Symmetrical Formulation of the Quantum Theory of Three-Wave Optical Parametric Interactions*

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The quantum theory of three-wave optical parametric interactions in a crystal such as $LiNbO_3$ is formulated in a symmetrical manner with respect to interaction time and distance. The formalism leads to a unique treatment of parametric interactions of both cavity type and propagation type, in that the resemblance between time in the cavity type and distance in the propagation type of interaction in the classical description is preserved. Three-wave effects of harmonic generation, parametric amplification (including spontaneous emission), and frequency conversion are treated in a unique and symmetric manner. Using Feynman's operator techniques, quantum states resulting from parametric interactions, previously unavailable, are derived, from which characteristics of the processes are deduced.

I. INTRODUCTION

IN the past decade, two kinds of three-wave parametric interactions at microwave frequencies have been intensively investigated[1]: cavity type and propagation type. The cavity type of interaction treats standing waves whose spatial distributions are determined by the boundary conditions of the resonant structure; the wave amplitudes develop in time. The propagation type of interaction treats traveling waves which are stationary in time; the wave amplitudes develop as they propagate in a specific direction in space. Recently, significant progress has been made on parametric interactions at optical frequencies.[2]

Classically, there exists a formal resemblance between the two types of interactions since time in one case plays the role of distance in the other. This resemblance, noticed by previous workers,[3] is to be expected, since the two types of parametric interactions differ only in their boundary conditions but arise from the same physical process. The quantum theory of parametric interactions due to Louisell *et al.* and others[4] uses the Schrödinger formulation (in either Heisenberg picture or Schrödinger picture) under which the dynamics of the system propagates in time, and appears to be adequate for the cavity-type interactions. Recent optical experiments by Harris *et al.* and Magde *et al.*[5] on spontaneous parametric emission can only be explained as propagation-type effects. Plausibility arguments can be developed which explain the observed spontaneous emission as arising from the amplification of one noise quantum per idler mode by a pump wave.[6] This approach, although useful, is confusing and leaves unanswered questions about the meaning and origin of these noise photons, and about the specification of the quantum states after the parametric interaction. This paper attempts to clarify these points by treating the problem in a unified manner.

In this paper, the quantum theory of three-wave parametric interactions is formulated in a manner that is symmetrical upon an interchange of time and distance (divided by the propagation velocity). We assume that the pump is a traveling wave and that the induced fields are either standing waves or traveling waves corresponding to the interaction in a nonlinear dielectric crystal with or without end coatings. The interaction will be described as the simultaneous annihilation of one photon and creation of two other photons. Harmonic generation,[7] parametric amplification, and frequency conversion[1] are included within the single format. For these physical situations of interest, we shall point out that the time-ordered transformation, associated with the solution to the Schrödinger equation (in the interaction picture) may be recast into a distance-ordered form suitable for handling the propagation type of interactions. It will be shown that the classical resemblance between time in the cavity type and distance in the propagation type of parametric interactions is preserved in the present quantum formulation of the theory. Using the operator tech-

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[1] H. Suhl, J. Appl. Phys. **28**, 1225 (1957); H. Heffner and G. Wade, *ibid.* **29**, 1321 (1958); P. K. Tien, *ibid.* **29**, 1347 (1958); A. van der Ziel, *ibid.* **19**, 999 (1948).

[2] J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. **127**, 1918 (1962); N. M. Kroll, *ibid.* **127**, 1207 (1962); C. C. Wang and C. W. Racette, Appl. Phys. Letters **8**, 169 (1965); J. A. Giordmaine and R. C. Miller, Phys. Rev. Letters **14**, 973 (1965); G. D. Boyd and A. Ashkin, Phys. Rev. **146**, 187 (1966).

[3] W. H. Louisell, *Coupled Mode and Parametric Electronics* (John Wiley & Sons, Inc., New York, 1960); Y. R. Shen, Phys. Rev. **155**, 921 (1967).

[4] W. H. Louisell, A. Yariv, and A. E. Siegman, Phys. Rev. **124**, 1646 (1961); W. G. Wagner and R. W. Hellwarth, *ibid.* **133**, A915 (1964); B. R. Mollow and R. Glauber, *ibid.* **160**, 1076 (1967); T. G. Giallorenzi and C. L. Tang, *ibid.* **166**, 225 (1968).

[5] S. E. Harris, M. K. Oshman, and R. L. Byer, Phys. Rev. Letters **18**, 732 (1967); D. Magde and H. Mahr, *ibid.* **18**, 905 (1967).

[6] R. L. Byer and S. E. Harris, Phys. Rev. **168**, 1064 (1968); R. G. Smith, J. G. Skinner, J. E. Geusic, and W. G. Nilsen, Appl. Phys. Letters **12**, 97 (1968).

[7] P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich, Phys. Rev. Letters **7**, 118 (1961); D. A. Kleinman, Phys. Rev. **128**, 1761 (1962); P. D. Maker, R. W. Terhune, M. Nisenoff, and C. M. Savage, Phys. Rev. Letters **8**, 21 (1962).

niques of Feynman,[8] properties of parametric interactions and quantum states after the interaction are analyzed in detail.

II. FORMULATION OF THE PROBLEM

To formulate a quantum-mechanical problem in the interaction picture, one has to determine for the process (1) an interaction Hamiltonian (or equivalently Lagrangian) density operator, subject to a set of commutation relations, and (2) a transformation law which describes the evolution of the interacting system.

