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Spontaneous Emission in the Frequency Up-Conversion Process in Nonlinear Optics*

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The spontaneous emission in the frequency up-conversion process in nonlinear optics due to the second-order process consisting of the mismatched spontaneous down-conversion process followed by the phase-matched up-conversion process is studied in detail as a quantum-mechanical scattering problem. The calculated integrated intensity of the spontaneous emission within the spectral and angular widths for the first-order phase-matched up-conversion process is found to be exactly the same as that obtained by Smith and Townes classically. By carefully choosing the parameters involved, it may be possible to eliminate the spontaneous emission from the detected output at the sum frequency of the up-conversion process. It therefore appears that the up-conversion process may indeed be made essentially noise-free and, with further improvements in the conversion efficiency, it is a very promising means of detecting low-level infrared radiation.

I. INTRODUCTION

THE frequency up-conversion process in nonlinear optics has been observed and studied extensively by a number of authors in the past[1] and has lately received considerable attention as a promising new means of detecting low-level infrared radiation.[2–7] In a recent study on the general problem of detecting infrared radiation, Smith and Townes[7] discussed some of the processes which could improve detection sensitivity in the infrared and analyzed in particular the technique making use of the up-conversion process. In this connection, one of the most intriguing questions concerns the spontaneous emission in the up-conversion process.

In the first-order up-conversion process, an infrared photon is combined with a pump photon to form a visible photon. It is clear from conservation of energy that no photons at the sum frequency in the visible can be produced spontaneously from the pump photons in the absence of any photons at the infrared frequency. At room temperature, the contribution at the sum frequency due to the blackbody radiation at the infrared frequency is insignificant; thus, in principle, the up-conversion process does not introduce any additional noise, at least to the first order. However, noise of an unknown origin has been observed in the experiment of Midwinter and Warner.[2] A number of possible sources for this observed noise has been suggested.[2] Some of these such as fluoresence in the filters, heating of dust particles, etc., are not basic to the up-conversion process and can presumably be eliminated; others are inherent to the up-conversion process and will thus determine the ultimate sensitivity of this detection scheme. The most important of these is the second-order process whereby a pump photon of angular frequency ω_p first decays spontaneously into an infrared photon at ω_i and another at $\omega_- = \omega_p - \omega_i$; the infrared photon then combines with another pump photon to form a spontaneously emitted visible photon at the sum frequency $\omega_+ = \omega_i + \omega_p$. Since, in general, it is not possible to satisfy the energy and momentum matching conditions for

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[1] W. H. Louisell, A. Yariv, and A. E. Siegman, Phys. Rev. **124**, 1646 (1961); J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, *ibid.* **127**, 1918 (1962); J. Giordmaine, Phys. Rev. Letters **8**, 19 (1962); A. W. Smith and Breslav, IBM J. Res. Develop. **6**, 361 (1962); G. D. Boyd and A. Ashkin, Phys. Rev. **146**, 187 (1966); D. Kleinman and G. Boyd, J. Appl. Phys. (to be published).

[2] J. E. Midwinter and J. Warner, J. Appl. Phys. **38**, 519 (1967).

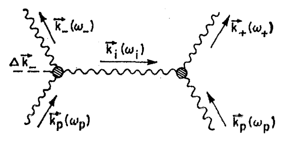
[3] F. M. Johnson, IEEE J. Quant. Electron. **QE-2**, xxi (1966).

[4] R. C. Miller and W. A. Nordland, IEEE J. Quant. Electron. **QE-3**, 642 (1967).

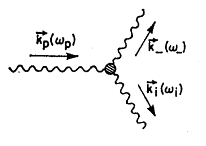
[5] J. E. Midwinter, Appl. Phys. Letters **12**, 68 (1968).

[6] J. E. Midwinter, in Proceedings of the Fifth International Congress of Quantum Electronics, Miami, Florida, 1968 (unpublished); IEEE J. Quant. Electron. **QE-4**, 716 (1968).

[7] H. A. Smith and C. H. Townes (unpublished).



(a)



(b)

FIG. 1. Schematic diagrams representing (a) the second-order process consisting of the mismatched spontaneous down-conversion process followed by the phase-matched up-conversion process. The momentum mismatch for the down-conversion process is

$$\Delta\mathbf{k}_-\equiv\mathbf{k}_p-\mathbf{k}_i-\mathbf{k}_-.$$

(b) The spontaneous down-conversion process.

both the down- and up-conversion processes simultaneously and the up-conversion process must be phase-matched for efficient conversion of the infrared radiation to the visible, the down-conversion process is usually highly mismatched. This second-order process consisting of the mismatched down-conversion process followed by the matched up-conversion process is the most important source of inherent noise, or spontaneous emission, in the frequency up-conversion process. It is represented schematically in the diagram shown in Fig. 1(a).

Smith and Townes have made a detailed study of this noise process on the basis of a classical picture for interaction of the fields together with some reasonable assumptions about the physical origin and nature of the fluctuations that eventually show up in the form of the spontaneous emission at the sum frequency. In the present study, this process is treated directly as a quantum-mechanical scattering process that is represented by the second-order diagram shown in Fig. 1(a). There is then no need to make any assumptions about the initial fluctuations. Hopefully, this alternative approach will lead to a better understanding of some of the theoretical aspects of this problem.

The general procedure used here is essentially the same as that used in Ref. 8 for the spontaneous parametric scattering of light,[9] or the first-order spontaneous down-conversion process, which is represented schematically by the first-order diagram shown in Fig. 1(b) and was treated[8] as an elementary quantum-mechanical scattering problem. In this approach, the fields are all quantized but the nonlinearity of the optical crystal is simply characterized by a constant nonlinear susceptibility in the usual way.[10] Since most of the nonlinear optical crystals of interest are uniaxial crystals, in what follows we shall tacitly assume that the medium under consideration has such a symmetry.

II. TRANSITION PROBABILITIES AND DISTRIBUTION FUNCTIONS

A. Field Quantization and Interaction Lagrangian

The formal problem of quantizing the field is exactly the same as that encountered in the study of the spontaneous parametric scattering process considered in detail in Ref. 8. We summarize here the necessary preliminaries; they are, of course, subject to the same restricting conditions as those of Ref. 8.

