Photon Correlations in the Parametric Frequency Splitting of Light

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Correlation effects in parametric photon-pair production are studied within the framework of a physically realistic model. The analysis, which is fully quantum mechanical, takes into account the finite sizes of the target and the beam cross section, and allows for dispersion and anisotropy in the linear susceptibility. The correlations in position and time at which the two members of a parametrically generated pair may be detected are carefully evaluated. These correlations, which have been measured experimentally, are intrinsically quantum mechanical; i.e., they can be explained by no theory in which the subharmonic fields are described purely by c-number functions. A complete solution, from which field correlation functions of arbitrarily high order may be evaluated, is obtained by a method which at the same time allows for an arbitrary degree of parametric gain. The solution is expressed entirely in terms of a particular two-point field-correlation-function, as evaluated in lowest order in the incident field strength, at points distant from the target. The function in question is found by directly examining the fluctuating currents in the material medium, rather than by eliminating the matter variables at the outset through the introduction of a nonlinear electomagnetic susceptibility.

I. INTRODUCTION

Spontaneous subharmonic generation or "frequency splitting" of light, whereby a photon of frequency ω_0 propagating within a nonlinear medium divides its energy between two photons of smaller energy ω_1 and ω_2, is a particularly simple physically realizable process which demonstrates the inherently quantum-mechanical nature of the electromagnetic field. The correlations in the directions and in the times at which the two photons are emitted—and therefore detected—can be explained by no theory in which the radiated field is described by a purely classical function. The coincidence counting rate recorded by a pair of optimally placed detectors, for sufficiently weak incident fields, is proportional in the quantum theory to the incident field power P_0, and hence can be made arbitrarily large compared to the accidental or uncorrelated coincidence rate, which is proportional to P_0^2. Ample confirmation of the predictions of the quantum theory of spontaneous frequency splitting has been achieved by Burnham and Weinberg[1] in an experiment which, finding more than a hundred-to-one ratio between actual and accidental coincidence counting rates, leaves little room for any conceivable semiclassical explanation of the process.[2]

Stimulated frequency splitting, on the other hand, in which a field is initially present at one or both of the subharmonic frequencies, may be described by a straightforward (though not fully correct) semiclassical theory based on the elimination of the atomic variables through the introduction of a nonlinear electromagnetic susceptibility.[3] This is done by taking the expectation value of the atomic current operator, evaluated in the presence of two of the (c-number) fields, to be the source for the third. A solvable quantum theory including spontaneous as well as stimulated processes can be obtained from the semiclassical theory simply by replacing the c-number subharmonic field amplitudes by Heisenberg operators.[4] Spontaneous frequency splitting then emerges as amplification of the "vacuum fluctuations" associated with the noncommutation of the field operators. The simplest model of the parametric amplifier, in which the field at each subharmonic frequency is represented by a single discrete mode of oscillation, has been extensively discussed in quantum-mechanical terms,[5] for arbitrary degrees of amplification.

More realistic models, in which infinitely many modes are excited, are far more difficult to treat, and analyses of them seldom allow for the possibility of amplification[6] or are concerned with correlation effects. The coupled-mode approach outlined above is particularly difficult to apply beyond the simplest approximation in cases where the nonlinear medium occupies a limited region of space, as it does in real frequency-splitting experiments. Previous theoretical analyses[7] of the frequency-splitting process are not adequate, in particular, to describe correlations such as those measured in the Burnham-Weinberg experiment, nor do they allow for the possibility of parametric gain.

In this paper a rather complete theoretical analysis of the frequency-splitting process is presented, which, while physically realistic, provides a solution for field-correlation effects of arbitrary order and allows for an arbitrary degree of para-

metric gain. The analysis is carried out by first obtaining solutions in the limit of low incident field intensity, where the process is entirely spontaneous, and then using the solutions so obtained to construct the solution which applies when parametric gain may be appreciable. The solutions in all cases are expressed in terms of the values of the quantum-mechanical field-correlation functions[8,9] at points far from the target, and may be used to find the complete statistical distribution of photocounts.

Field-correlation functions of arbitrary order, it is found, can be expressed in terms of the single function $G^{(0,2)}(x_2, x_1) \equiv \langle A^{(+)}(x_2)A^{(+)}(x_1)\rangle$, which represents the product of two photon-annihilation operators. In order to facilitate comparison with semiclassical theories, this function is evaluated not by using a formalism in which the atomic variables have been eliminated but instead by a direct examination of the fluctuating electric current within the material medium, as induced by the incident or "pump" field. (The latter is represented by a c-number function.) A quite general relation is derived which expresses the function $G^{(0,2)}$ at points distant from an arbitrary current distribution in terms of a suitable two-point electric current-correlation function. (The derivation of this relation requires careful attention to operator ordering.) In the case of spontaneous frequency splitting, the mean values of the atomic current operators vanish, while the fluctuations described by the aforementioned current-correlation function do not. The intrinsically quantum-mechanical nature of the spontaneous frequency-splitting process is thus exhibited in a particularly fundamental way.