We are concerned with electromagnetic fields in a dielectric medium, such as crystal KDP or $LiNbO_3$. For parametric interactions, the medium undergoes a virtual transition from ground state to ground state (via intermediate states). The detailed dynamics of the virtual transition is bypassed and its effects on electromagnetic fields are represented as usual[2,4] by macroscopic polarizations through frequency-dependent susceptibilities. For our purpose, there are two terms in the polarization that are of importance: A linear term accounts for the change in index of refraction from its vacuum value and a nonlinear term, represented by susceptibility tensor χ, serves as a source for three-wave mixing. We quantize the electromagnetic waves in a space of volume V and expand the fields into the normal modes of Maxwell's equations including the linear polarization of the medium to accounts for anisotropy and dispersion of the system. The electric field can then be expressed in Gaussian units as

$$\mathbf{E}(\mathbf{r},t)=i(4\pi)^{1/2}\sum_{\lambda} n_\lambda^{-1}(\tfrac{1}{2}\hbar\omega_\lambda)^{1/2} \times[a_\lambda \exp(-i\omega_\lambda t)\mathfrak{u}_\lambda(\mathbf{r})-a_\lambda^\dagger \exp(i\omega_\lambda t)\mathfrak{u}_\lambda^*(\mathbf{r})], \quad (1)$$

where $\mathfrak{u}_\lambda(\mathbf{r})$'s are normal distributions polarized with respect to the principal axes of the crystal; $\mathfrak{u}_\lambda(\mathbf{r})=e_\lambda \exp(i\mathbf{k}_\lambda\cdot\mathbf{r})/\sqrt{V}$ for propagation modes and $\mathfrak{u}_\lambda(\mathbf{r})=e_\lambda[\exp(i\mathbf{k}_\lambda\cdot\mathbf{r})-\exp(-i\mathbf{k}_\lambda\cdot\mathbf{r})]/i(2V)^{1/2}$ for cavity modes. n_λ is the index of refraction which depends on polarization as well as frequency. The creation and annihilation operators $a_\lambda^\dagger$ and a_λ obey the usual commutation relations $[a_\lambda, a_\mu^\dagger]=\delta_{\lambda\mu}$.

Parametric interactions arise from the nonlinear term in the polarization mentioned above. The effect, represented phenomenologically by nonlinear susceptibility χ, may be described by a free energy term[2,9] which corresponds to the following Hamiltonian density for three-wave processes:

$$\mathcal{H}=i^3(2\pi)^{3/2}\chi_{\lambda\mu\sigma}\frac{(\hbar\omega_\lambda\hbar\omega_\mu\hbar\omega_\sigma)^{1/2}}{n_\lambda n_\mu n_\sigma} \times[a_\lambda a_\mu^\dagger a_\sigma^\dagger e^{i\Delta\omega t}\mathfrak{u}_\lambda(\mathbf{r})\mathfrak{u}_\mu^*(\mathbf{r})\mathfrak{u}_\sigma^*(\mathbf{r}) -a_\lambda^\dagger a_\mu a_\sigma e^{-i\Delta\omega t}\mathfrak{u}_\lambda^*(\mathbf{r})\mathfrak{u}_\mu(\mathbf{r})\mathfrak{u}_\sigma(\mathbf{r})], \quad (2)$$

where $\Delta\omega=\omega_\mu+\omega_\sigma-\omega_\lambda$. The three waves are characterized by wave vectors $\mathbf{k}_\lambda$, $\mathbf{k}_\mu$, $\mathbf{k}_\sigma$ and frequencies ω_λ, ω_μ, ω_σ; they are polarized in directions[2,5,7] which give nonzero χ, corresponding to experimental situations. The solution to the quantum-mechanical problem, involving the interaction described by (2), which transforms the state before, $|\psi_b\rangle$, to the state after the interaction $|\psi_a\rangle$, may be expressed[10] as

$$|\psi_a\rangle=\sum_{n=0}^{\infty}\left(\frac{-i}{\hbar}\right)^n(n!)^{-1}\int_{\Omega_0}^{\Omega}\cdots\int_{\Omega_0}^{\Omega}dV_1dt_1\cdots dV_ndt_nP \times\{\mathcal{H}(\mathbf{r}_1,t_1)\cdots\mathcal{H}(\mathbf{r}_n,t_n)\}|\psi_b\rangle, \quad (3)$$

where $P\{\ \}$ is the Dyson chronological product and the operators are to be evaluated according to their time order. The limit of integrations (Ω_0, Ω) cover the space-time region of the interaction: The entire volume $V=l_xl_yl_z$ and time interval $t=(-\frac{1}{2}T, \frac{1}{2}T)$ for the cavity type, and all values of y, z, t, and $x=(-\frac{1}{2}L, \frac{1}{2}L)$ for the propagation type of interactions. In practice, the choice of propagation direction x depends on the crystal used, the interaction time T is finite, and the interaction length L corresponding to the length of the dielectric crystal is much shorter than l_x.