The total Lagrangian density L_{tot} of the fields in the nonlinear medium can be split up into a part L_0 for the free fields in the medium and a part L_1 that describes the interaction due to the nonlinearity

$$L_{\text{tot}}=L_0+L_1. \tag{1}$$

Consider first the part independent of the nonlinearity:

$$L_0=(1/8\pi)(\mathbf{D}\cdot\mathbf{E}-(1/\mu_0)\mathbf{B}\cdot\mathbf{B}) \tag{2}$$

and the corresponding Hamiltonian

$$H_0=(1/8\pi)\int\left(\mathbf{D}\cdot\mathbf{E}+\frac{1}{\mu_0}\mathbf{B}\cdot\mathbf{B}\right)d\mathbf{r}, \tag{3}$$

where μ_0 is the permeability of the medium and the $\mathbf{D}$ vector is related to the $\mathbf{E}$ vector by a dielectric tensor.

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

$$\begin{aligned}\mathbf{E}(\mathbf{r},t)&=\mathbf{E}_0(\mathbf{r},t)+\mathbf{E}_e(\mathbf{r},t)\\&=\frac{i}{2\pi}\int\frac{(\hbar\omega_0)^{1/2}}{n_0(\mathbf{k})}[a_0(\mathbf{k})e^{i\mathbf{k}\cdot\mathbf{r}-i\omega_0 t}\\&\qquad-a_0^\dagger(\mathbf{k})e^{-i\mathbf{k}\cdot\mathbf{r}+i\omega_0 t}]\hat{O}(\hat{k})d\mathbf{k}\\&\quad+\frac{i}{2\pi}\int\frac{(\hbar\omega_e)^{1/2}}{n_e(\mathbf{k})}[a_e(\mathbf{k})e^{i\mathbf{k}\cdot\mathbf{r}-i\omega_e t}\\&\qquad-a_e^\dagger(\mathbf{k})e^{-i\mathbf{k}\cdot\mathbf{r}+i\omega_0 t}]\hat{e}(\hat{k})d\mathbf{k},\end{aligned} \tag{4}$$

where $a_0^\dagger(\mathbf{k})$ and $a_0(\mathbf{k})$ are the creation and annihilation operators for a quantum of ordinary wave with wave vector $\mathbf{k}$ and polarization $\hat{O}(\hat{k})$; similarly, $a_e^\dagger(\mathbf{k})$ and $a_e(\mathbf{k})$ refer to the extraordinary wave. According to the usual rules of field quantization,[11] the creation and

[8] T. G. Giallorenzi and C. L. Tang, Phys. Rev. **166**, 225 (1968).

[9] D. Magde and H. Mahr, Bull. Am. Phys. Soc. **12**, 273 (1967); Phys. Rev. Letters **18**, 905 (1967); Phys. Rev. (to be published); S. E. Harris, M. K. Oshman, and R. L. Beyer, Phys. Rev. Letters **18**, 732 (1967); Phys. Rev. (to be published); D. N. Klyshko, Zh. Eksperim. i Teor. Fiz. Pis'ma v Redakstiyu **6**, 490 (1966); S. Akhmanov, V. Fadeev, R. Khokhlov, and O. Chunaev *ibid.* **6**, 575 (1966) [English transls.: Soviet Phys.—JETP Letters **6**, 23 (1967); **6**, 85 (1967)]; D. Magde, R. Scarlet, and H. Mahr, Appl. Phys. Letters **11**, 381 (1967); R. G. Smith, J. G. Skinner, J. E. Geusic, and W. G. Nilsen, *ibid.* **12**, 97 (1968); D. A. Kleinman, Phys. Rev. (to be published); T. G. Giallorenzi and C. L. Tang, Appl. Phys. Letters **12**, 376 (1968).

[10] D. A. Kleinman, Phys. Rev. **126**, 1977 (1962); P. A. Franken and F. J. Ward, Rev. Mod. Phys. **35**, 23 (1963); N. Bloembergen, *Nonlinear Optics* (W. A. Benjamin, Inc., New York, 1965); A. Yariv, *Quantum Electronics* (Wiley-Interscience, Inc., New York, 1967).

annihilation operators satisfy the commutation rules

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

$$[a_0(\mathbf{k}),\, a_0^\dagger(\mathbf{k}')] = \delta(\mathbf{k}-\mathbf{k}'), \tag{5b}$$

$$[a_e(\mathbf{k}),\, a_e^\dagger(\mathbf{k}')] = \delta(\mathbf{k}-\mathbf{k}'), \tag{5c}$$

$$[a_e(\mathbf{k}),\, a_0^\dagger(\mathbf{k}')] = 0. \tag{5d}$$

The angular frequencies ω_0 and ω_e are related to the magnitude of the corresponding wave vector $\mathbf{k}$ and the indices of refraction, $n_0(\mathbf{k})$ and $n_e(\mathbf{k})$, for the ordinary and extraordinary waves, respectively, as follows:

$$\omega_0 n_0(\mathbf{k}) - |\mathbf{k}|c = 0, \tag{6a}$$

$$\omega_e n_e(\mathbf{k}) - |\mathbf{k}|c = 0, \tag{6b}$$

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

$$H_0 = \tfrac{1}{2} \int \hbar\omega_0 [a_0^\dagger(\mathbf{k}) a_0(\mathbf{k}) + a_0(\mathbf{k}) a_0^\dagger(\mathbf{k})] d\mathbf{k} + \tfrac{1}{2} \int \hbar\omega_e [a_e^\dagger(\mathbf{k}) a_e(\mathbf{k}) + a_e(\mathbf{k}) a_e^\dagger(\mathbf{k})] d\mathbf{k}. \tag{7}$$