In the weak pump-field limit, expressions are obtained for the function $G^{(0,2)}$, and hence for the coincidence counting rate recorded by a pair of detectors, for the case of a target consisting of a plane slab which is illuminated by an incident beam of arbitrary cross-sectional profile. The effects of linear dispersion and anisotropy are fully taken into account. Suitable limiting cases are discussed, and the correlation time and angular width are evaluated within which one photon of a given parametrically generated pair may be detected, given the location at which the other is detected. The cross-spectral function $G^{(1,1)}(x_1', x_1) \equiv \langle A^{(-)}(x_1')A^{(+)}(x_1)\rangle$ which determines the intensity and frequency distribution of one subharmonic field is obtained directly, for weak pump fields, from the function $G^{(0,2)}$.

Field-correlation functions of higher order than $G^{(0,2)}$ and $G^{(1,1)}$ are most easily found by a method which at the same time allows for an arbitrary degree of parametric gain, and thus yields corrected expressions for the functions $G^{(0,2)}$ and $G^{(1,1)}$ themselves, valid to arbitrary order in the pump-field strength. The method in question consists of finding the unitary operator which generates the quantum state of the radiated field from the vacuum state, thus representing the coherent superposition of the two-photon wave functions emitted by the individual atoms. (In one-photon scattering, the analogous method leads to the introduction of the unitary *displacement operator*[10] and to the consequent representation of the scattered field by a *coherent state*.) The *characteristic functional* for the scattered field, from which field-correlation functions of arbitrary order can be directly evaluated, is found by this method to be expressible in terms of the (corrected) functions $G^{(0,2)}$ and $G^{(1,1)}$ alone. These in turn are found by the same method to be expressible as a power series consisting of multiple convolution integrals of the function $G^{(0,2)}$ as evaluated to lowest order in the pump-field strength, at points distant from the target. The series is governed by a parameter roughly equal to the number of photons located, according to the lowest-order theory, within a coherence volume in the scattering region. The solution in the case of strong pump fields implies considerable amplification of the spectral components of the field near the center of each subharmonic emission line, and thus a reduction in its width.

In Sec. II the current-correlation function which determines the function $G^{(0,2)}$ is derived in a quite general context. The value of this function for a single atom driven by a prescribed field is then found, to lowest order in the field strength, in Sec. III. In Sec. IV the scattering from a target consisting of many atoms is discussed, and the spatial and temporal correlations in the lowest-order coincidence rates are found. Sec. V is devoted to an analysis of the effect of parametric gain, and contains the solutions for field-correlation functions of arbitrary order.

II. CURRENT-CORRELATION FUNCTION FOR PHOTON PAIR EMISSION

The electromagnetic-field-correlation function which describes the photon-detection coincidence rate at two separated space-time points is directly obtainable from the four-point function[8,9]

$$G^{(2,2)}(x_1', x_2'; x_2, x_1) \equiv \langle A^{(-)}(x_1')A^{(-)}(x_2')A^{(+)}(x_2)A^{(+)}(x_1)\rangle, \quad (2.1)$$

where $x \equiv (\vec{r}, t)$ and the superscripts on the (Heisenberg) vector-potential field operators are frequency signatures. In the cases to be considered, it will prove possible to express the function defined by Eq. (2.1) in terms of the simpler two-point

functions

$$G^{(0,2)}(x_2, x_1) \equiv \langle A^{(+)}(x_2) A^{(+)}(x_1) \rangle \tag{2.2}$$

and

$$G^{(2,0)}(x_1, x_2) \equiv \langle A^{(-)}(x_1) A^{(-)}(x_2) \rangle . \tag{2.3}$$

In particular, when the state of the field is the superposition of the vacuum state and a state $|2\rangle$ containing two photons with small probability,

$$|t\rangle \approx |0\rangle + |2\rangle, \tag{2.4}$$

it is not difficult to deduce the approximate factorization relation

$$G^{(2,2)}(x_1', x_2'; x_2, x_1) = G^{(2,0)}(x_1', x_2') G^{(0,2)}(x_2, x_1). \tag{2.5}$$

The function $G^{(0,2)}(x_2, x_1)$, which for equal time arguments may then be roughly described as the wave function for the photon pair, has squared value proportional to the simultaneous two-photon-detection probability.

