal and the pump wavelength as well as the value of θ_p. In the case of Fig. 2, for example, the total beam width of the spontaneous emission in the colinear tuning range could be as large as 3.8°, even in the limit of an infinitely narrow ($\Delta\Omega_p \to 0$) pump beam.

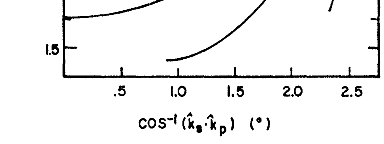

FIG. 3. Noncolinear tuning characteristics of $LiNbO_3$ pumped at $\lambda_p = 6943$ Å.

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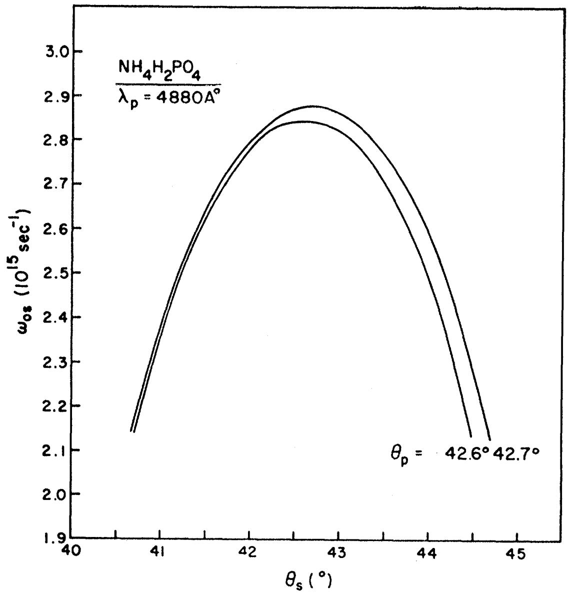

FIG. 4. The region between the two curves corresponds to the frequency and direction of the spontaneous emission in the plane containing the crystal ($NH_4H_2PO_4$) optic axis and the axis of the pump beam ($\lambda_p = 4880$ Å). The pump beam spreads from $\theta_p = 42.6°$ to 42.7°.

(2) For a fixed pump beam direction, the emission frequency changes with the direction of emission. The way the frequency changes depends upon the crystal and the pump wavelength. Of the cases we studied, for ADP and KDP pumped at 4880 and 5300 Å, the frequency decreases from the center toward the edge of the emitted beam when $\omega_{os} > \frac{1}{2}\omega_p$; for ADP and KDP pumped at 3472 Å and $LiNbO_3$ at 4880, 5300, and 6943 Å, the frequency increases from the center toward the edge of the beam when $\omega_{os} > \frac{1}{2}\omega_p$. The total change in frequency could be very large even in the limit of a very narrow beam, as can be seen from Figs. 2 and 3. To cite another dramatic example, in the case of ADP pumped at 3472 Å in the direction of $\theta_p = 50.5°$, the total beam width is 5° and the wavelength varies from 6540 Å at the center to 4700 Å at the edge of the emitted beam; thus, the emitted beam in this case would vary in color continuously from red at the center to blue at the edge.

(3) There could be spontaneous emission due to noncolinear interaction even beyond the colinear tuning range. In this case, the spontaneous emission would be emitted in a cone with a dark region near the center when a narrow pump beam is used. Figures 2 and 3 show examples of this.

(4) If the pump beam width is finite, the spectral width $\Delta\omega_{os}(\mathbf{k}_s)$ of the spontaneous emission also changes continuously with the direction of emission; when $\theta_p \approx \pi/2$, the spectral width is essentially constant. The detailed manner in which the spectral width changes with the direction of emission relative to the crystal optical

axis could be very different, depending on the crystal and the other parameters involved. Figure 4 shows an example of the approximate region in the $(\omega_{os}\theta_s)$ plane corresponding to the spontaneous emission in the plane defined by the optical axis and the pump beam axis of ADP pumped at 4880 Å in the direction $42.6° \leq \theta_p \leq 42.7°$ with a pencil beam. It is seen that there is a substantial increase in the spectral width from the edge nearest the optical axis to the farthest edge. It is clear also that to measure $\Delta\omega_{os}(\mathbf{k}_s)$ the detector aperture must be very small.

Most of these properties of the spontaneous emission have not yet been, but could easily be, observed experimentally.