The interaction Lagrangian density is determined by the following considerations. In a nonlinear crystal, a component of the electric displacement vector contains an additional term $D_i^{\mathrm{NL}}(\mathbf{r},t)$, corresponding to the nonlinear polarization, $P_i^{\mathrm{NL}}(\mathbf{r},t)$, which is related to the electric field components through a nonlinear susceptibility coefficient

$$D_i^{\mathrm{NL}}(\mathbf{r},t) = 4\pi P_i^{\mathrm{NL}}(\mathbf{r},t) = 4\pi \sum_{jk} d_{ijk} E_j(\mathbf{r},t) E_k(\mathbf{r},t), \tag{8}$$

where we have neglected the frequency dependence of the nonlinear coefficient d_{ijk}, which is a valid simplification within the context of the present problem. Since, in general,

$$D_i(\mathbf{r},t) = 4\pi[\partial L(\mathbf{r},t)/\partial E_i(\mathbf{r},t)],$$

we have

$$D_i^{\mathrm{NL}}(\mathbf{r},t) = 4\pi[\partial L_1(\mathbf{r},t)/\partial E_i(\mathbf{r},t)] \tag{9}$$

and

$$L_1(\mathbf{r},t) = \tfrac{1}{3} \sum_{ijk} d_{ijk} E_i(\mathbf{r},t) E_j(\mathbf{r},t) E_k(\mathbf{r},t), \tag{10}$$

where we have made use of the symmetry condition[10] that d_{ijk} is invariant under permutation of the subscripts i, j, and k.

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

[11] See, for example, N. N. Bogoliubov and D. N. Shirkov, *Introduction to the Theory of Quantized Fields* (Wiley-Interscience, Inc., New York, 1959).

B. Second-Order Scattering Matrix

To study the quantum noise in the up-conversion process due to the infrared photons generated by the spontaneous down-conversion process, one must consider the second-order term in the perturbation expansion of the scattering matrix, $S^{(2)}$. We first give the general expression of $S^{(2)}$, which contains all the terms corresponding to all the possible second-order processes; only some of these terms contribute to the quantum noise in the up-conversion process of interest here. The general expression of $S^{(2)}$ is the following[11]:

$$\begin{aligned} S^{(2)} &= -\frac{1}{\hbar^2} \int\!\!\int\!\!\int\!\!\int T[L_1(\mathbf{r},t) L_1(\mathbf{r}',t')] d\mathbf{r}dtd\mathbf{r}'dt' \\ &= -\frac{1}{9\hbar^2} \int\!\!\int\!\!\int\!\!\int \sum_{ijk}\sum_{lmn} d_{ijk} d_{lmn} T[E_i(\mathbf{r},t) E_j(\mathbf{r},t) \\ &\qquad \times E_k(\mathbf{r},t) E_l(\mathbf{r}',t') E_m(\mathbf{r}',t') E_n(\mathbf{r}',t')] d\mathbf{r}dtd\mathbf{r}'dt' \end{aligned} \tag{11}$$

with the help of Eq. (4) and T refers to the T product (or the chronological product); the T product of the six E operators is equal to the product of these operators taken in the order which corresponds to an increase in the time arguments.

According to Wick's theorem[11] for chronological products, the T product of a system of n linear operators is equal to the sum of their normal products with all possible chronological pairings. Chronological pairing of two E operators is by definition

$$\begin{aligned} [\text{pair } E(r,t)E(r',t')] &= \phi_0^\dagger E(\mathbf{r},t) E(\mathbf{r}',t') \phi_0, \quad \text{if } t > t' \\ &= \phi_0^\dagger E(\mathbf{r}',t') E(\mathbf{r},t) \phi_0, \quad \text{if } t' > t \end{aligned} \tag{12}$$

where ϕ_0 represents the vacuum state. It, therefore, describes the creation of a photon at $(\mathbf{r}',t')$ followed by its annihilation at $(\mathbf{r},t)$ or vice versa depending on whether t is greater or less than t'; it is also related to the causal Green's function[11] of the wave equation for the field. Since in the process of interest here there can be only one such act of creating and annihilating an intermediate infrared photon, of all the possible terms in the T product of the E operators only those involving one such pairing are of interest and $S^{(2)}$ can be rewritten

$$\begin{aligned} S^{(2)} = -\frac{1}{9\hbar^2} \sum_{ijk}\sum_{lmn} d_{ijk} d_{lmn} \times 9 \times \int\!\!\int\!\!\int\!\!\int :E_i(\mathbf{r},t) E_j(\mathbf{r},t) \\ \times [\text{pair } E_k(\mathbf{r},t) E_l(\mathbf{r}',t')] \\ \times E_m(\mathbf{r}',t') E_n(\mathbf{r}',t') : d\mathbf{r}dtd\mathbf{r}'dt'. \end{aligned}$$

The extra factor of 9 multiplying the integral is there because there are nine equivalent choices of pairing the E operators with arguments $(\mathbf{r},t)$ and $(\mathbf{r}',t')$. The normal product of the E operators is designated by : :; it means that in each term within the : : signs the unpaired annihilation operators should be placed to the right of the unpaired creation operators. Note that each E operator is the sum of an annihilation operator E^- and a creation operator E^+.

For definiteness, we assume that one of the four unpaired E operators corresponds to an extraordinary wave and all the others including the paired E operators are associated with ordinary waves. This choice for the up-conversion process corresponds to the experimental situation in Ref. 2. For this particular choice, the scattering matrix of interest becomes

$$S^{(2)}=-\frac{4}{\hbar^2}\sum_{ijk}\sum_{lmn}d_{ijk}d_{lmn}\int\int\int\int :E_{0i}(\mathbf{r},t)E_{0j}(\mathbf{r},t)\times[\text{pair } E_{0k}(\mathbf{r},t)E_{0l}(\mathbf{r}',t')]\times E_{0m}(\mathbf{r}',t')E_{en}(\mathbf{r}',t'):d\mathbf{r}dtd\mathbf{r}'dt'\,; \quad (13)$$

corresponding results can easily be obtained for other choices. The factor 4 arises because there are four equivalent choices of assigning the subscript e referring to the extraordinary wave to one of the four unpaired E operators. At this point, we can also give the pairing in the representation given in Eq. (4):