The field-correlation functions will all be evaluated subject to the initial condition of vanishing field strength at an initial time in the distant past, at which the Heisenberg and Schrödinger pictures of the motion will be taken to coincide. The state vector $|\rangle$ for the system at this time then satisfies the relations

$$A_I^{(+)}(x)|\rangle = 0 = \langle |A_I^{(-)}(x), \tag{2.6}$$

where $A_I(x) = A_I^{(+)}(x) + A_I^{(-)}(x)$ is the freely propagatting or homogeneous part of the solution to the wave equation. It is convenient to introduce the function

$$D(x) \equiv i[A_I(x), A_I(x')]/\hbar c^2 = D_c(x) - D_A(x), \tag{2.7}$$

where $D_c(x)$ and $D_A(x)$ are the causal and advanced Green's functions

$$D_c(x) = \left(1 - \frac{\vec{\nabla}\vec{\nabla}}{\nabla^2}\right) \cdot \frac{\delta(r - ct)}{4\pi c r} = \Theta(t) D(x), \tag{2.8a}$$

$$D_A(x) = \left(1 - \frac{\vec{\nabla}\vec{\nabla}}{\nabla^2}\right) \cdot \frac{\delta(r + ct)}{4\pi c r} = -\Theta(-t) D(x). \tag{2.8b}$$

Here $\Theta(t)$ is the unit step function,

$$\begin{aligned} \Theta(t) &= 1 \quad \text{for } t \geq 0 \\ &= 0 \quad \text{for } t < 0. \end{aligned} \tag{2.9}$$

In accordance with the boundary condition of vanishing field for $t \to -\infty$, the solution to the wave equation for the Heisenberg field operator $A(x)$ has the form

$$A(x) = A_I(x) + \int d^4\bar{x}\, D_c(x - \bar{x}) J(\bar{x}), \tag{2.10}$$

where $J(x)$ is the Heisenberg electric current operator, $d^4x \equiv c\, d^3r\, dt$, and the integrations on $\vec{r}$ and t extend from $-\infty$ to ∞.

Equations (2.6) enable one immediately to write down the solution for the cross-spectral field-correlation function

$$G^{(1,1)}(x', x) \equiv \langle A^{(-)}(x') A^{(+)}(x) \rangle \tag{2.11}$$

as

$$G^{(1,1)}(x', x) = \int\int d^4\bar{x}'\, d^4\bar{x}\, D_c(x' - \bar{x}') D_c(x - \bar{x}) \times \langle J^{(-)}(\bar{x}') J^{(+)}(\bar{x}) \rangle, \tag{2.12}$$

since the contributions from the freely propagating operators $A_I(x)$ in Eq. (2.10) vanish identically.

The function defined by Eq. (2.2), on the other hand, takes a more complicated form when $A(x)$ is obtained from Eq. (2.10), for while the contribution from $A_I^{(+)}(x_1)$ on the right-hand side vanishes by virtue of Eqs. (2.6), the contribution from $A_I^{(+)}(x_2)$ on the left-hand side does not. The function $G^{(0,2)}(x_2, x_1)$ is therefore given as

$$\begin{aligned} G^{(0,2)}(x_2, x_1) = &\int\int d^4\bar{x}_2 d^4\bar{x}_1 D_c^{(+)}(x_2 - \bar{x}_2) \\ &\times D_c^{(+)}(x_1 - \bar{x}_1) \langle J(\bar{x}_2) J(\bar{x}_1) \rangle \\ &+ \int d^4\bar{x}_1 D_c^{(+)}(x_1 - \bar{x}_1) \langle [A_I^{(+)}(x_2), J(\bar{x}_1)] \rangle , \end{aligned} \tag{2.13}$$

where $D_c^{(+)}(x)$ is the positive-frequency part of $D(x)$. In the second term use has been made of the relation

$$\langle A_I^{(+)}(x_2) J(\bar{x}_1) \rangle = \langle [A_I^{(+)}(x_2), J(\bar{x}_1)] \rangle, \tag{2.14}$$

which follows from Eqs. (2.6).

