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Quantum Theory of the Traveling-Wave Frequency Converter

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The Hamiltonian describing parametric frequency conversion in a nonlinear dielectric is derived, and a complete solution for the electric fields present is obtained in the limit of an infinite medium. The qualitative features of the classical solution are recovered by considering the motion of wave packets.

I. INTRODUCTION

RECENTLY, simple quantum-mechanical models of the parametric amplifier and frequency converter have been proposed[1] and analyses of the quantum statistics based on these models have been made.[2–4] In these models the signal and idler fields are characterized by monochromatic modes. This adequately describes the standing-wave problem of two modes coupled in a cavity. However, in practice the parametric amplifier and frequency converter are traveling-wave devices. In a model which represents both input and output by monochromatic modes, it is impossible to impose physically meaningful initial conditions in a traveling-wave configuration.

Experimentally, a signal composed of photon wave packets is incident on the medium, and what is observed in the steady state is a spatial variation in the intensity of the incident beam and the output beam. This is easily accounted for classically[5] by simply taking the field amplitudes to depend on spatial coordinates in steady-state traveling-wave problems, instead of on time as in standing-wave configurations. However, quantum theory is much less versatile in this respect, the Heisenberg equations of motion involving only time variation. The result is that simple quantum-mechanical model calculations, such as those of Refs. 1–4, yield results in exact agreement with classical theory[5] in standing-wave configurations when the initial states of the fields are taken to be coherent, while in traveling-wave problems such agreement has only been obtained by strategic substitution of space for time variables in the final result. In this paper we derive a complete quantum-mechanical solution for a more realistic model of parametric frequency conversion in a nonlinear medium. Though this paper deals explicitly with the case of parametric frequency conversion, the method is equally applicable to parametric amplification. A mathematical simplication of the problem is accomplished by considering the medium to be infinite in extent. We shall attempt to provide some insight into the relationship between the quantum-mechanical and classical analyses. In particular, we will show how the spatial variation of the fields predicted classically may be recovered from the quantum-mechanical solution by considering the motion of wave packets through the medium.

II. HAMILTONIAN DESCRIBING PARAMETRIC FREQUENCY CONVERSION

The presence of an electromagnetic field in a dielectric causes a polarization of the medium. We assume that the polarization may be expanded in powers of the instantaneous electric field[5]

$$\mathbf{P}(r,t)=\chi\mathbf{E}(r,t)+\chi:\mathbf{E}(r,t)\mathbf{E}(r,t)+\cdots, \qquad (2.1)$$

where the first term defines the usual linear susceptibility and the second term the lowest-order nonlinear susceptibility. These susceptibility tensors are taken to depend only on frequency and direction of polarization, and to be spatially independent.

The interaction of the electromagnetic field with the dielectric medium may be described by the Hamiltonian

$$H_I=-\int\mathbf{E}\cdot\mathbf{P}d^3r$$
$$=-\int\mathbf{E}\cdot\chi\cdot\mathbf{E}d^3r-\int\mathbf{E}\cdot\chi:\mathbf{EE}d^3r. \qquad (2.2)$$

The electric field operator may be expanded in terms of normal modes as[6]

$$\mathbf{E}(r,t)=i\sum_k(\tfrac{1}{2}\hbar\omega_k)^{1/2}[a_k(t)\mathbf{u}_k(r)-a_k{}^\dagger(t)\mathbf{u}_k{}^*(r)], \qquad (2.3)$$

where $a_k(t)$ and $a_k{}^\dagger(t)$ are the usual annihilation and creation operators for the kth mode, obeying the com-

[1] W. H. Louisell, A. Yariv, and A. E. Siegman, Phys. Rev. **124**, 1646 (1961); W. H. Louisell, *Radiation and Noise in Quantum Electronics* (McGraw-Hill Book Co., New York, 1964).

[2] B. R. Mollow and R. J. Glauber, Phys. Rev. **160**, 1077 (1967).

[3] J. Tucker and D. F. Walls, Ann. Phys. (N. Y.) (to be published).

[4] R. Graham and H. Haken, Z. Physik **210**, 276 (1968); R. Graham, *ibid.* **210**, 319 (1968); **211**, 469 (1968).

[5] N. Bloembergen, *Non-linear Optics* (W. A. Benjamin, Inc., New York, 1965).

[6] R. J. Glauber, Phys. Rev. **131**, 2766 (1963).

mutation rules

$$[a_k(t),a_l^\dagger(t)]=\delta_{kl}\,,\qquad [a_k(t),a_l(t)]=[a_k^\dagger(t),a_l^\dagger(t)]=0\,. \tag{2.4}$$

The mode functions $\mathbf{u}_k(r)$ are solutions to the wave equation. The effects of the linear polarizability in a nondissipative medium may be accounted for by simply including a dielectric constant $\epsilon=1+4\pi\chi$.[7] The mode functions are thus taken to satisfy

$$(\nabla^2+\omega_k^2\epsilon_k/c^2)\mathbf{u}_k(r)=0 \tag{2.5}$$

with the orthonormality condition

$$\int(\epsilon_k\epsilon_l)^{1/2}\mathbf{u}_k^*(r)\cdot\mathbf{u}_l(r)d^3r=\delta_{kl}\,. \tag{2.6}$$

Their polarization is chosen so as to diagonalize the dielectric tensor, so that ϵ appears as a scalar.