$$[\text{pair } E_{0k}(\mathbf{r},t)E_{0l}(\mathbf{r}',t')]=\left(\frac{1}{2\pi}\right)^4\int\int\left[\frac{4\pi\hbar\omega_i^2}{c^2}\right]\times\left[|\mathbf{k}_i|^2-\frac{n_0^2(\omega_i)\omega_i^2}{c^2}+i\epsilon\right]^{-1}e^{-i\mathbf{k}_i\cdot(\mathbf{r}-\mathbf{r}')+i\omega_i(t-t')}\times O_k(\hat{k}_i)O_l(\hat{k}_i)d\mathbf{k}_id\omega_i\,, \quad (14)$$

where ϵ is the usual small parameter introduced to specify the path of integration in the complex ω_i plane near the poles; and $O_k(\hat{k}_i)$ and $O_l(\hat{k}_i)$ are the projection of $\hat{O}(\hat{k}_i)$ on the unit vectors $\hat{k}$ and $\hat{l}$, respectively, referred to in the subscripts of the nonlinear coefficients. Equation (14) is obtained from Eqs. (14.25) and (14.26) of Ref. 11 after introducing all the necessary multiplying constants to account for the differences in the normalizing constants and the systems of units adopted here and in Ref. 11. With the scattering matrix given in Eq. (13) and the pairing of the E operators describing the creation, propagation, and annihilation of the intermediate infrared photon known, we can now proceed to calculate the matrix element and the corresponding transition probability between specific initial and final states.

We must first specify the initial and final states of interest. In the absence of the nonlinearity the only field present should be that of the incident pump beam. Unlike the spontaneous parametric scattering problem, which is a first-order process, the spontaneous intensity in the second-order up-conversion process would depend on the coherence properties of the pump beam. To simplify the algebra, however, we shall use the simpler fixed-number state for the pump beam; the influence of the coherence properties of the pump beam will be considered in a later study. Thus, the initial state representing N_p pump photons per unit volume *to be used in conjunction with* $S^{(2)}$ *given in Eq. (13)* is

$$\phi_{\text{in}}=[(2\pi)^3/(N_p!)^{1/2}][a_0^\dagger(\mathbf{k}_p)]^{N_p}\phi_0. \quad (15)$$

It should be pointed out that the factor $(2\pi)^3$ appears here in the normalizing constant[12] because, in the second-order process considered here, two pump photons are annihilated in each elementary act of scattering; if only one input photon is involved, $(2\pi)^{3/2}$ would appear. It should also be pointed out that Eq. (15) is to be used in the situation where the pump beam divergence can be neglected; for ordinary pump waves, this is a valid simplification here. Otherwise, creation operators corresponding to two different wave vectors $\mathbf{k}_p$ and $\mathbf{k}_p'$, which would eventually be integrated over the same range of beam directions, would appear in Eq. (15); in such a case, the need for the $(2\pi)^3$ factor would be even more transparent.[12] In the presence of the nonlinearity, for each pair of pump photons annihilated due to the process characterized by $S^{(2)}$, Eq. (13), there will be one output photon at the sum frequence ω_{e+} with a wave vector designated $\mathbf{k}_+$ and a corresponding down-converted photon with a wave vector $\mathbf{k}_-$. Thus, the final state of interest is

$$\phi_{\text{final}}^\dagger=\phi_0^\dagger\frac{[a_0(\mathbf{k}_p)]^{N_p-2}a_0(\mathbf{k}_-)a_e(\mathbf{k}_+)}{[(N_p-2)!]^{1/2}}, \quad (16)$$

which is normalized to unity: $|\phi_{\text{final}}|^2=1$, and we assume that the up-converted wave is the extraordinary wave.

Substituting Eqs. (4) and (14) into Eq. (13), we obtain the matrix element of $S^{(2)}$ between the initial and final states, Eqs. (15) and (16), of interest:

$$\phi_{\text{final}}^\dagger S^{(2)}\phi_{\text{in}}=-\frac{2\hbar\omega_{0p}[\omega_{0-}\omega_{e+}N_p(N_p-1)]^{1/2}}{n_0^2(\mathbf{k}_p)n_0(\mathbf{k}_-)n_e(\mathbf{k}_+)}\int\int\Delta\epsilon_o\Delta\epsilon_e\frac{2\omega_i^2}{c^2}\left[|\mathbf{k}_i|^2-\frac{n_0^2(\omega_i)\omega_i^2}{c^2}+i\epsilon\right]^{-1}$$
$$\times\left[\frac{2\sin\frac{1}{2}(\mathbf{k}_p-\mathbf{k}_i-\mathbf{k}_-)\cdot\hat{k}_pL}{(\mathbf{k}_p-\mathbf{k}_i-\mathbf{k}_-)\cdot\hat{k}_p}\right]\delta[(\mathbf{k}_p-\mathbf{k}_i-\mathbf{k}_-)\cdot\hat{t}_p]\delta[(\mathbf{k}_p-\mathbf{k}_i-\mathbf{k}_-)\cdot\hat{t}_p']$$
$$\times\left[\frac{2\sin\frac{1}{2}(\mathbf{k}_p+\mathbf{k}_i-\mathbf{k}_+)\cdot\hat{k}_pL}{(\mathbf{k}_p+\mathbf{k}_i-\mathbf{k}_+)\cdot\hat{k}_p}\right]\delta[(\mathbf{k}_p+\mathbf{k}_i-\mathbf{k}_+)\cdot\hat{t}_p]\delta[(\mathbf{k}_p+\mathbf{k}_i-\mathbf{k}_+)\cdot\hat{t}_p']$$
$$\times\delta(\omega_{0p}-\omega_i-\omega_{0-})\delta(\omega_{0p}+\omega_i-\omega_{e+})d\mathbf{k}_id\omega_i\,, \quad (17)$$

[12] See, for example, Ref. 11, pp. 267–271.