The commutator of the freely propagating field operator $A_I(x_2)$ and the full Heisenberg current operator $J(\bar{x}_1)$ may be expressed in terms of the current operators alone in a straightforward manner. One may begin by noting that the full Heisenberg operators $A(x_2)$ and $J(\bar{x}_1)$ must commute at equal times,

$$[A(x_2), J(\bar{x}_1)] = 0 \quad \text{for } t_2 = \bar{t}_1 . \tag{2.15}$$

By making use of Eq. (2.10) for $A(x_2)$ in this relation, one finds the relation

$$[A_I(x_2), J(\bar{x}_1)] = -\int d^4\bar{x}_2 D_c(x_2 - \bar{x}_2) \langle [J(\bar{x}_2), J(\bar{x}_1)] \rangle \quad (t_2 = \bar{t}_1). \tag{2.16}$$

It then follows from Eq. (2.8a) that at $t_2 = \bar{t}_1$ one may write

$$[A_I(x_2), J(\bar{x}_1)] = -\int d^4\bar{x}_2 \theta(\bar{t}_1 - \bar{t}_2) D(x_2 - \bar{x}_2) \times \langle [J(\bar{x}_2), J(\bar{x}_1)] \rangle . \tag{2.17}$$

It is easily shown that the latter equation (unlike Eq. (2.16)) is valid not only at $t_2 = \bar{t}_1$, but for all times t_2. The proof of this assertion follows immediately from the fact that both sides obey the wave equation, i.e., vanish under the operation

$$\Box_2^2 = \nabla_2^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t_2^2}, \tag{2.18}$$

and both have the same value at $t_2 = \bar{t}_1$. [That both sides have the same time derivative with respect to t_2 at $t_2 = \bar{t}_1$ follows directly from an argument similar to the one leading to Eq. (2.17).]

By taking the positive-frequency part (with respect to t_2) of Eq. (2.17) and substituting the result into Eq. (2.13), one finds, with the aid of Eq. (2.7), the relation

$$G^{(0,2)}(x_2, x_1) = \int\int d^4\bar{x}_2 d^4\bar{x}_1 \{D_c^{(+)}(x_2 - \bar{x}_2) D_c^{(+)}(x_1 - \bar{x}_1) \times \langle (J(\bar{x}_2) J(\bar{x}_1))_T \rangle + \theta(\bar{t}_1 - \bar{t}_2) D_A^{(+)}(x_2 - \bar{x}_2) \times D_c^{(+)}(x_1 - \bar{x}_1) \langle [J(\bar{x}_2), J(\bar{x}_1)] \rangle\}, \tag{2.19}$$

where the subscript T denotes time ordering (later times are to the left).

It is not difficult to show that in the limit of infinite distance from the current distribution, the term containing the advanced Green's function in Eq. (2.19) (which represents waves traveling toward the target) must make a vanishing contribution. By transferring the frequency signatures in the remaining term to the current-correlation function, one finds the relation

$$G^{(0,2)}(x_2, x_1) = \int\int d^4\bar{x}_2 d^4\bar{x}_1 D_c(x_2 - \bar{x}_2) D_c(x_1 - \bar{x}_1) \times \langle (J(\bar{x}_2) J(\bar{x}_1))_T \rangle^{(++)}, \tag{2.20}$$

in which the positive-frequency parts are to be taken after time ordering.

It may be noted that the function $G^{(0,2)}(x_2, x_1)$ as given by Eq. (2.20) is symmetric under the interchange $x_2 \longleftrightarrow x_1$, as one should expect it to be at points far from the current distribution.

III. SPONTANEOUS PAIR-EMISSION COEFFICIENT AND NONLINEAR SUSCEPTIBILITY

For a single atom at the point $\vec{r}$, the electric current-correlation function in Eq. (2.20) may be expressed, in the electric dipole approximation, as

$$\langle (J(x_2) J(x_1))_T \rangle^{(++)} = -\delta^{(3)}(\vec{r}_2 - \vec{r}) \delta^{(3)}(\vec{r}_1 - \vec{r}) \times (2\pi)^{-1} \int\int d\omega_2 d\omega_1 e^{-i\omega_2 t_2 - i\omega_1 t_1} \times \omega_2 \omega_1 \tilde{g}_T^{(0,2)}(\omega_2, \omega_1), \tag{3.1}$$

where $\tilde{g}_T^{(0,2)}(\omega_2, \omega_1)$ is defined in terms of the atomic dipole-moment correlation function $\langle \mu(t_2) \mu(t_1) \rangle$ as

$$\tilde{g}_T^{(0,2)}(\omega_2, \omega_1) = \theta(\omega_2)\theta(\omega_1)(2\pi)^{-1} \times \int_{-\infty}^{\infty} dt_2 \int_{-\infty}^{t_2} dt_1 e^{i\omega_2 t_2 + i\omega_1 t_1} \times \langle \mu(t_2) \mu(t_1) \rangle + (2 \longleftrightarrow 1). \tag{3.2}$$

[The interchange $2 \longleftrightarrow 1$ includes the (suppressed) vector indices as well as the frequency arguments ω_2 and ω_1.]