The time-ordered expression of (3) is just what is needed for solving the cavity-type problems, and will give results identical to those obtained previously.[4] In order to handle the propagation-type problems and demonstrate the kind of symmetry of interest to this paper, we recall that there is a classical analogy between time in a cavity of the problem of coherent scattering and distance in a problem of steady-state propagation, and we assume that the same analogy holds in the quantum theory. This can be realized simply by evaluating (3) according to their distance order in the direction of propagation x. Although a rigorous mathematical derivation is outside the scope of this paper, the plausibility of using this procedure is intuitively evident. The condition under which (3) may be evaluated according to x ordering can be seen by examining the integrand of the third term in (3). Let $(\mathbf{r}_1, t_1)$ and $(\mathbf{r}_2, t_2)$ be abbreviated by (I) and (II), we may write out the time-ordered product of $\mathcal{H}(\mathbf{r}_1, t_1)$ and $\mathcal{H}(\mathbf{r}_2, t_2)$ as

$$\begin{aligned} P\{\mathcal{H}(\mathrm{I})\mathcal{H}(\mathrm{II})\} &=\mathcal{H}(\mathrm{I})\mathcal{H}(\mathrm{II}); && t_1>t_2,\ x_1>x_2\\ &\mathcal{H}(\mathrm{II})\mathcal{H}(\mathrm{I})+[\mathcal{H}(\mathrm{I}),\mathcal{H}(\mathrm{II})]; && t_1>t_2,\ x_2>x_1\\ &\mathcal{H}(\mathrm{II})\mathcal{H}(\mathrm{I}); && t_2>t_1,\ x_2>x_1\\ &\mathcal{H}(\mathrm{I})\mathcal{H}(\mathrm{II})+[\mathcal{H}(\mathrm{II}),\mathcal{H}(\mathrm{I})]; && t_2>t_1,\ x_1>x_2. \end{aligned}$$

This suggests that the t ordering can be replaced by the x ordering if the commutators in the above expression

[8] R. P. Feynman, Phys. Rev. **84**, 108 (1951).
[9] P. S. Pershan, Phys. Rev. **130**, 919 (1963).
[10] See, for example, F. Mandl, *Introduction to Quantum Field Theory* (Interscience Publishers, Inc., New York, 1959), Chap. 12.

vanish for problems in question. Physically, the parametric interactions of three traveling waves deal with events propagating forward along the unique direction x specified by $\Delta\mathbf{k}$, and thus it is only necessary to demonstrate that the above commutators, for the traveling waves so propagated, vanish. A direct mathematical proof should be possible, but we cannot consider it here. That $[\mathcal{H}(\mathrm{I}), \mathcal{H}(\mathrm{II})]=0$ can be visualized from the generally accepted fact that the commutator of two operators vanishes if observation of one does not effect observation of the other. With the forward propagation assumption, the effect of observing $\mathcal{H}(\mathrm{II})$, when $t_1>t_2$ can be felt at (I) only if x_1 is located ahead of x_2, i.e., $x_1>x_2$. Thus, $[\mathcal{H}(\mathrm{I}), \mathcal{H}(\mathrm{II})]=0$ if $x_2>x_1$ and $t_1>t_2$; the same is true if $x_1>x_2$ and $t_2>t_1$. This justification, although not mathematically rigorous, can be easily extended to deal with the nth term of (3); therefore, (3) may be evaluated according to x ordering when dealing with waves traveling forward along a unique direction.

When (3) is evaluated by the x ordering, the states before and after the interaction may be associated with the ordering parameter $x=-\frac{1}{2}L$ and $x=\frac{1}{2}L$. This is just what is needed to handle the propagation-type problems in a symmetrical manner. One can then view waves at one end of the crystal as "amplification" of the waves coming in from the other end.

Since we may evaluate (3) by either t ordering or x ordering, we shall rewrite (3) in the following form:

$$|\psi_a(\Omega)\rangle=\exp\left\{-\frac{i}{\hbar}\int_{\Omega_0}^{\Omega}\mathcal{H}(\mathbf{r},t)\,dV\,dt\right\}|\psi_b(\Omega_0)\rangle, \quad (4)$$

where Ω and Ω_0 are ordering parameters, denoting the order of operation and the states of interest. Equation (4) is time ordered or distance ordered to handle the cavity-type or the propagation-type problems, respectively. It is thus evident that the kind of classical resemblance between time and distance in the two types of parametric interactions is preserved in the present quantum description. It should be pointed out that although the justification of x ordering in evaluating (3) and (4) makes a symmetrical theory, the physics remains unchanged. The states before (initial) and after (final) the interaction, although labeled by $x=-\frac{1}{2}L$ and $x=\frac{1}{2}L$ are still time-dependent states of definite momenta. Our treatment is really equivalent to that of a scattering problem between states at $t=-\infty$ and at $t=\infty$, except that, the x ordering has the further advantages of making the coherence of the problem and the effective interaction interval in time $(-L/2v, L/2v)$ explicit.[11] Very recently, spontaneous parameteric emission of light has been treated as a scattering problem by Giallorenzi *et al.*[4] in a first-order calculation.

[11] The velocity of propagation in the x direction, v, sets the pace of interaction of traveling waves.