where

$$\Delta\epsilon_0=4\pi\sum_{ijk} d_{ijk}O_i(\hat{k}_p)O_j(\hat{k}_i)O_k(\hat{k}_-)\,,\qquad \Delta\epsilon_e=4\pi\sum_{lmn} d_{lmn}O_l(\hat{k}_p)O_m(\hat{k}_i)e_n(\hat{k}_+)\,;\tag{18}$$

the length of the nonlinear crystal in the pump beam direction $\hat{k}_p$ is assumed to be L, and $\hat{t}_p$ and $\hat{t}_p'$ are two orthogonal unit vectors perpendicular to $\hat{k}_p$. Equation (17) is also represented by the second-order diagram shown in Fig. 1(a). In obtaining Eq. (17), we have also made use of the following considerations. The orientation of the nonlinear crystal and the choice of the pump frequency are such that the first-order up-conversion process can be phase-matched; this means that the frequency and momentum matching conditions $\omega_{0p}+\omega_{0i}-\omega_{e+}=0$ and $\mathbf{k}_p+\mathbf{k}_i-\mathbf{k}_+=0$ can be simultaneously satisfied for some ω_i and $\mathbf{k}_i$ that also satisfy a dispersion relation of the form of Eq. (6a). This is always the case, since the question of the spontaneous emission is only of interest in connection with the use of the first-order phase-matched up-conversion process for detecting infrared radiation. In general, when the first-order up-conversion process is phase-matched, phase-matching for the down-conversion process is usually not possible. For the same reason, we have neglected in Eq. (17) the term in this matrix element that corresponds to the process whereby two pump photons combine to produce a photon at the second harmonic of the pump frequency, $\mathbf{k}_p+\mathbf{k}_p\rightarrow\mathbf{k}_i=2\mathbf{k}_p$, followed by the spontaneous decay of the second harmonic photon into a $\mathbf{k}_+$ and a $\mathbf{k}_-$ photon.[13] In this connection, it should also be mentioned that the four-photon process $\mathbf{k}_p+\mathbf{k}_p\rightarrow\mathbf{k}_-+\mathbf{k}_+$, or the single step light-by-light scattering process,[13,14,7] can also contribute to the noise at the sum frequency of the first-order up-conversion process. Depending primarily on the relative magnitudes of the nonlinear coefficients $|d^{(3)}|^2$ and $|d^{(2)}|^4$, the total integrated intensity of the spontaneous emission due to this four-photon process may even be higher than that due to the two-step second-order process considered here. However, the spontaneous emission due to the four-photon process is expected to be diffused in frequency and direction, its intensity within the narrow frequency and angular widths for the first-order phase-matched up-conversion process is expected to be much smaller and is, hence, neglected in the present study. Conceivably, there could be special situations where this source of spontaneous emission must also be included.

Before we evaluate the integrals in Eq. (17), it is important to consider the physical significance of the function

$$\begin{aligned}[][|\mathbf{k}_i|^2-n_0^2(\omega_i)\omega_i^2/c^2+i\epsilon]^{-1}\\ &=i\pi\delta[|\mathbf{k}_i|^2-n_0^2(\omega_i)\omega_i^2/c^2]\\ &\quad+P[|\mathbf{k}_i|^2-n_0^2(\omega_i)\omega_i^2/c^2]^{-1},\end{aligned}\tag{19}$$

where P designates the principal value. These two terms in Eq. (19) describe how the infrared photons in the intermediate states can contribute to spontaneous emission at the sum frequency in two different ways. Physically, the first term describes photons in the intermediate state that satisfy the dispersion relation Eq. (6a). The second term describes photons in the intermediate state that do not satisfy the dispersion relation Eq. (6a); they may be regarded as "virtual" photons.[15]

We return now to Eq. (17); after carrying out the integrations in (17) with the help of Eq. (19), one finds

$$\begin{aligned}\phi_{\text{final}}^\dagger S^{(2)}\phi_{\text{in}}=&-\frac{8\pi\Delta\epsilon_0\Delta\epsilon_e\hbar\omega_{0p}\omega_R^2[\omega_{0-}\omega_{e+}N_p(N_p-1)]^{1/2}}{c^2n_0^2(\mathbf{k}_p)n_0(\mathbf{k}_-)n_e(\mathbf{k}_+)(\mathbf{k}_R\cdot\hat{k}_p)}\\ &\times\left[i\frac{\sin\frac{1}{2}\Delta\mathbf{k}_+\cdot\hat{k}_pL}{\Delta\mathbf{k}_+\cdot\hat{k}_p}+\frac{2\sin^2(\frac{1}{4}L\Delta\mathbf{k}_+\cdot\hat{k}_p)}{(\Delta\mathbf{k}_+\cdot\hat{k}_p)}\right]\\ &\times\delta(\Delta\mathbf{k}_-\cdot\hat{t}_p)\delta(\Delta\mathbf{k}_-\cdot\hat{t}_p')\frac{\sin\frac{1}{2}\Delta\mathbf{k}_-\cdot\hat{k}_pL}{\Delta\mathbf{k}_-\cdot\hat{k}_p}\\ &\times\delta(2\omega_{0p}-\omega_{0-}-\omega_{e+})\,,\end{aligned}\tag{20}$$

where in Eq. (20) and what follows

$$\begin{aligned}\omega_R&=\omega_{e+}-\omega_{0p}\,,\\ \mathbf{k}_R\cdot\hat{k}_p&=[n_0^2(\omega_R)\omega_R^2/c^2-(\mathbf{k}_+\cdot\hat{t}_p)^2-(\mathbf{k}_+\cdot\hat{t}_p')^2]^{1/2}\,,\\ \Delta\mathbf{k}_\pm\cdot\hat{k}_p&=(\mathbf{k}_p-\mathbf{k}_\pm)\cdot\hat{k}_p\pm\mathbf{k}_R\cdot\hat{k}_p\,,\\ \Delta\mathbf{k}_-\cdot\hat{t}_p&=(\mathbf{k}_-+\mathbf{k}_+)\cdot\hat{t}_p\,,\\ \Delta\mathbf{k}_-\cdot\hat{t}_p'&=(\mathbf{k}_-+\mathbf{k}_+)\cdot\hat{t}_p'\,.\end{aligned}\tag{21}$$

Since we will generally be dealing with narrow beams, we neglected in Eq. (20) the $\mathbf{k}$ dependence in $\Delta\epsilon_0$ and $\Delta\epsilon_e$.