The Hamiltonian for the field including the effects of the linear polarizability is then found to assume the same form as in the case of the free field

$$H_0=\sum_k \hbar\omega_k a_k^\dagger a_k\,. \tag{2.7}$$

The remaining term in the interaction (2.2) may now be considered as a perturbation on (2.7) describing a nonlinear coupling of the normal modes:

$$H_1=-\int \mathbf{E}\cdot\chi:\mathbf{E}\mathbf{E}d^3r\,. \tag{2.8}$$

From (2.5) and (2.6) the appropriately normalized mode functions are seen to be

$$\mathbf{u}_k(r)=\hat{e}_k\frac{1}{(V\epsilon_k)^{1/2}}e^{i\mathbf{k}\cdot\mathbf{r}}\,, \tag{2.9}$$

where

$$\omega_k=ck/\epsilon_k^{1/2}\,. \tag{2.10}$$

V is the normalization volume and ϵ_k a unit polarization vector, where for simplicity we drop the usual polarization indices. The expansion of the electric field (2.3) then becomes

$$\mathbf{E}(r,t)=i\sum_k\left(\frac{\hbar\omega_k}{2V\epsilon_k}\right)^{1/2}\hat{e}_k(e^{i\mathbf{k}\cdot\mathbf{r}}a_k(t)-e^{-i\mathbf{k}\cdot\mathbf{r}}a_k^\dagger(t))\,. \tag{2.11}$$

Substitution of this expression into the interaction (2.8) yields

$$H_1=\sum_{kk'k''}\kappa(\omega,\omega',\omega'')a_k^\dagger(t)a_{k'}(t)a_{k''}(t)\times\frac{1}{V}\int e^{-i(\mathbf{k}-\mathbf{k}'-\mathbf{k}'')\cdot\mathbf{r}}d^3r+\text{H.c.} \tag{2.12}$$

plus other terms corresponding to three-photon absorption and emission which are highly energy-non-conserving and so will be neglected. The coupling constant in (2.12) is

$$\kappa(\omega,\omega',\omega'')=-i\left(\frac{(\frac{1}{2}\hbar)^3\omega\omega'\omega''}{V\epsilon\epsilon'\epsilon''}\right)^{1/2}\hat{e}_k\cdot\chi:\hat{e}_{k'}\hat{e}_{k''}\,. \tag{2.13}$$

We now suppose that an intense, monochromatic laser beam of frequency ω_L is present in the medium and that it is the source of all non-negligible coupling. The Hamiltonian appropriate to the frequency conversion process we wish to describe may be obtained by setting k' or k'' equal to k_L in (2.12)

$$H_1=\sum_{kk'}2\kappa(\omega,\omega',\omega_L)a_k^\dagger(t)a_{k'}(t)a_L(t)\times\frac{1}{V}\int e^{-i(\mathbf{k}-\mathbf{k}'-\mathbf{k}_L)\cdot\mathbf{r}}d^3r+\text{H.c.} \tag{2.14}$$

This interaction describes processes in which a photon of frequency ω' is annihilated along with a quantum from the laser field to produce a photon at frequency ω, so that (2.14) describes "frequency conversion" effects. It may also be seen that the coupling described by this interaction is extremely complex even with the assumption of a purely monochromatic laser field. This complexity is greatly simplified in the limit of an infinite interaction volume. The mode function integral in (2.14) reduces in this case to a δ function, and the interaction Hamiltonian becomes

$$H_1=\sum_{kk'}2\kappa(\omega,\omega',\omega_L)a_k^\dagger(t)a_{k'}(t)a_L(t)\delta_{\mathbf{k},\mathbf{k}'+\mathbf{k}_L}+\text{H.c.} \tag{2.15}$$

The simplification which results from considering the case of an infinite medium is seen to be just the requirement of exact momentum conservation in each term of the interaction Hamiltonian

$$\mathbf{k}=\mathbf{k}'+\mathbf{k}_L\,. \tag{2.16}$$

Our subsequent analysis will be based on the Hamiltonian $H=H_0+H_1$ given by (2.7) and (2.15). In Sec. III, we obtain the solutions to the equations of motion for all field operators in the parametric approximation.

III. SOLUTIONS TO EQUATIONS OF MOTION

We have seen that all terms in the interaction Hamiltonian (2.15) conserve momentum. However, not all of these terms conserve energy. Terms which are substantially non-energy-conserving will oscillate rapidly and provide no significant coupling. Suppose, however, that we succeed in obtaining two modes, which we shall call the signal and idler, which not only satisfy the momentum-matching condition (2.16) but are energy-matched as well

$$\omega_I^0=\omega_S^0+\omega_L\,. \tag{3.1}$$

[7] Y. R. Shen, Phys. Rev. **155**, 921 (1967); C. Y. She, *ibid.* **176**, 461 (1968).

Quantities referring to these energy-matched modes will be denoted by the superscript 0. We take the propagation vectors of signal, idler, and laser modes to all lie in the z direction, so that (2.16) becomes

$$k_I^0 = k_S^0 + k_L \quad \text{or} \quad \omega_I^0 \epsilon_I^{0\frac{1}{2}}/c = \omega_S^0 \epsilon_S^{0\frac{1}{2}}/c + \omega_L \epsilon_L^{\frac{1}{2}}/c. \qquad (3.2)$$

Conditions (3.1) and (3.2) can be simultaneously satisfied, for example, in some uniaxial crystals by appropriate use of ordinary and extraordinary modes to cancel the effects of dispersion.[8]