III. STATES OF THE INDUCED WAVES

We are interested in optical phenomena produced by a coherent laser beam, and we assume that the pump wave, denoted by index λ, is a traveling wave and that the induced waves, denoted by indices μ and σ, are either standing waves or traveling waves. The integration of the Hamiltonian density over y, z, and x for one case and t for the other in the exponent of (4) can be carried out directly; it gives obviously the conservation law of the process in question. The remaining one-dimensional integration with respect to the ordering parameter t or x specifies the order for operators.[8] The exponent of (4) may now be written as

$$-\frac{i}{\hbar}\int_{\Omega_0}^{\Omega}\mathcal{H}(\mathbf{r},t)\,dV\,dt=-i\int_{-\xi/2}^{\xi/2}(\kappa a_\lambda a_\mu^\dagger a_\sigma^\dagger e^{i\Delta\eta\xi'}+\kappa^* a_\lambda^\dagger a_\mu a_\sigma e^{-\Delta\eta\xi'})\,d\xi', \quad (5)$$

where[12]

$$\kappa=(1/\hbar\sqrt{V})(2\pi)^{3/2}\zeta[(\hbar\omega_\lambda\hbar\omega_\mu\hbar\omega_\sigma)^{1/2}/n_\lambda n_\mu n_\sigma]\delta_{\eta_\gamma,\eta_\mu+\eta_\sigma}.$$

In the expression, one notes that $\eta=\omega$, $\xi=T$, $\zeta=\chi$ for the cavity type, and that $\eta=k$, $\xi=L$, $\Delta k=|\mathbf{k}_\lambda-\mathbf{k}_\mu-\mathbf{k}_\sigma|$ and $\zeta=\chi/v$, where v is the velocity of interaction[11,12] in the x direction, for the propagation-type interactions. Written in this way, the operators in (4) and (5) are ξ ordered, which may be regarded as either t ordered or x ordered, and they operate according to time or distance depending on the individual situation.

Since the pump wave is produced by a single-mode laser, we may reasonably assume that it is initially in a Glauber state[13] $|\alpha\rangle_\lambda$, and (4) becomes

$$|\tfrac{1}{2}\xi\rangle_{\lambda\mu\sigma}=\exp\left\{-i\kappa^*\int_{-\xi/2}^{\xi/2}a_\lambda^\dagger a_\mu a_\sigma e^{-i\Delta\eta\xi'}d\xi'\right\}\times\exp\left\{-i\kappa\int_{-\xi/2}^{\xi/2}a_\lambda a_\mu^\dagger a_\sigma e^{i\Delta\eta\xi'}d\xi'\right\}|\alpha\rangle_\lambda|-\tfrac{1}{2}\xi\rangle_\mu|-\tfrac{1}{2}\xi\rangle_\sigma, \quad (6)$$

where the operators are ξ ordered. We may disentangle the pump operators, i.e., a_λ and $a_\lambda^\dagger$, first by the techniques of Feynman.[8] Using Eqs. (19) and (20) of Ref. 8, one can write (6) as

$$|\tfrac{1}{2}\xi\rangle_{\lambda\mu\sigma}=\exp(-i\beta^\dagger a_\lambda^\dagger)\times\exp(-i\beta a_\lambda)G_{00}|\alpha\rangle_\lambda|-\tfrac{1}{2}\xi\rangle_\mu|-\tfrac{1}{2}\xi\rangle_\sigma, \quad (7)$$

[12] We have used the relations

$$V^{-1}\int_V e^{i\mathbf{k}\cdot\mathbf{r}}dV=\delta_{0,k}$$

and

$$l_x^{-1}\lim_{\tau\to\infty}\int_{-\tau/2}^{\tau/2}e^{i\omega t}dt=v^{-1}\lim_{\tau\to\infty,\,l_x\to\infty}(v\tau/l_x)\delta_{0,\omega}=v^{-1}\delta_{0,\omega}.$$

[13] R. J. Glauber, Phys. Rev. **131**, 2766 (1963); W. H. Louisell, *Radiation and Noise in Quantum Electronics* (McGraw-Hill Book Co., New York, 1964).

where

$$\beta^\dagger = \kappa^* \int_{-\xi/2}^{\xi/2} a_\mu a_\sigma e^{-i\Delta\eta\xi'} d\xi',$$

and

$$G_{00} = \exp\left\{-|\kappa|^2 \int_{-\xi/2}^{\xi/2} a_\mu{}^\dagger a_\sigma{}^\dagger e^{i\Delta\eta\xi'} d\xi' \times \int_{-\xi/2}^{\xi'} a_\mu a_\sigma e^{-i\Delta\eta\xi''} d\xi''\right\}.$$

Equation (7) is position ordered[8,13] with respect to pump operators a_λ, $a_\lambda{}^\dagger$ and is still ξ ordered with respect to operators of the induced waves $a_{\mu'}a_\mu{}^\dagger$ and $a_{\sigma'}a_\sigma{}^\dagger$. The operation on the pump state in (7) can be carried out easily as one notices that $|\alpha\rangle_\lambda$ is an eigenstate[13] of a_λ. The density operator (or matrix) for the induced waves of interest can be expressed as

$$\begin{aligned}\rho_{\text{ind.}}(\tfrac{1}{2}\xi) &= \mathrm{Tr}_\lambda\{|\tfrac{1}{2}\xi\rangle_{\lambda\mu\sigma}\,{}_{\lambda\mu\sigma}\langle\tfrac{1}{2}\xi|\} \\ &= \pi^{-1}\int \langle\alpha'|\tfrac{1}{2}\xi\rangle\langle\tfrac{1}{2}\xi|\alpha'\rangle d^2\alpha' \\ &= \pi^{-1}\int d^2\alpha' \,|\langle\alpha'|\alpha\rangle|^2 G^\dagger(\alpha,\alpha')\,|-\tfrac{1}{2}\xi\rangle_{\mu\sigma} \\ &\quad \times {}_{\mu\sigma}\langle -\tfrac{1}{2}\xi|\,G(\alpha,\alpha'), \qquad (8)\end{aligned}$$

where

$$G^\dagger(\alpha,\alpha') = \exp(i\alpha'^*\beta^\dagger)\exp(-i\alpha\beta)G_{00}.$$