Since the transition probability per unit time (T) per unit cross-sectional area (A) perpendicular to $\hat{k}_p$ is

$$|\phi_{\text{final}}^\dagger S^{(2)}\phi_{\text{in}}|^2/AT\,,$$

the corresponding flux $dN(\mathbf{k}_+)$ of up-converted photons spontaneously emitted into the differential solid angle $d\Omega_+$ and bandwidth $d\omega_{e+}$ is then from Eq. (20) after

[13] H. Robl, in *Proceedings of the Third International Congress of Quantum Electronics*, edited by P. Grivet and N. Bloembergen (Columbia University Press, New York, 1964).

[14] A. Grinberg and N. I. Kramer, Fiz. Tverd. Tela **8**, 1555 (1966); **10**, 2022 (1968) [English transls.: Soviet Phys.—Solid State **8**, 1235 (1966); **10**, 1573 (1969)]; A. Grinberg and S. Ryvkin, Zh. Eksperim. i Teor. Fiz. Pis'ma v Redaktsiyu **7**, 324 (1968) [English transl.: JETP Letters **7**, 253 (1968)]; D. Klyshko, *ibid.* (to be published).

[15] See, for example, Ref. 11, p. 260.

carrying out the integration in $\mathbf{k}_-$:

$$dN(\mathbf{k}_+)=\int \frac{|\phi_{\text{final}}{}^\dagger S^{(2)}\phi_{\text{in}}|^2}{AT}\frac{n_e{}^3(\mathbf{k}_+)\omega_{e+}{}^2}{c^3}d\mathbf{k}_-d\omega_{e+}d\Omega_+$$

$$=\bar{N}\frac{1-\cos\Delta k_-L}{(\Delta k_-)^2}\left[\frac{\sin^2(\frac{1}{2}L\Delta\mathbf{k}_+\cdot\hat{k}_p)}{(\Delta\mathbf{k}_+\cdot\hat{k}_p)^2}+\frac{4\sin^4(\frac{1}{4}L\Delta\mathbf{k}_+\cdot\hat{k}_p)}{(\Delta\mathbf{k}_+\cdot\hat{k}_p)^2}\right]d\omega_{e+}d\Omega_+, \quad (22)$$

where

$$\bar{N}=\frac{4\Delta\epsilon_0{}^2\Delta\epsilon_e{}^2\hbar^2\omega_{0p}{}^2\omega_{e+}{}^3\omega_-\omega_R{}^4n_e(\mathbf{k}_+)N_p(N_p-1)}{\pi c^8n_0{}^4(\omega_{0p})n_0(\omega_-)(\mathbf{k}_R\cdot\hat{k}_p)^2}, \quad (23)$$

$$\Delta k_-=|\mathbf{k}_p|-[n_0{}^2(\omega_-)\omega_-{}^2/c^2-(\mathbf{k}_+\cdot t_p)^2-(\mathbf{k}_+\cdot t_p{}')^2]^{1/2}-\mathbf{k}_R\cdot\mathbf{k}_p, \quad (24a)$$

$$\omega_-=2\omega_{0p}-\omega_{e+}, \quad (24b)$$

with the help of Eqs. (6a) and (6b).

These, Eqs. (22)–(24b), contain all the information on the intensity of the spontaneous emission at the sum frequency of the up-conversion process. We consider in Sec. III the physical consequences of this result.

III. SPONTANEOUS EMISSION

A. Spectral and Angular Characteristics

The spectral and angular distributions of the spontaneous emission described by Eq. (22) are primarily determined by the terms in the brackets which add up to

$$4\sin^2(\tfrac{1}{4}L\Delta\mathbf{k}_+\cdot\hat{k}_p)/(\Delta\mathbf{k}_+\cdot\hat{k}_p)^2. \quad (25a)$$

This is, of course, a sharply peaked function which in the limit of $L\to\infty$ is proportional to

$$\sim L\delta(\Delta\mathbf{k}_+\cdot\hat{k}_p). \quad (25b)$$

It is clear then that the peak of the spontaneous emission follows the phase-matching characteristic $\Delta\mathbf{k}_+=0$, or equivalently the equation

$$G=0, \quad (26a)$$

where

$$G=\omega_{e+}-\omega_{0p}-[1/n_0(\omega_R)][n_e{}^2(\mathbf{k}_+)\omega_{e+}{}^2+n_0{}^2(\mathbf{k}_p)\omega_{0p}{}^2-2n_e(\mathbf{k}_+)n_0(\mathbf{k}_p)\omega_{e+}\omega_{0p}(\hat{k}_+\cdot\hat{k}_p)]^{1/2}. \quad (26b)$$

Equation (26a) is also the phase-matching condition that determines the tuning characteristics of the first-order up-conversion process.

In the case of the up-conversion process, for a given visible beam of frequency ω_+ collinear with a pump beam of frequency $\omega_{0p}=\omega_p$, which we assume to be monochromatic, the solution of Eq. (26a) with $\hat{k}_+\cdot\hat{k}_p=1$ gives the orientation of the pump beam relative to the crystal optic axis required to achieve efficient up conversion of the infrared to the visible. Figure 2 gives examples of such collinear tuning characteristics: $LiNbO_3$ at room temperature pumped by the 6943 Å output of the ruby laser and the 1.06-μ output of the YAG:Nd^{3+} laser. The turn-around near the long-wavelength edge of each tuning curve is due to the anomalous dispersion of the corresponding infrared beyond $\sim 5\ \mu$. The index of refraction data were taken from Refs. 16 and 17. Since these tuning curves are very sensitive to the indices of refraction involved, there may be some small differences between these computed results and measured results for particular crystals.