From (2.7) and (2.15) it is seen that these modes are effectively coupled only to each other, and so may be treated independently. We ignore for the moment the other pairs of modes which are not frequency-matched. The laser mode is assumed to be in a coherent state and of sufficient intensity that we may neglect the reaction of the nonlinear coupling back on the state of this mode. In this parametric approximation, the field operators for the laser mode may be replaced by their expectation values

$$a_L(t) \to \alpha_L e^{-i\omega_L t}. \qquad (3.3)$$

The terms in the Hamiltonian which describe the frequency-matched modes may be then written as

$$H' = \hbar\omega_I^0 a_I^{0\dagger}(t) a_I^0(t) + \hbar\omega_S^0 a_S^{0\dagger}(t) a_S^0(t) + \hbar\kappa[a_I^{0\dagger}(t) a_S^0(t) e^{-i\omega_L t} + a_I^0(t) a_S^{0\dagger}(t) e^{+i\omega_L t}], \qquad (3.4)$$

where the coupling constant is given by

$$\hbar\kappa = -\left(\frac{(\frac{1}{2}\hbar)^2 \omega_I^0 \omega_S^0}{\epsilon_I^0 \epsilon_S^0}\right)^{1/2} E_L \hat{e}_I^0 \cdot \chi : \hat{e}_S^0 \hat{e}_L. \qquad (3.5)$$

In this expression, E_L represents the maximum amplitude of the electric field in the laser mode:

$$E_L = 2|i(\hbar\omega_L/2\epsilon_L V)^{1/2} \alpha_L|. \qquad (3.6)$$

The constant $\hbar\kappa$ has been taken to be real for convenience. This can always be accomplished by adjusting the arbitrary phases of the mode functions.

The Hamiltonian (3.4) is seen to be the model presented in Ref. 1 and treated in detail in Ref. 3. The only difference is that here we have derived an expression (3.5) for the coupling constant in terms of the parameters of the medium and the strength of the laser field. The solutions to the Heisenberg equations of motion are found to be[1,3]

$$\begin{aligned} a_I^0(t) &= e^{-i\omega_I^0 t}[a_I^0(0)\cos\kappa t - i a_S^0(0)\sin\kappa t], \\ a_S^0(t) &= e^{-i\omega_S^0 t}[a_S^0(0)\cos\kappa t - i a_I^0(0)\sin\kappa t]. \end{aligned} \qquad (3.7)$$

Now, however, we wish to go beyond this simple model and consider also modes which are very nearly frequency-matched, but which differ slightly in frequency from ω_I^0 or ω_S^0. Consider a mode with propagation vector in the z direction and of the same polarization as the signal mode, but with frequency

$$\omega_S = \omega_S^0 + \omega'. \qquad (3.8)$$

In the infinite medium, this mode will be coupled to one with frequency near that of the idler mode

$$\omega_I = \omega_I^0 + \omega. \qquad (3.9)$$

The relation between ω and ω' is determined by the momentum conservation condition (3.2). For small ω and ω', the relation is found to be

$$\omega = (v_I/v_S)\omega', \qquad (3.10)$$

where v_I and v_S are the group velocities $d\omega/dk$ evaluated at the idler and signal frequencies, respectively. Using the parametric approximation (3.3), the terms in the Hamiltonian involving these two modes are seen from (2.7) and (2.15) to be

$$H'' = \hbar\omega_S a_S^\dagger(t) a_S(t) + \hbar\omega_I a_I^\dagger(t) a_I(t) + \hbar\kappa[a_I^\dagger(t) a_S(t) e^{-i\omega_L t} + a_I(t) a_S^\dagger(t) e^{+i\omega_L t}]. \qquad (3.11)$$

The same coupling constant (3.6) appears again in (3.11), as we assume that the change in the nonlinear polarizability is negligible for very small deviations in frequency. The coupling terms in (3.11) no longer conserve energy since

$$\begin{aligned} \omega_I - \omega_S - \omega_L &= \omega - \omega' \\ &= \omega(1 - v_S/v_I) \\ &\equiv \Delta\omega. \end{aligned} \qquad (3.12)$$

The degree of mixing is found to depend on the amount of frequency mismatch $\Delta\omega$. The solutions to the Heisenberg equations of motion corresponding to (3.11) are found to be

$$\begin{aligned} a_I(t) = e^{-i(\omega_I - \frac{1}{2}\Delta\omega)t}\Bigg[& a_I(0)\Bigg(\cos([\kappa^2 + (\tfrac{1}{2}\Delta\omega)^2]^{1/2} t) - \frac{i\Delta\omega}{2[\kappa^2 + (\frac{1}{2}\Delta\omega)^2]^{1/2}} \sin([\kappa^2 + (\tfrac{1}{2}\Delta\omega)^2]^{1/2} t)\Bigg) \\ & - i a_S(0) \frac{\kappa}{[\kappa^2 + (\frac{1}{2}\Delta\omega)^2]^{1/2}} \sin([\kappa^2 + (\tfrac{1}{2}\Delta\omega)^2]^{1/2} t)\Bigg], \\ a_S(t) = e^{-i(\omega_S + \frac{1}{2}\Delta\omega)t}\Bigg[& a_S(0)\Bigg(\cos([\kappa^2 + (\tfrac{1}{2}\Delta\omega)^2]^{1/2} t) + \frac{i\Delta\omega}{2[\kappa^2 + (\frac{1}{2}\Delta\omega)^2]^{1/2}} \sin([\kappa^2 + (\tfrac{1}{2}\Delta\omega)^2]^{1/2} t)\Bigg) \\ & - i a_I(0) \frac{\kappa}{[\kappa^2 + (\frac{1}{2}\Delta\omega)^2]^{1/2}} \sin([\kappa^2 + (\tfrac{1}{2}\Delta\omega)^2]^{1/2} t)\Bigg]. \end{aligned} \qquad (3.13)$$

In the limit as $\Delta\omega \to 0$, we recover (3.7) for the case of

[8] J. A. Giordmaine and R. C. Miller, Phys. Rev. Letters **14**, 973 (1965); J. A. Giordmaine and R. C. Miller, *Physics of Quantum Electronics*, edited by P. L. Kelley, B. Lax, and P. E. Tannenwald (McGraw-Hill Book Co., New York, 1966); J. E. Midwinter and J. Warner, J. Appl. Phys. **38**, 519 (1967).

perfect energy matching. However, for $\Delta\omega \gg \kappa$, the solutions (3.13) become

$$a_I(t) \to a_I(0)e^{-i\omega_I t}, \quad a_S(t) \to a_S(0)e^{-i\omega_S t}, \tag{3.14}$$

just as for the free field. Therefore, the mixing becomes negligible for pairs of modes whose energy mismatch is large compared to the coupling constant.