C. Spontaneous Intensity

To calculate the spontaneously emitted power, one must first evaluate the integral in Eq. (30). To do so exactly is difficult due to certain complications involving the geometric factors of the beam. A very good approximation of the integral can, however, be obtained by assuming $\hat{k}_s \cdot \hat{k}_p \approx 1$ in F and by neglecting the θ_p dependence in the limits of integration in the azimuthal angle ϕ_p, when a narrow pencil-shaped pump beam is used, which is always the case in the experimental studies. Thus, within the beam width and spectral width of the spontaneous emission, or where the equation $F=0$ can be satisfied, one obtains the approximate distribution function for the emitted power:

$$P(\mathbf{k}_s)d\omega_{os}d\Omega_s \equiv d\sigma(\mathbf{k}_s)\hbar\omega_{os}V\bar{N}_pCn_e^{-1}$$
$$\cong \frac{(\Delta\epsilon_1)^2\hbar^2\omega_p\omega_{os}^4\omega_{oi}n_o(\omega_{os})V\bar{N}_pC}{2\pi C^4 n_o^2(\omega_{oi})\Delta\theta_p[n_e^2(\omega_p\theta_p)\,|\,\partial F/\partial\theta_p\,|\,]_{F=0}}d\omega_{os}d\Omega_s$$
$$\cong \frac{(\Delta\epsilon_1)^2\hbar^2\omega_{os}^4\omega_{oi}n_o(\omega_{os})V\bar{N}_pC}{2\pi C^4 n_o(\omega_{oi})n_e^2(\omega_p\bar{\theta})_p\Delta\theta_p\,|\,\partial n_e/\partial\theta_p\,|_{\bar{\theta}_p}}d\omega_{os}d\Omega_s, \tag{31}$$

where V is the volume of the interaction region, $\bar{\theta}_p$ refers to the direction of the axis of the pump beam, and $\Delta\bar{\theta}_p$ is the corresponding linear width of the beam in the medium. This result is not valid when $\bar{\theta}_p$ is in the direction where $\partial n_e/\partial\theta_p=0$. In this particular case, F in the δ function has essentially no angular dependence and the distribution function for the emitted power should be

$$P(\mathbf{k}_s)d\omega_{os}d\Omega_s = \frac{(\Delta\epsilon_1)^2\hbar^2\omega_p\omega_{os}^4\omega_{oi}n_o(\omega_{os})V\bar{N}_pC}{2\pi C^4 n_o^2(\omega_{oi})n_e^2(\omega_p\tfrac{1}{2}\pi)}\delta(\bar{F})d\omega_{os}d\Omega_s, \tag{32}$$

where in terms of Eq. (28),

$$\bar{F} = F(\theta_p = \tfrac{1}{2}\pi,\ \hat{k}_s\cdot\hat{k}_p = 1).$$

For a fixed direction of $\mathbf{k}_s$, the spectral width of the spontaneous emission is typically fairly narrow so that all the factors in these expressions, Eqs. (31) and (32), are essentially constant over the spectral width; one can, therefore, integrate these expressions in ω_{os} and obtain the angular distribution of the spontaneous emission. For example, the spontaneously emitted power varies with $\mathbf{k}_s$ as

$$\int P(\mathbf{k}_s)d\omega_{os}d\Omega_s = \frac{(\Delta\epsilon_1)^2\hbar^2\omega_{os}^4\omega_{oi}n_o(\omega_{os})V\bar{N}_pC}{2\pi C^4 n_o(\omega_{oi})n_e^2(\omega_p\bar{\theta}_p)\Delta\theta_p\,|\,\partial n_e/\partial\theta_p\,|_{\bar{\theta}_p}}\Delta\omega_{os}(\mathbf{k}_s)d\Omega_s \tag{33}$$

from Eq. (31), where ω_{os} is a solution of Eq. (29) for the particular value of $\mathbf{k}_s$. The emitted power per unit solid angle is directly proportional to the spectral width $\Delta\omega_{os}(\mathbf{k}_s)$ as a function of $\mathbf{k}_s$; when $\theta_s=\bar{\theta}_p$, the spectral width is equal to the colinear tuning range corresponding to the given value of $\Delta\theta_p$. As can be seen from the example given in Fig. 4 and the related discussions, the intensity is also expected to change with the direction of emission with respect to the optical axis. In the case corresponding to Eq. (32), or $\bar{\theta}_p=\frac{1}{2}\pi$, there is no such a change for a typically narrow pump beam and the angular distribution of the emitted power is a constant:

$$\int P(\mathbf{k}_s)d\omega_{os}d\Omega_s = \frac{(\Delta\epsilon_1)^2\hbar^2\omega_p\omega_{os}^4\omega_{oi}n_0(\omega_{os})V\bar{N}_pCd\Omega_s}{2\pi C^4 n_e^2(\omega_p\tfrac{1}{2}\pi)n_o(\omega_{oi})\,|\,n_o(\omega_{os})-n_o(\omega_{oi})\,|}, \tag{34}$$

where ω_{os} is a solution of Eq. (29).