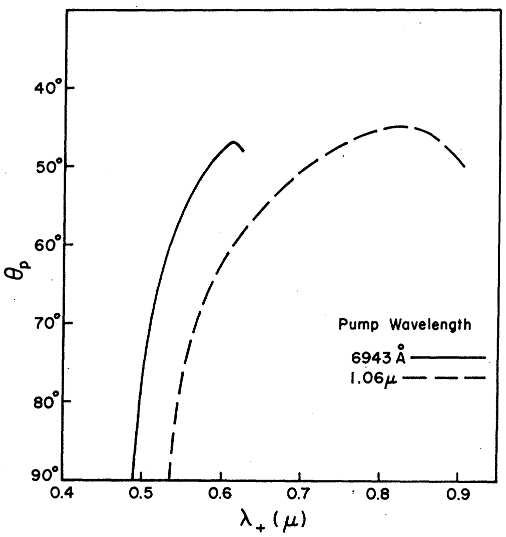


FIG. 2. Collinear tuning characteristics of $LiNbO_3$ at room temperature for the first-order up-conversion process. λ_p is the pump wavelength; λ_+ is the wavelength corresponding to the sum frequency; and θ_p is the angle between the pump ray and the optic axis of the crystal.

For a fixed pump beam frequency and orientation relative to the crystal optic axis, the solution of Eq. (26a) with $\hat{k}_+\cdot\hat{k}_p\leq 1$ gives also the required orientation of the sum-frequency beam (and hence the infrared beam) relative to the pump beam for a given sum frequency (or infrared frequency). Figure 3 gives examples of such noncollinear tuning characteristics. The general features of these collinear and noncollinear tuning characteristics are similar to those of the spontaneous down-conversion process, since the basic phase-matching equations are the same [Eq. (26a) is the same as Eq. (29) of Ref. 8 with the roles of the pump beam and signal beam reversed]. The tuning characteristics for the spontaneous down-conversion process have already been studied and discussed extensively previously.[8,9] The angular range for noncollinear interaction also defines the acceptance angle if the up-conversion process is used to convert two-dimensional images from the infrared to the visible.[5,6] As can be seen from Fig. 3, the noncollinear tuning range can be quite large.

[16] G. D. Boyd, R. C. Miller, K. Nassau, W. L. Bond, and A. Savage, Appl. Phys. Letters **5**, 234 (1964).

[17] W. S. Barker and R. Loudon, Phys. Rev. **158**, 433 (1967).

The frequency bandwidth for the first-order up-conversion process[1–6] is approximately

$$\Delta\omega_+(\hat{k}_+\cdot\hat{k}_p)=2\pi c/|n_e(\mathbf{k}_+)-n_0(\omega_R)+[\partial n_e(\mathbf{k}_+)/\partial\omega]\omega_+-[\partial n_0(\omega_R)/\partial\omega]\omega_R|L, \quad (27)$$

where ω_+ is equal to ω_{e+} that satisfies the phase-matching condition for the up-conversion process Eq. (26a). This follows from the condition that, for a well-collimated pump beam of ordinary wave, the total phase-mismatch in the direction $\hat{k}_p$ for the first-order process should be

$$|(\mathbf{k}_+-\mathbf{k}_p-\mathbf{k}_i)\cdot\hat{k}_p|L\lesssim\pi,$$

and assuming $\hat{k}_+\cdot\hat{k}_p\approx 1$. In this connection, it should be pointed out that the corresponding bandwidth for the spontaneous emission described by Eqs. (22) and (25a) is about twice that for the first-order process given above, Eq. (27). The extra spectral width is due to the virtual photons in the intermediate state.

To facilitate the discussions which are to follow, we show in Fig. 4 the subspace D in the phase space covered by a detector with a bandwidth $\Delta\omega_{\text{det}}$ and linear collection angle $\Delta\theta_{\text{det}}$ in the plane containing the crystal optic axis and $\hat{k}_p$. The shaded region R of width $\Delta\omega_+(\hat{k}_+\cdot\hat{k}_p)$ corresponds to the subspace for phase-matched first-order up-conversion process. To optimize the signal-to-noise ratio in the detection of low-level infrared radiation by the up-conversion process, one would generally make $\Delta\omega_{\text{det}}<\Delta\omega_+$ or make sure that D falls entirely in R in the case, of course, when the signal-to-noise ratio is limited by the spontaneous emission noise considered here.

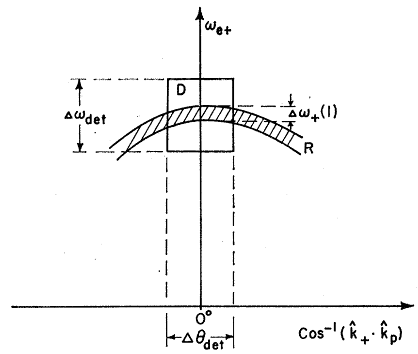


FIG. 4. Relationship between the subspace D covered by the detector and the subspace R corresponding to the phase-matched up-conversion process in the phase space for the spontaneous emission at the sum frequency ω_{e+}.

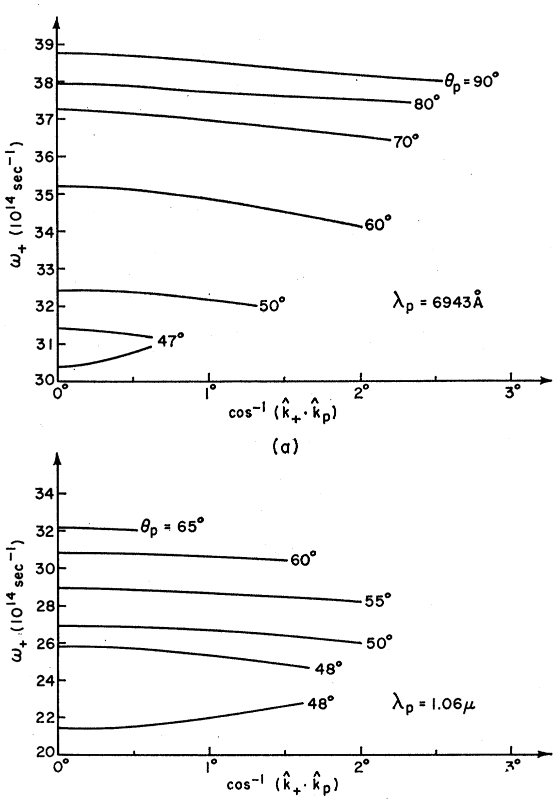


FIG. 3. Noncollinear tuning characteristics of $LiNbO_3$ pumped at (a) 6943 Å and (b) 1.06 μ.