The result of (8) can be applied to evaluate properties of the induced waves, once given their initial states $|-\tfrac{1}{2}\xi\rangle_{\mu\sigma} = |-\tfrac{1}{2}\xi\rangle_\mu |-\tfrac{1}{2}\xi\rangle_\sigma$, although the disentangling of operator $G(\alpha,\alpha')$ is not very simple. The physical situation corresponding to experiments carried out so far[2,5,7] uses not only a coherent pump, but also a strong pump which cannot be depleted during the course of the interaction. For these cases, one can describe the pump as a classical traveling wave with a constant amplitude, and the discussion may be greatly simplified. In the following sections, we shall discuss the induced harmonic generation, parametric amplification (including spontaneous emission), and frequency conversion by assuming that the pump wave may be treated classically.

IV. INDUCED HARMONIC GENERATION

The interaction of a second harmonic with a classically describable primary may be dealt with by replacing a_λ by a_2, $a_\mu = a_\sigma$ by α_1, and correspondingly ω, n, $\mathbf{k}$ in (4) and (5). α_1 is a c number, denoting the classical amplitude of the primary. Since only the second harmonic is treated quantum mechanically, (4) becomes simply

$$|\tfrac{1}{2}\xi\rangle = \exp\left\{-i\int_{-\xi/2}^{\xi/2}(\kappa_2{}^*a_2{}^\dagger e^{-i\Delta\eta\xi'} + \kappa_2 a_2 e^{i\Delta\eta\xi'})d\xi'\right\}|-\tfrac{1}{2}\xi\rangle, \qquad (9)$$

where

$$\kappa_2{}^* = -(i/\hbar\sqrt{V})(2\pi)^{3/2}\zeta_2[(\hbar\omega_1)(\hbar\omega_2)^{1/2}/n_1{}^2 n_2] \times \alpha_1{}^2\delta_{\eta_2,2\eta_1}.$$

Equation (9) is readily disentangled[8] and it results in

$$|\tfrac{1}{2}\xi\rangle = \exp(-i\beta_2{}^*a_2{}^\dagger)\exp(-i\beta_2 a_2)\,|-\tfrac{1}{2}\xi\rangle e^{-I_2}, \qquad (10)$$

where

$$\begin{aligned}\beta^* &= \kappa_2{}^*\int_{-\xi/2}^{\xi/2} e^{-i\Delta\eta\xi'}d\xi' \\ &= \kappa_2{}^*\xi[\sin\tfrac{1}{2}(\Delta\eta\xi)/\tfrac{1}{2}(\Delta\eta\xi)] \\ &\equiv \kappa_2{}^*\xi f^{1/2}(\Delta\eta,\xi)\end{aligned}$$

and

$$I_2 = |\kappa_2|^2\int_{-\xi/2}^{\xi/2} e^{i\Delta\eta\xi'}d\xi'\int_{-\xi/2}^{\xi'} e^{-i\Delta\eta\xi''}d\xi''.$$

For second harmonic generation, the system is initially in the vacuum state, i.e., $|-\tfrac{1}{2}\xi\rangle = |0\rangle$, and (10) describing the state of second harmonics after the interaction becomes

$$|\tfrac{1}{2}\xi\rangle = \exp(-i\beta_2{}^*a_2{}^\dagger)\,|0\rangle e^{-I_2}. \qquad (11)$$

It is a Glauber state corresponding to a Poisson distribution in occupation space; it is a typical result of forced excitation[13,14] when the interaction is linear in the quantized field. This implies that the second harmonic generation under the present assumption is really a very coherent process. The power flow (Poynting vector) of the generated second harmonics can be obtained as

$$\begin{aligned}P_2 &= v(\hbar\omega_2/V)\langle\tfrac{1}{2}\xi|\,a_2{}^\dagger a_2\,|\tfrac{1}{2}\xi\rangle \\ &= v(\hbar\omega_2/V)\,|\kappa_2|^2\xi^2 f(\Delta\eta,\xi),\end{aligned}$$

where $v = c/n_2$. For the propagation-type interaction, the above expression may be related to primary power flow P_1 as

$$\begin{aligned}P_2 &= \left(\frac{2\pi}{c}\right)^3\frac{\omega_2{}^2}{n_2 n_1{}^2}|\chi_2|^2\left(\frac{c}{n_1}\frac{\hbar\omega_1}{V}|\alpha_1|^2\right)^2 L^2\left(\frac{\sin\frac{1}{2}(\Delta kL)}{\frac{1}{2}(\Delta kL)}\right)^2 \\ &= \left(\frac{2\pi}{c}\right)^3\frac{\omega_2{}^2}{n_2 n_1{}^2}|\chi_2|^2 L^2 f(\Delta k, L)P_1{}^2.\end{aligned}$$

This expression has been experimentally verified in many materials and is essentially the classical expression used by many authors.[2,7] If the dispersion due to linear polarization of the nonlinear material is such that $\Delta\eta = 0$, $f = 1$ and one deals with a matched situation.

The case of subharmonic generation however is quantum mechanically much more complicated. Even with a classical pump, the interaction is quadratic in subharmonic field. The forced excitation, in this case,

[14] R. P. Feynman, Phys. Rev. 80, 440 (1950).

may not generate field describable by a coherent state. Although the problem can be handled in similar manner, we shall not include it here.