Finally, to facilitate direct comparison with the experimental results, we reexpress these results in the form of the emitted power that would be measured by a detector:

$$P_s = \frac{(\Delta\epsilon_1)^2\hbar\omega_{os}^4\omega_{oi}LP_p\Delta\omega_{os}(\mathbf{k}_s)\Delta\Omega_{\text{det}}}{2\pi C^4\omega_p n_o(\omega_{os})n_o(\omega_{oi})\,|\,\partial n_e/\partial\theta_p\,|_{\bar{\theta}_p}\Delta\theta_{\text{inc}}} \tag{35}$$

from Eq. (33), where $\Delta\Omega_{\text{det}}$ is the solid angle subtended by the detector and we assume that $\Delta\Omega_{\text{det}}$ is sufficiently small so that the intensity variation within it is negligible. We have also used in this expression the incident pump beam angle $\Delta\theta_{\text{inc}}$ outside the medium in the free space. L is the length of the interaction region measured along the pump beam; and P_p is the total pump power. Similarly, for Eq. (34), we have

$$P_s=\frac{(\Delta\epsilon_1)^2\hbar\omega_{os}{}^4\omega_{oi}LP_p\Delta\Omega_{\text{det}}}{\pi C^4 n_o(\omega_{os})n_o(\omega_{oi})n_e{}^2(\omega_p\tfrac{1}{2}\pi)\ |\ n_o(\omega_{os})-n_o(\omega_{oi})\ |}, \tag{36}$$

which does not apply at the degenerate point where $\omega_{os}=\omega_{oi}$. It must be emphasized that $\Delta\omega_{os}(\mathbf{k}_s)$ in (35) is not the total bandwidth of the spontaneous emission measured with a finite detector; it corresponds to the bandwidth in the limit of $\Delta\Omega_{\text{det}}\rightarrow 0$.

From an experimental point of view, these formulas are the two most important new results of the present theory on the quantum noise in the parametric scattering of intense light. We now apply these results to two typical experimental situations.

D. Numerical Examples

Consider, for example, the case of ADP pumped at 3472 Å; extensive data on the quantum noise for this case are given by Magde and Mahr.[5] The colinear tuning characteristics have already been discussed. Consider now the emitted power at 7800 Å. ADP has a V_d point-group symmetry; in addition, when the pump wave is an extraordinary wave while the spontaneously emitted waves are ordinary waves, the coefficient $\Delta\epsilon_1$ defined in Eq. (17) becomes

$$\Delta\epsilon_1=8\pi\chi_{zxy}e_zo_xo_y.$$

If the pump wave makes $\bar{\theta}_p$ with respect to the optical axis, the $\hat{z}$ axis, and 45° with respect to the $\bar{x}$ and $\bar{y}$ axes of the crystal, we have $e_z=\sin\bar{\theta}_p$, $o_x=o_y=1/\sqrt{2}$ and, hence,

$$\Delta\epsilon_1=4\pi\chi_{zxy}\sin\bar{\theta}_p\cong 1.7\times10^{-8}\sin\bar{\theta}_p \text{ esu}$$

from the data given in Ref. 22. The remaining parameters for the experiment of Ref. 5 are

$\bar{\theta}_p\approx 49.5°$, $L=1.15$ cm,

$\Delta\omega_{os}(\bar{\theta}_p)\approx 1.2\times10^{14}$ sec^{-1},

$n_e(\omega_p\bar{\theta}_p)\approx 1.53$, $n_o(\omega_{os})\approx n_o(\omega_{oi})\approx 1.52$,

$|\,\partial n_e/\partial\theta_p\,|_{\bar{\theta}_p}\approx 0.05$, $P_p=2.5\times10^5$ W,

$\Delta\Omega_{\text{det}}\approx 1.5\times10^{-5}$ sr, $\Delta\theta_{\text{inc}}\approx 5.7\times10^{-3}$ rad.

From the formula for the spontaneously emitted power, Eq. (35), one obtains $P_s\cong 10^{-6}$ W, which is exactly the measured value. Furthermore, the frequency dependence of the emitted power predicted by this formula, Eq. (35), also agrees with that given in Fig. 3 of Ref. 5. Equation (35) obviously also has the correct length L dependence.

We consider also an example that requires the use of Eq. (36). Consider $LiNbO_3$ pumped by the 4880 Å line of the $Ar^{\dagger}$ laser at $\bar{\theta}_p=\frac{1}{2}\pi$. As pointed out earlier, with a gas laser as the pump source, there is very little gain. On the other hand, the spontaneous emission could be sufficiently intense for the purpose of studying the characteristics of the spontaneous emission. For $LiNbO_3$ pumped by an extraordinary wave at $\bar{\theta}_p=\frac{1}{2}\pi$, one has

$$\Delta\epsilon_1=8\pi\chi_{zxy}e_zo_xo_y\approx 4.1\times10^{-7}\text{ esu}$$

using the data listed in Ref. 22. For the remaining parameters, let us take

$$\omega_{os}=2\pi\times4.1\times10^{14}\text{ sec}^{-1},\ L=0.5\text{ cm},$$

$$n_o(\omega_{os})\approx 2.286,\ n_o(\omega_{oi})\approx 2.197,\ n_e(\omega_p\tfrac{1}{2}\pi)\approx 2.25,$$

$$\Delta\Omega_{\text{det}}\approx 2.4\times10^{-4}\text{ sr},\qquad P_p=1\text{ W}.$$

Using these numbers, Eq. (36) predicts a spontaneous power of

$$P_s=2\times10^{-10}\text{ W}$$

at 7300 Å, for example, which is quite reasonable and it is certainly adequate for the purpose of studying the quantum noise in the parametric scattering of intense light in $LiNbO_3$.

ACKNOWLEDGMENTS

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