The solutions to the equations of motion (3.13) completely characterize our model frequency converter. In the infinite medium the modes are coupled in pairs, and each of these two mode systems may be treated independently. A complete analysis of the quantum statistics for each of these pairs of coupled modes may be carried out along the lines described in Ref. 3. The solution for the general time-dependent P representation for the fields in a pair of such modes corresponding to the solutions (3.13) is given in Appendix A.

IV. FREQUENCY CONVERSION OF WAVE PACKETS

The solutions (3.13) of the equations of motion for the field operators provide, in principle, a complete quantum-mechanical solution to the problem of parametric frequency conversion in the limit of an infinite medium. We will now proceed to indicate the manner in which the well-known spatial variation of the signal and idler fields calculated classically and observed experimentally may be seen to emerge from this quantum-mechanical solution.

In the simplest quantum-mechanical model of parametric frequency conversion,[1,3] the signal and idler fields are both represented by monochromatic modes. The only part of the Hamiltonian considered in this case is (3.4) corresponding to the pair of modes which are perfectly phase-matched, satisfying the conditions (3.1) and (3.2). If we assume that the signal mode is initially in a coherent state of argument α_s and that the idler is in the vacuum state, then after time t there will be an electric field in the idler mode given by

$$\langle \mathbf{E}_I^+(z,t)\rangle = \langle \alpha_s,0|\, i\hat{e}_I\left(\frac{\hbar}{2\epsilon_I^0}\frac{\omega_I^0}{V}\right)^{1/2} a_I^0(t)e^{ik_I^0 z}|\alpha_s,0\rangle. \tag{4.1}$$

Using the solution for the time-dependent field operators (3.8), this is seen to be

$$\begin{aligned}\langle \mathbf{E}_I^+(z,t)\rangle &= \hat{e}_I\left(\frac{\hbar\omega_I^0}{2\epsilon_I^0 V}\right)^{1/2} \alpha_s \sin\kappa t\, e^{i(k_I^0 z-\omega_I^0 t)} \\ &= -i\hat{e}_I\left(\frac{\omega_I^0\epsilon_s^0}{\omega_s^0\epsilon_I^0}\right)^{1/2}\langle E_s^+(0,0)\rangle \\ &\qquad \times \sin\kappa t\, e^{i(k_I^0 z-\omega_I^0 t)}. \end{aligned}\tag{4.2}$$

This result is very similar to that obtained with the simplest sort of classical calculation describing parametric frequency conversion for perfectly phase-matched modes. In fact, the steady-state solution with the signal field incident at $z=0$ and otherwise ignoring boundary effects turns out to be just (4.2) with the substitution $t \to z/c$ in the argument of the sine.

We see that the quantum-mechanical solution for the idler electric field varies sinusoidally in time rather than in space and thus does not adequately describe a traveling-wave device. To enable the quantum-mechanical solution to describe a traveling-wave device, the above substitution of space for time variables is often advocated, and in some instances rather elaborate arguments have been used to support it.[7]

This problem inevitably arises in any simple quantum-mechanical treatment of a traveling-wave problem, in which each field is characterized by a single plane-wave mode. This is because the magnitude of the plane-wave mode function is constant over all space, and thus it is impossible to achieve the spatial variation found in classical solution or even impose physically meaningful initial conditions with such simple models. If, indeed, the electric field is expanded in terms of plane-wave modes, then the spatial variations which appear in traveling-wave situations can only be found in a quantum-mechanical solution by considering a large number of modes. A solution to the many-mode problem allows us to construct spatially localized wave packets at the initial time, and then to watch their behavior as they propagate through the medium. This avoids the unphysical feature of the single-mode calculation, in which it is necessary to specify some sort of fields existing throughout the medium before the interaction is "turned on."

We now proceed to give an example illustrating the manner in which the qualitative features of the classical solution emerge from a complete quantum-mechanical solution by considering the motion of wave packets through our model parametric frequency converter. We suppose that at time $t=0$ the idler field is in the ground state and that the electric field of the signal is comprised of a single wave packet centered at the origin. This field is nearly monochromatic, and is composed of a superposition of coherent states for modes of the field having frequencies near ω_s^0. The wave packet is defined formally by the expression[6]

$$|\{\beta(\omega')\}\rangle = \exp\left(\int \beta(\omega')a_{s\omega'}^\dagger(0)d\omega' - \text{H.c.}\right)|0\rangle, \tag{4.3}$$

so that the signal-field mode of frequency

$$\omega_s = \omega_s^0 + \omega' \tag{4.4}$$

is in a coherent state of argument $\beta(\omega')$. To be explicit, we take this distribution to be of the form

$$\beta(\omega') = \beta_0/(\omega'^2+\gamma^2), \tag{4.5}$$

where

$$\gamma \ll \omega_s^0. \tag{4.6}$$

The initial electric field is therefore given by

$$\langle \mathbf{E}_s^+(z,0)\rangle = \langle\{\beta(\omega')\}|i\hat{e}_s\int\frac{d\omega'}{2\pi}\left(\frac{\hbar(\omega_s^0+\omega')}{2\epsilon_s(\omega_s^0+\omega')V}\right)^{1/2}\times a_{s\omega'}(0)e^{i(k_s^0+k')z}|\{\beta(\omega')\}\rangle. \quad (4.7)$$