The atomic correlation function in Eq. (3.2) is readily evaluated when the atom is driven by a prescribed electric field $E(t)$. By using the dipole coupling $H_1 = -\mu E(t)$, it is not difficult to show that the Heisenberg operator $\mu(t)$ may be expressed in increasing powers of the field strength by means of the series expansion

$$\mu(t) = \mu_I(t) + (i/\hbar) \int_{-\infty}^{t} dt' [\mu_I(t), \mu_I(t')] E(t') + (i/\hbar)^2 \int_{-\infty}^{t} dt' \int_{-\infty}^{t'} dt'' [[\mu_I(t), \mu_I(t')], \mu_I(t'')] \times E(t') E(t'') + \cdots, \tag{3.3}$$

where $\mu_I(t)$ is the interaction picture (freely oscillating) operator

$$\mu_I(t) = \sum_{j,k} \mu_{jk} e^{i\omega_{jk} t} |j\rangle\langle k| \tag{3.4a}$$

$$\omega_{jk} = (E_j - E_k)/\hbar. \tag{3.4b}$$

For the case in which $E(t)$ is the harmonic function

$$E(t) = E_0 e^{-i\omega_0 t} + \text{c.c.}, \tag{3.5}$$

one finds by retaining terms only up to the first power in E_0 in Eq. (3.3) that the function defined by Eq. (3.2) is in this approximation nonvanishing only for $\omega_1 + \omega_2 = \omega_0$, and may be expressed by means of the relation

$$\tilde{g}_T^{(0,2)}(\omega_2, \omega_1) = -i\hbar\varphi(-\omega_2, -\omega_1, \omega_0) E_0, \tag{3.6}$$

in which the third-rank-tensor function φ is defined as

$$\varphi^{\lambda_2\lambda_1\lambda_0}(\omega_2, \omega_1, \omega_0) = -\frac{1}{\hbar^2} \delta(\omega_2 + \omega_1 + \omega_0) \times \sum_{p} \sum_{j_2, j_1} \frac{\mu_{g j_2}^{\lambda_{p(2)}} \mu_{j_2 j_1}^{\lambda_{p(1)}} \mu_{j_1 g}^{\lambda_{p(0)}}}{(\omega_{p(2)} + \omega_{j_2 g})(\omega_{p(0)} - \omega_{j_1 g})}. \tag{3.7}$$

In the latter relation, the superscripts are vector indices, g represents the ground state of the atom, and p is a permutation on the indices 0, 1, 2. All six such permutations are included in the sum in Eq. (3.7), which thus defines a function which is fully symmetric in all three of its arguments, i.e., which is invariant under the interchange $(\omega_j, \lambda_j) \longleftrightarrow (\omega_k, \lambda_k)$ for any pair of indices j and k.

For real values of the frequency arguments ω_1, ω_2, and ω_0, the function φ can be shown to obey the identity

$$\varphi^*(\omega_2, \omega_1, \omega_0) = \varphi(-\omega_2, -\omega_1, -\omega_0). \tag{3.8}$$

In the analysis which follows, all field frequencies

will be assumed far enough from the atomic resonance frequencies to justify omitting the small imaginary terms, proportional to the damping constants, which would otherwise appear in Eq. (3.7). It should be noted that in this approximation the function φ, which in Eq. (3.6) specifies the amplitude for a frequency-splitting process (in which a photon of frequency ω_0 divides its energy between two photons of smaller energies ω_1 and ω_2) specifies the amplitude for the inverse process as well. If an atom is stimulated by the field

$$E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t}, \tag{3.9}$$

then, as one finds by directly substituting this expression into Eq. (3.3) and retaining terms quadratic in E, the mean atomic dipole moment has a component oscillating at the sum frequency $\omega_1 + \omega_2$, which is given by the relation[3]

$$\langle \mu_0(t) \rangle = \int d\omega_0 e^{-i\omega_0 t} \varphi(-\omega_0, \omega_1, \omega_2) E_1 E_2, \tag{3.10}$$

where φ is the function defined by Eq. (3.7). [The functions φ in Eqs. (3.6) and (3.10) are complex conjugates of one another, as is apparent from Eq. (3.8) and the symmetry of the function defined by Eq. (3.7).]