V. PARAMETRIC AMPLIFICATION AND FREQUENCY CONVERSION

In this section, we consider interactions between a strong pump wave to be treated classically, and a weak signal wave and a weak idler wave which shall be treated quantum mechanically. If the pump photon has higher energy, the interaction gives rise to parametric amplification. For this case, we replace a_λ, a_μ, a_σ by α_p, a_s, a_i and, respectively, the indices of the parameters ω, n, $\mathbf{k}$ in (5). α_p is a c number, denoting classical pump amplitude. Equation (4) describing the quantum state for signal and idler waves becomes

$$|\tfrac{1}{2}\xi\rangle_{\rm si}=\exp\left\{-i\int_{-\xi/2}^{\xi/2}(\kappa_p{}^*a_s{}^\dagger a_i{}^\dagger e^{i\Delta\eta\xi'}+\kappa_pa_sa_ie^{-i\Delta\eta\xi'})d\xi'\right\}|-\tfrac{1}{2}\xi\rangle_{\rm si},\quad(12)$$

where

$$\kappa_p{}^*=(i/\hbar\sqrt{V})(2\pi)^{3/2}\zeta_p[(\hbar\omega_p\hbar\omega_s\hbar\omega_i)^{1/2}/n_pn_sn_i]\times\alpha_p\delta_{\eta_p,\eta_s+\eta_i}.$$

We shall limit our discussions to the following two cases only: (1) spontaneous parametric emission for which both signal and idler are assumed to be in vacuum states initially, and (2) parametric amplification for which the signal is in a Glauber state and the idler is in the vacuum state initially. To deal with these situations, we use Feynman's techniques again and we disentangle the idler operators first. Equation (12) becomes

$$|\tfrac{1}{2}\xi\rangle_{\rm si}=\exp(-i\beta_p{}^\dagger a_i{}^\dagger)\exp(-i\beta_pa_i)G_{00}{}^p|-\tfrac{1}{2}\xi\rangle_{\rm si},\quad(13)$$

where

$$\beta_p{}^\dagger=\kappa_p{}^*\int_{-\xi/2}^{\xi/2}a_s{}^\dagger e^{i\Delta\eta\xi'}d\xi';$$

$$G_{00}{}^p=\exp\left\{-|\kappa_p|^2\int_{-\xi/2}^{\xi/2}a_se^{-i\Delta\eta\xi'}d\xi'\int_{-\xi/2}^{\xi'}a_s{}^\dagger e^{i\Delta\eta\xi''}d\xi''\right\}$$

and the idler operators are position ordered and the signal operators are ξ ordered. Since the idler is initially in the vacuum, operation on idler mode may be carried out and (13) becomes

$$|\tfrac{1}{2}\xi\rangle_{\rm si}=\sum_n|n\rangle_i[(-i\beta_p{}^\dagger)^n/(n!)^{1/2}]G_{00}{}^p|-\tfrac{1}{2}\xi\rangle_s,\quad(14)$$

where $|n\rangle_i$ is the state containing n idler photons and $|-\xi/2\rangle_s$ is the initial state of the signal wave. Equation (14) contains only ξ-ordered signal operators and they can be further disentangled by the same techniques. When this is done, we have

$$|\tfrac{1}{2}\xi\rangle=\sum_n|n\rangle_i\exp[-I(\xi/2)a_sa_s{}^\dagger](1/\sqrt{n!})\times\left\{-i\kappa_p{}^*\int_{-\xi/2}^{\xi/2}a_s{}^\dagger e^{-I(\xi')}e^{i\Delta\eta\xi'}d\xi'\right\}^n|-\tfrac{1}{2}\xi\rangle_s$$
$$=\sum_n|n\rangle_ie^{-I(\xi/2)}\exp[-I(\xi/2)a_s{}^\dagger a_s](1/\sqrt{n!})\times\{Ja_s{}^\dagger\}^n|-\tfrac{1}{2}\xi\rangle_s,\quad(15)$$

where

$$I(\xi')=|\kappa_p|^2\int_{-\xi/2}^{\xi'}e^{-i\Delta\eta\xi''}d\xi''\int_{-\xi/2}^{\xi''}e^{i\Delta\eta\xi'''}d\xi'''$$

and

$$J=-i\kappa_p{}^*\int_{-\xi/2}^{\xi/2}e^{-I(\xi')}e^{i\Delta\eta\xi'}d\xi'.$$

For spontaneous parametric emissions, we take $|-\frac{1}{2}\xi\rangle_s=|0\rangle_s$ and the density operator for signal wave ρ_s invoked from (15) and $|-\frac{1}{2}\xi\rangle_s=|0\rangle_s$ may be calculated. Knowing the trace of ρ_s is unity, we obtain

$$\rho_s(\tfrac{1}{2}\xi)=\mathrm{Tr}_i\{|\tfrac{1}{2}\xi\rangle\langle\tfrac{1}{2}\xi|\}=\sum_me^{-K}(|J|^2e^{-K})^m|m\rangle\langle m|=\sum_me^{-K}(1-e^{-K})^m|m\rangle\langle m|,\quad(16)$$

where

$$K=I(\tfrac{1}{2}\xi)+I^*(\tfrac{1}{2}\xi)=|\kappa_p|^2\left|\int_{-\xi/2}^{\xi/2}e^{i\Delta\eta\xi'}d\xi'\right|^2=|\kappa_p|^2\xi^2f(\Delta\eta,\xi).$$