With the condition (4.6), we may approximate

$$\left(\frac{\hbar(\omega_s^0+\omega')}{2\epsilon_s(\omega_s^0+\omega')V}\right)^{1/2}\approx\left(\frac{\hbar\omega_s^0}{2\epsilon_s(\omega_s^0)V}\right)^{1/2}. \quad (4.8)$$

We may write $\exp[i(k_s^0+k')z]$ as $\exp[i\epsilon_s(\omega_s^0+\omega')\times((\omega_s^0+\omega')z/c)]$. If we expand $\epsilon_s(\omega_s^0+\omega')$ in a Taylor series in the small frequency shift ω' and consider only terms linear in ω', we find

$$\exp[i\epsilon_s(\omega_s^0+\omega')((\omega_s^0+\omega')z/c)]\approx\exp(\omega'z/v_s). \quad (4.9)$$

With the above two approximations, Eq. (4.7) becomes

$$\langle \mathbf{E}_s^+(z,0)\rangle = i\hat{e}_s\left(\frac{\hbar\omega_s^0}{2\epsilon_s^0V}\right)^{1/2}e^{ik_s^0z}\int\frac{d\omega'}{2\pi}e^{i\omega'z/v_s}\beta(\omega')$$

$$= i\hat{e}_s\frac{\beta_0}{2\gamma}\left(\frac{\hbar\omega_s^0}{2\epsilon_s^0V}\right)^{1/2}e^{-\gamma|z|/v_s}e^{ik_s^0z}, \quad (4.10)$$

where $v_s=(\partial\omega/\partial k)|_{\omega=\omega_s^0}$ is the group velocity.

We now wish to examine the field generated in the idler mode after time t. As shown in Sec. III, each mode of the signal field at frequency ω_s given by (4.4) is coupled to a mode of the idler field of frequency

$$\omega_I=\omega_I^0+\omega, \quad (4.11)$$

where the relation between ω and ω' is given by (3.10). The electric field in the idler mode at time t is thus seen to be

$$\langle \mathbf{E}_I^+(z,t)\rangle = \langle\{\beta(\omega')\}|i\hat{e}_I\int\frac{d\omega}{2\pi}\left(\frac{\hbar(\omega_I^0+\omega)}{2\epsilon_I(\omega_I^0+\omega)V}\right)^{1/2}\times a_{I\omega}(t)e^{i(k_I^0+k)z}|\{\beta(\omega')\}\rangle. \quad (4.12)$$

Substituting $a_{I\omega}(t)$ from the solutions to the equations of motion (3.11) yields

$$\langle \mathbf{E}_I^+(z,t)\rangle = \hat{e}_I\int\frac{d\omega}{2\pi}\left(\frac{\hbar(\omega_I^0+\omega)}{2\epsilon_I(\omega_I^0+\omega)V}\right)^{1/2}\beta(\omega')\times\frac{\kappa}{[\kappa^2+(\frac{1}{2}\Delta\omega)^2]^{1/2}}\sin([\kappa^2+(\tfrac{1}{2}\Delta\omega)^2]^{1/2}t)\times e^{i[(k_I^0+k)z-(\omega_I-\frac{1}{2}\Delta\omega)t]}. \quad (4.13)$$

Using the condition (4.6), an expression similar to Eq. (4.9) for $e^{i(k_I^0+k)z}$, and the relations (3.10) and (3.12) for ω' and $\Delta\omega$ in terms of ω, we find

$$\langle \mathbf{E}_I^+(z,t)\rangle = \hat{e}_I\left(\frac{\hbar\omega_I^0}{2\epsilon_I^0V}\right)^{1/2}\frac{\kappa\beta_0}{2\pi}e^{i(k_I^0z-\omega_I^0t)}\times\int_{-\infty}^{\infty}d\omega\, e^{i(\omega/v_I)[z-\frac{1}{2}(v_I+v_s)t]}\times\frac{\sin(\kappa^2+\chi^2\omega^2)^{1/2}t}{(\gamma^2+\eta^2\omega^2)(\kappa^2+\chi^2\omega^2)^{1/2}}, \quad (4.14)$$

where

$$\chi=\tfrac{1}{2}(1-v_s/v_I),\quad \eta=v_s/v_I. \quad (4.15)$$

The integration over ω in (4.14) may be performed using the tabulated form (B1) given in Appendix B. The final result is

$$\langle \mathbf{E}_I^+(z,t)\rangle = \hat{e}_I\left(\frac{\hbar\omega_I^0}{2\epsilon_I^0V}\right)^{1/2}\frac{\kappa\beta_0}{2\gamma\eta}e^{-(\gamma/v_s)|z-\frac{1}{2}(v_I+v_s)t|}\times\frac{\sin[\kappa^2-(\gamma\chi/\eta)^2]^{1/2}t}{[\kappa^2-(\gamma\chi/\eta)^2]^{1/2}}e^{i(k_I^0z-\omega_I^0t)}. \quad (4.16)$$