B. Intensity Formulas

To calculate the total spontaneously emitted power P_{det} that would be measured by a detector with a finite aperture and bandwidth, we must integrate the flux density, Eq. (22), over the corresponding subspace in the phase space. Assuming that the subspace corresponding to the detector falls entirely in the subspace corresponding to the first-order up-conversion process, we have

$$P_{\text{det}}=\int_{D\cap R}\hbar\omega_{e+}A\,dN(\mathbf{k}_+). \quad (28)$$

In carrying out the integration in the frequency ω_{e+} and the direction of emission of the spontaneous emission over the detector bandwidth $\Delta\omega_{\text{det}}$ and collection angle $\Delta\Omega_{\text{det}}$, respectively, two possible situations must be considered. First, if the collection angle of the detector at the sum frequency is large in the sense

$$|\partial\Delta k_-L/\partial\theta_+|\Delta\theta_{\text{det}}=(L/c)|(\partial n_e/\partial\theta_+)\omega_+ +[n_e(\omega_+)-n_0(\omega_-)](\partial\omega_+/\partial\theta_+)|\Delta\theta_{\text{det}}\gg 2\pi, \quad (29)$$

then the $\cos\Delta k_-L$ factor in Eq. (22) will be averaged out upon integration of the direction of emission, or more specifically the direction θ_+ with respect to the crystal optic axis, over the collection angle of the detector $\Delta\Omega_{\text{det}}$. In this case, from Eqs. (22), (23), and (28), one obtains approximately

$$P_{\text{det}}=\frac{\Delta\epsilon_0^2\Delta\epsilon_e^2\hbar\omega_+^4\omega_-\omega_R^2P_P^2L^2\Delta\omega_{\text{det}}\Delta\Omega_{\text{det}}}{\pi c^8n_0^2(\omega_p)n_0^2(\omega_R)n_e(\mathbf{k}_+)n_0(\omega_-)(\Delta\bar{k}_-)^2A}, \quad (30a)$$

where P_P is the total pump power and $\Delta\Omega_{\text{det}}$ is the solid collection angle of the detector. In deriving P_{det}, Eq. (30), we have also ignored the slow variations of some of factors in the integrand in the subspace D; thus, in obtaining Eq. (30), we let $\mathbf{k}_R\cdot\hat{k}_p\cong n_0(\omega_R)\omega_R/c$,

$$\Delta k_-\cong\Delta\bar{k}_-\equiv 2|\mathbf{k}_p|-|\mathbf{k}_+|-[n_0(\omega_-)/c]\omega_-, \quad (30b)$$

and $\omega_{e+}=\omega_+$ which satisfies Eq. (26a). We have also replaced the terms in the brackets in Eq. (22), or the factor shown in (25a), by its asymptotic form $\frac{1}{4}L^2$, which is valid near $\Delta\mathbf{k}_+\cdot\hat{k}_p\to 0$. If $\Delta\omega_{\text{det}}\geqq 2\Delta\omega_+$, to obtain the total integrated spontaneous intensity, $\Delta\omega_{\text{det}}$ in (30a) must be replaced by $2\Delta\omega_+$ defined in Eq. (27).

If, on the other hand, the collection angle of the detector is small in the sense that the inequality in Eq. (29) is far from being satisfied,

$$|\partial\Delta k_-L/\partial\theta_+|\Delta\theta_{\text{det}}=(L/c)|(\partial n_e/\partial\theta_+)\omega_+ +[n_e(\omega_+)-n_0(\omega_-)](\partial\omega_+/\partial\theta_+)|\Delta\theta_{\text{det}}\ll 2\pi, \quad (31a)$$

and if the detector bandwidth is also relatively narrow in the sense that

$$|\partial\Delta k_-L/\partial\omega_+|\Delta\omega_{\text{det}} =(L/c)|n_e(\omega_+)-n_0(\omega_-)|\Delta\omega_{\text{det}}\ll 2\pi, \quad (31b)$$

then the total spontaneously emitted power measured will be

$$P_{\text{det}}=\frac{(1-\cos\Delta\bar{k}_-L)\Delta\epsilon_0{}^2\Delta\epsilon_e{}^2\hbar\omega_+{}^4\omega_-\omega_R{}^2P_P{}^2L^2\Delta\omega_{\text{det}}\Delta\Omega_{\text{det}}}{\pi c^8n_0{}^2(\omega_p)n_0{}^2(\omega_R)n_e(\mathbf{k}_+)n_0(\omega_-)(\Delta\bar{k}_-)^2A}. \quad (32)$$

In this case, there is a periodic variation of the measured spontaneous intensity with L. By carefully choosing the operating parameters involved, it should be possible to satisfy both Eqs. (31a) and (31b) for given $\Delta\omega_{\text{det}}$ and $\Delta\theta_{\text{det}}$ and the requirement that only either the ordinary or the extraordinary wave in the intermediate state can be excited. It is then possible to eliminate the spontaneous emission considered here from the detected output of the up-conversion process by making $\Delta\bar{k}_-L$ equal to some even multiples of π. However, other considerations[6] may prevent one from operating with these particular restrictions.

Equations (30a) and (32) are our final results. It is of particular interest to note that Eq. (30a) is exactly the same as the corresponding result obtained by Smith and Townes classically.[18] This provides a needed check on the physical assumptions that must be introduced in the simple classical picture on the one hand and a check on the more involved quantum-mechanical calculation on the other. The present formulation of the problem also provides a convenient starting point for future systematic extension of the theory. As for the noise observed by Midwinter and Warner[2] in the up-conversion process, it has already been shown by Smith and Townes on the basis of the classical results that it could not have been due to the noise process considered here and was probably due to some of the other sources suggested by Midwinter and Warner; the noise observed was, therefore, probably not inherent to the up-conversion process.

ACKNOWLEDGMENTS

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[18] In making the comparison, one should note in particular the differences in the notations: ϵ_1 of Ref. 7 corresponds to our $4\pi 2d_{ijk}$ here, where d_{ijk} is the nonlinear coefficient according to Kleinman's (see Ref. 10) definition; $2\pi\Delta\nu$ in Ref. 7 corresponds to our $\Delta\omega_{\text{det}}$ here, etc.