Similarly, the simultaneous presence of the two fields E_0 and E_1 induces a component oscillating at the difference frequency $\omega_0 - \omega_1$, which is given by the relation[3]

$$\langle \mu_2(t) \rangle = \int d\omega_2 e^{-i\omega_2 t} \varphi(-\omega_2, -\omega_1, \omega_0) E_1^* E_0. \tag{3.11}$$

The process represented here is stimulated subharmonic generation, the same process whose spontaneous part is represented in Eq. (3.6). The value of the spontaneous coefficient as $\hbar$ times the stimulated coefficient, it should be emphasized, has been derived from a direct calculation of each, and not from a quantum Hamiltonian formalism based on classical analogy.

IV. SPATIAL AND TEMPORAL CORRELATIONS IN SPONTANEOUS FREQUENCY SPLITTING

When a large number of atoms are stimulated by the same light wave, it becomes necessary, in evaluating the function $G^{(0,2)}(x_2, x_1)$ defined by Eq. (2.2), not merely to add coherently the contributions from each atom, but to replace the propagation function $D_c(x)$ in Eq. (2.20) by the one which represents the effect of the linear dielectric susceptibility on the emitted waves. Let us assume that the incident wave propagates in the z direction and that its amplitude is specified in the transverse plane by the function $f(x, y)$:

$$\vec{\mathrm{E}}_0(\vec{\mathrm{r}}, t) = \vec{\mathrm{E}}_0 f(x, y) e^{ik_0 z - i\omega_0 t}. \tag{4.1}$$

If the number density N of atoms is constant within the target, then at observation points far away the function $G^{(0,2)}(x_2, x_1)$ may be approximated as

$$G^{(0,2)}(x_2, x_1) = \frac{N}{(4\pi c)^2} P_2 P_1 \frac{1}{r_1 r_2} \int\int e^{-i\omega_2 \bar{t}_2 - i\omega_1 \bar{t}_1} T_2 T_1 i\hbar \omega_2 \omega_1 \varphi(-\omega_2, -\omega_1, \omega_0) \cdot E_0 \zeta(\vec{k}_2 + \vec{k}_1 - \vec{k}_0) d\omega_2 d\omega_1 / 2\pi, \tag{4.2}$$

where $P = 1 - \hat{r}\hat{r}/r^2$ for $\vec{r} = \vec{r}_{1,2}$; $\bar{t}_{1,2} \equiv t_{1,2} - r_{1,2}/c$; φ is the nonlinear coefficient defined by Eq. (3.7); $\vec{k}_1$ and $\vec{k}_2$ are the wave vectors within the target associated (through Snell's law) with the frequencies ω_1 and ω_2 and the external observation directions $\hat{r}_1 \equiv \vec{r}_1/r_1$ and $\hat{r}_2 \equiv \vec{r}_2/r_2$, respectively; T_1 and T_2 are quantities proportional to the corresponding amplitude transmission coefficients[11]; and the function $\zeta(\vec{k})$ is defined by the relation

$$\zeta(\vec{k}) \equiv \int d^3 r e^{-i\vec{k}\cdot\vec{r}} f(x, y) \eta(\vec{r}). \tag{4.3}$$

The function $\eta(\vec{r})$ is here defined as unity inside the target and as zero outside. For the case of a plane slab perpendicular to the beam and of thickness l and cross section larger than that of the incident field, one may put

$$\eta(\vec{r}) \to \eta(z) \equiv \Theta(\tfrac{1}{2} l - |z|). \tag{4.4}$$

It is apparent from Eqs. (4.2) and (4.3) and the frequency δ function in the definition of φ that a resonance in the emission occurs when momentum and energy are both conserved, i.e., when $\vec{k}_2 + \vec{k}_1 = \vec{k}_0$ and $\omega_2 + \omega_1 = \omega_0$. Wave vectors $\vec{k}_1$ for which these relations are both satisfied (for given $\vec{k}_0$) are said to lie on the "phase-matching surface"[7] for pair production. It is assumed in what follows that the external observation direction $\hat{r}_1$ (which is thought of as fixed) is such that for a particular frequency ω_1^*, the associated internal wave vector $\vec{k}_1^*$ lies on the matching surface. The complementary frequency ω_2^* and wave vector $\vec{k}_2^*$, defined by the relations

$$\omega_2^* + \omega_1^* \equiv \omega_0, \tag{4.5a}$$

$$\vec{k}_2^* + \vec{k}_1^* \equiv \vec{k}_0, \tag{4.5b}$$

are then, like ω_1^* and $\vec{k}_1^*$, functions of $\hat{r}_1$.

In the limit of infinite target thickness and incident-beam cross section, the emission is wholly phase matched. This means that the emission in the direction $\hat{r}_1$ occurs exactly at the frequency

ω_1^*, and that the complementary photon of frequency ω_2^* is emitted in a precisely defined direction $\hat{r}_2^*$ (associated externally, through Snell's law, with $\vec{k}_2^*$). The correlation time for the pair-emission process is correspondingly infinite in the limit under discussion, i.e., the detection of one photon at a given time provides no information about the time at which the other photon is likely to be detected.