It is seen that the wave packet generated in the idler field after time t has the same form as the original packet present in the signal field (4.10), and that it moves with the average group velocity $\bar{v}=\frac{1}{2}(v_I+v_s)$ of the two modes. If we consider the frequency spread γ of the initial wave packet to be sufficiently small so that

$$\frac{\chi\gamma}{\eta}=\frac{1}{2}\left(\frac{v_I}{v_s}-1\right)\gamma\ll\kappa, \quad (4.17)$$

then there is essentially total conversion, and (4.14) reduces to

$$\langle \mathbf{E}_I^+(z,t)\rangle = \hat{e}_I\left(\frac{\hbar\omega_I^0}{2\epsilon_I^0V}\right)^{1/2}\frac{\beta_0}{2\gamma\eta}\sin\kappa t\, e^{-(\gamma/v_s)|z-\bar{v}t|}e^{i(k_I^0z-\omega_I^0t)}$$

$$= -i\hat{e}_I\left(\frac{\omega_I^0\epsilon_s^0}{\omega_s^0\epsilon_I^0}\right)^{1/2}\frac{v_I}{v_s}\langle E_s^+(0,0)\rangle\times\sin\kappa t\, e^{-(\gamma/v_s)|z-\bar{v}t|}e^{i(k_I^0z-\omega_I^0t)}. \quad (4.18)$$

The behavior of the generated wave packet is apparent from (4.18). After $t=0$, the packet begins to form at the idler frequency with an envelope of the same form as the initial signal field. As time progresses, the packet moves with the average group velocity, its center being at $z_0=\bar{v}t$, and grows in amplitude as $\sin\kappa t=\sin(\kappa z_0/\bar{v})$. It is in this manner that the expected spatial variation in the strength of the idler field is manifested quantum-mechanically. If we pictured the signal field as composed of a succession of such wave packets with coherent phase, we would expect the time average of the electric field in the idler mode to vary as $\sin(\kappa z/\bar{v})$, in qualitative agreement with the classical steady-state solution.

These conclusions are, of course, independent of the particular form of the wave-packet envelope. In fact, it is not difficult to show that if the original wave packet present in the signal field at $t=0$ is of the form

$$\langle \mathbf{E}_s^+(z,0)\rangle = i\hat{e}_s f(z) e^{ik_s^0 z}\,, \tag{4.19}$$

where the envelope $f(z)$ has a frequency width γ satisfying (4.15), then the idler field at time t is given by

$$\langle \mathbf{E}_I^+(z,t)\rangle = -i\hat{e}_I\left(\frac{\omega_I^0\epsilon_s^0}{\omega_s^0\epsilon_I^0}\right)^{1/2}\frac{v_I}{v_s}\sin\kappa t\, f(z-\bar{v}t) \times e^{i(k_I^0 z-\omega_I^0 t)}\,. \tag{4.20}$$

Thus, the above qualitative arguments are seen to apply for an arbitrary initial wave packet, provided that its frequency spread is sufficiently small. The above considerations may be generalized to the case where there is an initial wave packet present in both the signal and idler fields. The initial density matrix is taken to be

$$\rho(0) = |\{\beta(\omega')\}\{\alpha(\omega)\}\rangle\langle\{\alpha(\omega)\}\{\beta(\omega')\}|\,, \tag{4.21}$$

where the amplitudes of the coherent states are again assumed to have a Lorentz profile in frequency space.

$$\alpha(\omega) = \frac{\alpha_0}{\omega^2+\gamma'^2}\,,\quad \beta(\omega') = \frac{\beta_0}{\omega'^2+\gamma^2}\,. \tag{4.22}$$

With the conditions

$$\gamma' \ll \omega_I^0\,,\quad \gamma \ll \omega_s^0$$

the expectation value of the electric field in the idler may be written as

$$\langle\{\alpha\}\{\beta\}|\mathbf{E}_I^+(z,t)|\{\beta\}\{\alpha\}\rangle = i\hat{e}_I\left(\frac{\hbar\omega_I^0}{2\epsilon_I^0 V}\right)^{1/2}\int d\omega\, e^{i(\omega/v_I)(z-\bar{v}t)}\left[\frac{\alpha_0}{\gamma^2+\omega^2}\left(\cos(t(\kappa^2+\chi^2\omega^2)^{1/2}) - \frac{i\omega\sin(t(\kappa^2+\chi^2\omega^2)^{1/2})}{(\kappa^2+\chi^2\omega^2)^{1/2}}\right) - i\frac{\kappa\beta_0}{\gamma'^2+\eta^2\omega^2}\,\frac{\sin(t(\kappa^2+\chi^2\omega^2)^{1/2})}{(\kappa^2+\chi^2\omega^2)^{1/2}}\right]. \tag{4.23}$$

The integration over ω in (4.23) may be performed using the tabulated integrals (B1), (B2), and (B3) given in Appendix B. The final result is

$$\begin{aligned}\langle\{\beta\}\{\alpha\}|\mathbf{E}_I^+(z,t)|\{\alpha\}\{\beta\}\rangle &= \hat{e}_I \exp i(k_I^0 z-\omega_I^0 t)\left\{\exp\left(-\frac{\gamma'}{v_I}|z-\bar{v}t|\right)\langle E_I(0)\rangle\left[\cos([\kappa^2-(\gamma'\chi)^2]^{1/2}t) - i\frac{\chi\gamma'}{[\kappa^2-(\chi\gamma')^2]^{1/2}}\sin([\kappa^2-(\gamma'\chi)^2]^{1/2}t)\right]\right.\\ &\left.\quad - i\exp\left(-\frac{\gamma}{v_s}|z-\bar{v}t|\right)\langle E_s^+(0)\rangle\frac{v_I}{v_s}\left(\frac{\omega_I^0\epsilon_s^0}{\omega_s^0\epsilon_I^0}\right)^{1/2}\frac{\kappa}{[\kappa^2-(\gamma/\eta)^2]^{1/2}}\sin([\kappa^2-(\gamma/\eta)^2]^{1/2}t)\right\}. \end{aligned}\tag{4.24}$$