The density matrix (16) describes a Bose-Einstein distribution,[13] and the emitted signal wave is thus "noise" in nature. The properties of this distribution have been discussed by Mollow and Glauber.[4] This is significant since (16) tells us many properties of spontaneous parametric emission including the emission intensity given in previous works.[5,6] The power per unit area, spontaneously emitted as the result of interaction between pump and one idler mode, may be calculated as

$$p_s=v(\hbar\omega_s/V)\;\mathrm{Tr}\{a_s{}^\dagger a_s\rho_s(\tfrac{1}{2}\xi)\}=v(\hbar\omega_s/V)(e^K-1),\quad(17)$$

where $v=c/n_s$.

This may be regarded as amplification of one noise photon per mode and it is the output noise power of an ideal amplifier.[13,15] For optical experiments, K is typically much less than unity and $e^K-1\simeq K$. This is in fact the assumption of Giallorenzi *et al.*[4] and Byer *et al.*[6] as they put the final idler waves in "one-photon"

[15] H. Heffner, Proc. IRE **50**, 1604 (1962).

eigenstates and the former used only the first-order scattering matrix in their calculation. For spontaneous parametric emissions pumped by a laser in a crystal such as $LiNbO_3$, one deals with a continuum of idler modes with essentially the same momentum each producing p_s given in (17). When $K \ll 1$, the incremental signal power flow due to idler modes $\mathbf{k}_i$ scattered into a capturing solid angle $d\Omega_{\text{cap}}$ is

$$dP_s = p_s[V/(2\pi)^3]k_i^2 dk_i d\Omega_{\text{cap}}$$
$$= v[\hbar\omega_s/(2\pi)^3]Kk_i^2 dk_i d\Omega_{\text{cap}}.$$

For the propagation-type parametric emission with a crystal of length L, we have

$$dP_s = v\frac{\hbar\omega_s}{(2\pi)^3}\left[\frac{(2\pi)^3}{\hbar^2}\frac{|\chi_p|^2\hbar\omega_i\hbar\omega_s}{v^2 c n_i^2 n_s^2 n_p}\frac{c}{n_p}\frac{\hbar\omega_p}{V}|\alpha_p|^2\right]L^2 \times\left(\frac{\sin\frac{1}{2}(\Delta kL)}{\frac{1}{2}(\Delta kL)}\right)^2 k_i^2 dk_i d\Omega_{\text{cap}}$$

$$= \left(\frac{\omega_s^2|\chi_p|^2}{c^3 n_p n_i n_s}L^2\right)P_p f(\Delta k, L)\left(\frac{c}{n_i}\hbar\omega_i\right)k_i^2 dk_i d\Omega_{\text{cap}}, \tag{18}$$

which is essentially Eq. (3) of Byer *et al.*[6] The total spontaneous power may be obtained from (18) by a simple integration. Since our aim is to obtain quantum states like (16) and to justify expressions like (18), we refer to Ref. 6 for further developments which are derivable from (18).

For parametric amplification, we take $|-\frac{1}{2}\xi\rangle_s = |\alpha_s\rangle_s = \exp(i\alpha_s^* a_s^\dagger)|0\rangle_s$. Substituting this last form into (15), one can show that the density matrix (16) for this case is

$$\rho_s(\tfrac{1}{2}\xi) = \sum_m e^{-K}(|J|^2 e^{-K})^m \exp(i\alpha_s^* e^{-I(\xi/2)a_s\dagger})|m\rangle\langle m| \times\exp(-i\alpha_s e^{[-I^*(\xi/2)a_s]}).$$

The induced signal power flow after interaction may be evaluated as

$$p_s = (c/n_s)(\hbar\omega_s/V)\,\mathrm{Tr}\{a_s^\dagger a_s\rho_s(\tfrac{1}{2}\xi)\}$$
$$= (c/n_s)(\hbar\omega_s/V)[(e^K-1)+|\alpha_s|^2 e^K]$$
$$= (c/n_s)(\hbar\omega_s/V)(e^K-1)+P_s e^K. \tag{19}$$

This expression differs from (17) by addition of a signal term which describes amplification of input signal with amplifier gain e^K. The noise introduced in the output is additive with a power level equal to that of an ideal amplifier.[15]

If the pump photon has energy lower than that of the signal or idler, one deals with frequency conversion. The treatment is similar to, and the results are somewhat different from that of parametric amplification discussed above. The question of the possibility of spontaneous emission deserves some discussion. Energy conservation suggests that a pump photon in this case does not have enough energy to "spontaneously split" into one signal photon plus one idler photon. The system should be left in the vacuum state after the interaction takes place, if neither signal nor idler was initially present. To see that this is true, we make substitutions in (5) for frequency conversion. The transformation which connects the states before and after the interaction is

$$|\tfrac{1}{2}\xi\rangle = \exp\left\{-i\int_{-\xi/2}^{\xi/2}(\kappa_c^* a_s a_i^\dagger e^{i\Delta\eta\xi'} + \kappa_c a_s^\dagger a_i e^{-i\Delta\eta\xi'})d\xi'\right\}|-\tfrac{1}{2}\xi\rangle,$$

where

$$\kappa_c^* = (i/\hbar\sqrt{V})(2\pi)^{3/2}\zeta_c[(\hbar\omega_p\hbar\omega_i\hbar\omega_s)^{1/2}/n_p n_i n_s] \times\alpha_p^*\delta_{\eta_p,\eta_s-\eta_i}.$$