For finite values of the target thickness and beam radius, on the other hand, what may be thought of as a diffractive effect leads to a nonzero spectral width in the emission in the specified direction $\hat{r}_1$, and to an uncertainty in the direction $\hat{r}_2$ in which the complementary photon is emitted. At the same time, the correlation time for the process becomes finite. (It is in fact comparable to the transit time of a photon through the scattering region.)

It is convenient to introduce the parameters

$$\vec{A} \equiv \frac{d\vec{k}_1}{d\omega_1} - \frac{d\vec{k}_2}{d\omega_2} \tag{4.6}$$

and

$$\vec{B} \equiv k_2 \left(1 - \frac{\hat{k}_2 \vec{v}_2}{v_2'}\right) \cdot \delta\hat{k}_2, \tag{4.7}$$

in which $\hat{k}_2 \equiv \vec{k}_2/k_2$; $\delta\hat{k}_2$ is the shift in internal direction associated (for $\omega_2 = \omega_2^*$) with the difference $\delta\hat{r}_2 = \hat{r}_2 - \hat{r}_2^*$ between actual and optimal external directions; $\vec{v}$ is the group velocity $\vec{\nabla}_k \omega$; and

$$v' \equiv \vec{v} \cdot \hat{k}\,. \tag{4.8}$$

Each of the derivatives $d\vec{k}/d\omega$ in Eq. (4.6) is evaluated with external direction held fixed. In the absence of linear dispersion (where the internal direction also remains fixed), $d\vec{k}/d\omega$ is the "phase slowness"[12] $\vec{k}/\omega = \hat{k}/v'$, and the parameter $\vec{A}$ has the value

$$\vec{A} = \hat{k}_1/v_1' - \hat{k}_2/v_2'\,. \tag{4.9}$$

The parameter $\vec{B}$, for the case of isotropy in the linear susceptibility, is simply

$$\vec{B} = k_2 \delta\hat{k}_2\,. \tag{4.10}$$

For sufficiently large target thickness and beam cross section, it is not difficult to show that the function $G^{(0,2)}$ as given by Eqs. (4.2) and (4.3) is well approximated by the expression

$$G^{(0,2)}(x_2, x_1) = \frac{i\hbar\omega_1^*\omega_2^*\chi(\omega_2^*, \omega_1^*)E_0}{(4\pi c)^2 r_1 r_2} \times e^{-i\omega_2^* \bar{t}_2 - i\omega_1^* \bar{t}_1} \psi(\bar{t}_2 - \bar{t}_1;\ \hat{r}_2, \hat{r}_1), \tag{4.11}$$

in which the parameter χ is defined by the relation

$$NP_2P_1T_2T_1\varphi(-\omega_2, -\omega_1, \omega_0)\cdot\vec{E}_0/E_0 \equiv \delta(\omega_2 + \omega_1 - \omega_0)\chi(\omega_2, \omega_1), \tag{4.12}$$

and the function ψ is defined as

$$\psi(\tau; \hat{r}_2, \hat{r}_1) = \int d^3r f(x, y)\eta(z) e^{-i\vec{B}\cdot\vec{r}} \delta(\tau - \vec{A}\cdot\vec{r}). \tag{4.13}$$

These relations follow readily from the approximations $\vec{k}_1 \approx \vec{k}_1^* + (\omega_1 - \omega_1^*)\, d\vec{k}_1/d\omega_1$ and $\vec{k}_2 \approx \vec{k}_2^* + \vec{B} + (\omega_2 - \omega^*{}_2)\, d\vec{k}_2/d\omega_2$ in Eqs. (4.2) and (4.3).

The coincidence counting rate at two points $\vec{r}_1$ and $\vec{r}_2$, with effective time delay

$$\tau = t_2 - t_1 - (r_2 - r_1)/c,$$

according to Eqs. (2.5) and (4.11), is directly proportional to the function $|\psi(\tau; \hat{r}_2, \hat{r}_1)|^2$.