Thus, we see the oscillatory exchange of wave packets between signal and idler. We note that there is not perfect conversion at $t=\pi/2\kappa$, because of the energy mismatch present in the coupling of the modes which make up the wave packets. In the limit $\gamma\chi/\eta\ll\kappa$, $\gamma'\chi\ll\kappa$, Eq. (4.24) reduces to

$$\begin{aligned}\langle\{\beta\}\{\alpha\}|\mathbf{E}_I^+(z,t)|\{\alpha\}\{\beta\}\rangle &= \hat{e}_I\exp[i(k_I^0 z-\omega_I^0 t)]\left[\langle E_I^+(0)\rangle\cos\kappa t\exp\left(-\frac{\gamma'}{v_I}|z-\bar{v}t|\right) - i\frac{v_I}{v_s}\left(\frac{\omega_I^0\epsilon_s^0}{\omega_s^0\epsilon_I^0}\right)^{1/2}\langle E_s^+(0)\rangle\sin\kappa t\exp\left(-\frac{\gamma}{v_s}|z-\bar{v}t|\right)\right]\end{aligned}\tag{4.25}$$

and we have essentially perfect conversion at $t=\pi/2\kappa$.

V. CONCLUSION

We have derived a quantum-mechanical model for parametric frequency conversion which includes the effects of coupling between bands of modes centered about the ideally phase-matched signal and idler frequencies. The case of an infinite medium was taken to simplify the coupling and enable a solution to the equations of motion for the field operators by elementary means. We utilized this model to indicate the manner in which the expected spatial variation in amplitude of the generated idler field emerges from a quantum-mechanical analysis.

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APPENDIX A

We shall derive the time-dependent density operator for two coupled modes with an energy mismatch, which are initially in a coherent state.

The initial density operator is assumed to be

$$\rho(0)=|\alpha_0\beta_0\rangle\langle\alpha_0\beta_0|\,. \qquad \text{(A1)}$$

The time-dependent characteristic function is defined by[2,3]

$$\chi_N(\eta,\xi,t)=\langle\alpha_0\beta_0|e^{\eta a^\dagger(t)+\xi b^\dagger(t)}e^{-\eta^* a(t)-\xi^* b(t)}|\alpha_0\beta_0\rangle. \qquad \text{(A2)}$$

The solutions for the time-dependent operators for the energy-mismatched modes are given by Eq. (3.10). Substituting these solutions into Eq. (A2), we obtain

$$\chi_N(\eta,\xi,t)=e^{\eta\bar{\alpha}^*(t)-\eta^*\bar{\alpha}(t)+\xi\beta^*(t)-\xi^*\bar{\beta}(t)}\,, \qquad \text{(A3)}$$

where

$$\bar{\alpha}(t)=e^{-i(\omega_I{}^0+\omega)t}e^{i\chi t\omega}\left(\alpha_0\frac{\cos(\chi t[(\kappa/\chi)^2+\omega^2]^{1/2})-i\omega\sin(\chi t[(\kappa/\chi)^2+\omega^2]^{1/2})}{[(\kappa/\chi)^2+\omega^2]^{1/2}}-i\beta_0\frac{\kappa}{\chi}\frac{\sin(\chi t[(\kappa/\chi)^2+\omega^2]^{1/2})}{[(\kappa/\chi)^2+\omega^2]^{1/2}}\right),$$

$$\bar{\beta}(t)=e^{-i(\omega_s{}^0+\omega')t}e^{-i\chi t\omega}\left[-i\alpha_0\frac{\kappa}{\chi[(\kappa/\chi)^2+\omega^2]^{1/2}}\sin(\chi t[(\kappa/\chi)^2+\omega^2]^{1/2})\right.$$
$$\left.+\beta_0\left(\cos(\chi t[(\kappa/\chi)^2+\omega^2]^{1/2})+\frac{i\omega}{[(\kappa/\chi)^2+\omega^2]^{1/2}}\sin(\chi t[(\kappa/\chi)^2+\omega^2]^{1/2})\right)\right]. \qquad \text{(A4)}$$

We may obtain the time-dependent P representation from[2,3]

$$P(\alpha,\beta,t)=\frac{1}{\pi^4}\int e^{\eta^*\alpha+\xi^*\beta-\eta\alpha^*-\xi\beta^*}\chi_N(\eta,\xi,t)d^2\eta d^2\xi. \qquad \text{(A5)}$$

Upon substitution of (A3) this becomes

$$P(\alpha,\beta,t)=\frac{1}{\pi^4}\int e^{\eta^*[\alpha-\bar{\alpha}(t)]-\eta[\alpha-\alpha(t)]^*}\times e^{\xi^*[\beta-\bar{\beta}(t)]-\xi[\beta-\bar{\beta}(t)]^*}d^2\eta d^2\xi, \qquad \text{(A6)}$$

which yields on integration

$$P(\alpha,\beta,t)=\delta^2(\alpha-\bar{\alpha}(t))\delta^2(\beta-\bar{\beta}(t)). \qquad \text{(A7)}$$

We shall now calculate the general solution for the time-dependent density operator given that an initial P representation exists.