Disentangling the idler operators and assuming that idler is initially in vacuum, one can obtain the state vector similar to (15) except that a_s and $a_s^\dagger$ are interchanged, i.e.,

$$|\tfrac{1}{2}\xi\rangle = \sum_n |n\rangle_i \exp[-I(\xi/2)a_s^\dagger a_s](1/\sqrt{n!}) \times\{Ja_s\}^n|-\tfrac{1}{2}\xi\rangle_s. \tag{20}$$

If the signal is also in the vacuum state initially, i.e., $|-\frac{1}{2}\xi\rangle_s = |0\rangle_s$, there is no signal photon to be annihilated and (20) becomes $|\xi/2\rangle = |0\rangle_i|0\rangle_s$. There can be no spontaneous emission in frequency conversion as expected.

VI. SUMMARY AND DISCUSSION

The quantum theory of three-wave optical parametric processes with the interaction Hamiltonian density given as usual, is proposed in a symmetrical manner with respect to time and distance. The formalism gives a unique treatment of both the cavity-type and the propagation-type parametric interactions. Preservation of classical resemblance between time in the former and distance in the latter is shown in the quantum theory.

Feynman's operator techniques are used to discuss states of waves induced by a strong and coherent pump. The state of the generated second harmonics is in a Glauber state corresponding to a very coherent interaction. The state of spontaneous parametric emission, however, gives a Bose-Einstein distribution in occupation corresponding to "noise." Since the system gain is typically very low at optical frequencies, the more exact expression reduces to what was usually assumed and to that previously arrived at from perturbation calculation. Parametric amplification of signal and frequency conversion have been discussed also. Here, intuitively expected results are studied by the symmetrical formulation proposed.

The success of the symmetrical formalism depends on the feasibility of evaluating (3) and (4) by means of x ordering; it is intuitively plausible. The arguments presented, although lacking mathematical rigor, have demonstrated and justified the validity of this procedure in dealing with traveling waves propagating along a unique forward direction. It is interesting to note that not only the parametric interactions considered here deal with waves traveling forward in a unique dimension, but most experiments in coherent nonlinear optics are in this situation.

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Spin–Lattice Coupling Constants of an Fe^{3+} Ion in MgO

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Estimates of the spin-lattice coupling constants of an Fe^{3+} ion in the host crystal MgO have been made. There are only two independent coupling constants C_{11} and C_{44} in the simple case of a cubic lattice. Various possible mechanisms contributing to the coupling constants in the point-charge model have been investigated. It has been shown that the Blume-Orbach mechanism is the dominant one among the mechanisms considered. The next most important contribution arises from the spin-spin interaction mechanism proposed by Pryce. The other mechanisms which we have considered are found to give an entirely negligible contribution to the coupling constants. The combined point-charge contributions arising from all the mechanisms are, in units of 10^{-13} cm/dyn, C_{11}(point-charge) $=+2.11$ and C_{44}(point-charge) $=-3.06$, as compared with the experimental results, in the same units, C_{11}(expt) $=+26$ and C_{44}(expt) $=-5.5$, due to Feher. Also, the estimated overlap contributions to C_{11} and C_{44} are found to be an order of magnitude less than the point-charge contributions. Finally, some suggestions have been made in order to bring the theoretical results into better agreement with the experimental results.

I. INTRODUCTION

IT has been shown earlier[1,2] that the zero-field splitting parameters D and E occurring in the spin Hamiltonian

$$H_S=D[3S_z^2-S(S+1)]+E(S_x^2-S_y^2) \tag{1}$$

can be explained reasonably well in the case of Mn^{2+} contained in ZnF_2 and MnF_2. The effects of the crystal fields at the site of the paramagnetic ion Mn^{2+} in the host lattice and the overlap due to the ligand ion wave functions were taken into account. It was concluded[1] that the dominant contribution arose from the Blume-Orbach[3] (BO) mechanism, which involves the first-order matrix element of the axial and rhombic fields, and second-order matrix elements of the spin-orbit interaction between excited quartet states which have been admixed into one another by the presence of the cubic field. The next most important mechanism was shown to be the spin-spin mechanism[4] (Pryce mechanism), linear in both spin-spin interaction and axial or rhombic crystal fields. The Orbach-Das-Sharma mechanism[4] (ODS) and the Watanabe mechanism[1,5] in the presence of cubic field (WC) follow the spin-spin mechanism in decreasing order of importance. The overlap contributions were also investigated[2] and found to be important.

The agreement between the theory and the experimental results for Mn^{2+} has prompted the analysis of the various mechanisms for Fe^{3+} present in different crystal symmetries. In this paper, the example of Fe^{3+} situated in the host crystal MgO distorted by an uniaxial stress is considered. The undistorted MgO crystal has a cubic lattice, and therefore by symmetry both D and E parameters vanish. However, one can create a noncubic environment about the paramagnetic-ion (Fe^{3+}) site by applying uniaxial stress, and therefore obtain nonvanishing parameters D and E. When the parameters D and E are expressed as linear functions of the applied stress, there are only two independent constants[6] of proportionality in the simple case of a cubic crystal. These are spin-lattice constants C_{11} and C_{44}. Experimental results are available for these con-

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