We assume that the fields in the two-mode system initially have the P representation

$$\rho(0)=\int|\alpha_0\beta_0\rangle\langle\alpha_0\beta_0|P(\alpha_0,\beta_0,0)d^2\alpha_0d^2\beta_0. \qquad \text{(A8)}$$

Since this just amounts to an ensemble of coherent states, we may use the previous result (A6) to obtain

$$P(\alpha,\beta,t)=\int P(\alpha_0,\beta_0,0)\delta^2(\alpha-\bar{\alpha}(t))\times\delta^2(\beta-\bar{\beta}(t))d^2\alpha_0d^2\beta_0. \qquad \text{(A9)}$$

In order to integrate out the δ functions, we require the inverse of the transformations (A4), which may be written as follows:

$$\alpha_0=\left(\cos(t[\kappa^2+(\chi\omega)^2]^{1/2})+\frac{i\omega\chi\sin(t[\kappa^2+(\chi\omega)^2]^{1/2}}{[\kappa^2+(\chi\omega)^2]^{1/2}}\right)\times\bar{\alpha}(t)e^{i(\omega_I{}^0+\omega)t}e^{-i\chi\omega t}+\frac{i\kappa\sin(t[\kappa^2+(\chi\omega)^2]^{1/2})}{[\kappa^2+(\chi\omega)^2]^{1/2}}\times\bar{\beta}(t)e^{i(\omega_s{}^0+\omega')t}e^{i\chi\omega t},$$

$$\beta_0=\frac{i\kappa\sin(t[\kappa^2+(\chi\omega)^2]^{1/2})}{[\kappa^2+(\chi\omega)^2]^{1/2}}\bar{\alpha}(t)e^{i(\omega_I{}^0+\omega)t}e^{-i\chi\omega t}+\left(\cos(t[\kappa^2+(\chi\omega)^2]^{1/2})+\frac{i\chi\omega\sin(t[\kappa^2+(\chi\omega)^2]^{1/2})}{[\kappa^2+(\chi\omega)^2]^{1/2}}\right)\times\beta(t)e^{i(\omega_I{}^0+\omega')t}e^{i\chi\omega t}. \qquad \text{(A10)}$$

Carrying out the integration over α_0 and β_0 in (A9) with the aid of (A10), we obtain

$$P(\alpha,\beta,t)=P(\mathcal{A}(\alpha,\beta,t)\mathcal{B}(\alpha,\beta,t),0)\,, \qquad \text{(A11)}$$

where $\mathcal{A}(\alpha,\beta,t)$, $\mathcal{B}(\alpha,\beta,t)$ are identical to α_0, β_0, respectively, given by Eq. (A10), with the substitutions $\bar{\alpha}(t)\to\alpha$, $\bar{\beta}(t)\to\beta$.

APPENDIX B

The following integrals used in the text are tabulated in Ref. 9:

$$\int_0^\infty \cos(px)\frac{\sin[b(x^2+a^2)^{1/2}]}{(x^2+c^2)(x^2+a^2)^{1/2}}dx = \pi e^{-|cp|}\frac{\sin[b(a^2-c^2)^{1/2}]}{2c(a^2-c^2)^{1/2}}, \quad \text{(B1)}$$

where $c\neq a$, $b\leqslant p$;

$$\int_0^\infty \cos(px)\cos[b(a^2+x^2)^{1/2}]\frac{c}{x^2+c^2}dx = \tfrac{1}{2}\pi e^{-|cp|}\cos[b(a^2-c^2)^{1/2}], \quad \text{(B2)}$$

where $b\leqslant p$;

$$\int_0^\infty \sin(px)x\frac{\sin[b(a^2+x^2)^{1/2}]}{(a^2+x^2)^{1/2}}\frac{c}{x^2+c^2}dx = \tfrac{1}{2}\pi\frac{c}{(a^2-c^2)^{1/2}}e^{-|cp|}\sin[b(a^2-c^2)^{1/2}], \quad \text{(B3)}$$

where $b<p<\infty$.

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Quantum Mechanics of Paraparticles*

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We show that it is possible to formulate a consistent first-quantized theory of paraparticles, i.e., particles which are neither bosons nor fermions. We examine a number of properties of the theory and show that the formulation of Messiah and Greenberg in terms of generalized rays can be replaced by an equivalent formulation in which states are represented by rays in the usual way. We use this alternative formulation to establish some results of ordinary quantum mechanics. We examine in detail the consistency of the theory with the cluster law and show that paraparticles must have states associated with whole families of different permutation symmetries, according to the following rule: If a given particle has N-particle states associated with a given Young diagram, then it must have $(N-1)$-, $(N-2)$-, $\cdots$, two-particle states associated with *all* Young diagrams which can be obtained from the first by successively removing squares. This gives rise to infinitely many different kinds of paraparticle, all with rather complicated properties.

I. INTRODUCTION

THERE is no evidence that particles other than bosons or fermions exist in nature. There is also no evidence that any but the most stable known particles actually are either bosons or fermions.[1] The name "paraparticle" has been introduced for a particle which is neither boson nor fermion, and the possible existence of such particles has been discussed by several authors.[1–5]

Theories of paraparticles have been constructed in two ways. The first approach considers the allowed permutation symmetries of the multiparticle wave functions and is formulated within the framework of first-quantized quantum mechanics. (See Refs. 1–3.) The second approach is a second-quantized theory in which commutation relations more general than the customary Bose or Fermi type are considered. (See Refs. 2–5.) In this paper we discuss only the former (first-quantized) approach; we do not discuss the as-yet unexplained connection between the two types of theory.

Our aim is to examine the consistency of the first-quantized theory of paraparticles. In particular, we reexamine the argument of Steinmann[2] that parastatistics are incompatible with the important cluster law, which requires that two well-separated groups of particles can be treated as separate isolated systems.

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