The function ψ is readily evaluated in the limiting cases of interest. If the target thickness l is much greater than the beam radius R, for example, one finds, taking the x-z plane to include the vector $\hat{r}_1$ (and hence $\hat{k}_1^*$, $\hat{k}_2^*$, $\hat{r}_2^*$, and $\vec{A}$), that ψ is well approximated by the relation

$$\psi(\tau; \hat{r}_2, \hat{r}_1) = (2\pi/A_z) e^{-i\tau B_z/A_z} \times \tilde{f}(B_x - B_z A_x/A_z,\ B_y)\eta(\tau/A_z) \quad (l \gg R) \tag{4.14}$$

in which $\tilde{f}$ is defined by the wave-vector decomposition

$$f(x, y) = (2\pi)^{-1} \int\int e^{ik_x x + ik_y y} \tilde{f}(k_x, k_y) dk_x\, dk_y\,. \tag{4.15}$$

The coincidence counting rate in this limit therefore vanishes, according to Eqs. (4.14), (4.4) and (4.9), unless the time delay τ is small enough to satisfy the inequality

$$|\tau| < \tfrac{1}{2} l |(\cos\theta_{1i})/v_1' - (\cos\theta_{2i})/v_2'|\,, \tag{4.16}$$

where θ_{1i} and θ_{2i} are internal polar angles.

It should be emphasized that what is evaluated here is the coincidence detection probability for the two photons which are generated during the same elementary parametric process. The approximation that at most two photons are present in the scattered field is implicit in the analysis of of this entire section, which is based on the weak-pump-field approximation in Eqs. (3.6) and (2.5). A more complete theory, adequate to describe pump fields of arbitrary intensity, will be presented in Sec. V.

To determine the dependence of ψ on the shift in the external observation direction $\hat{r}_2$ relative to its optimal value, one must use Snell's law to express $\delta\hat{r}_2$ in terms of $\delta\hat{k}_2$, and then make use of Eq. (4.7) in Eq. (4.14). In the case of isotropy in the linear susceptibility [where Eq. (4.10) holds], one finds that for a beam with Gaussian

cross section

$$f(x,y)=\exp[-\tfrac{1}{2}(x^2+y^2)/R^2], \tag{4.17}$$

the function given by Eq. (4.14) has the value

$$\psi(\tau;\hat{r}_2,\hat{r}_1)=(2\pi/A_z)\eta(\tau/A_z)e^{i\tau\delta\theta_{2i}\mu}\times\exp\{-\tfrac{1}{2}R^2k_2^2[\gamma^2(\delta\theta_{2i})^2+\sin^2\theta_{2i}(\delta\varphi_2)^2]\}\quad(l\gg R), \tag{4.18}$$

where $\delta\theta_{2i}$ and $\delta\varphi_2$ are the shifts in the internal polar and azimuthal angles, respectively, $\mu\equiv k_2(\sin\theta_{2i})/A_z$, and $\gamma\equiv\vec{A}\cdot\hat{k}_2/A_z$. The angular widths $\Delta\theta_2$ and $\Delta\varphi_2$ are thus both inversely proportional to the beam radius R in this limit, while the correlation time (or effective correlation length in the direction of $\hat{r}_2$) is proportional to the thickness l of the target.

In the limit in which the beam radius is large compared to the thickness of the target, on the other hand, one finds that the function ψ as given by Eqs. (4.13), (4.10), and (4.17) is well approximated by the relation

$$\psi(\tau;\hat{r}_2,\hat{r}_1)=(2\pi)^{1/2}RlA_x^{-1}e^{i\tau\delta\theta_{2i}\mu'}\times\exp[-\tfrac{1}{2}\tau^2/R^2A_x^2-\tfrac{1}{2}R^2k_2^2\sin^2\theta_{2i}(\delta\varphi_2)^2]\times\frac{\sin(\frac{1}{2}\gamma' k_2 l\delta\theta_{2i})}{\frac{1}{2}\gamma' k_2 l\delta\theta_{2i}}\quad(R\gg l), \tag{4.19}$$

where $\mu'\equiv k_2(\cos\theta_{2i})/A_x$ and $\gamma'\equiv\vec{A}\cdot\hat{k}_2/A_x$. The azimuthal angular width in this limit is still proportional to R^{-1}, while the polar angular width and correlation time are proportional to l^{-1} and R, respectively.

The cross-spectral field-correlation function $G^{(1,1)}(x_1',x_1)$, which for $\vec{r}_1'=\vec{r}_1$ determines the counting rate at a single point, can easily be found, in the weak field limit under discussion, from the solutions for the function $G^{(0,2)}(x_2,x_1)$, which determines the coincidence counting rate at two different points. In particular, it is a simple matter to show that in the state represented in Eq. (2.4), the functions defined by Eqs. (2.11), (2.2), and (2.3) obey the identity

$$G^{(1,1)}(x_1',x_1)=\frac{2i}{\hbar c^2}\int d^3r_2\,G^{(2,0)}(x_1',x_2)\times\frac{\partial}{\partial t_2}G^{(0,2)}(x_2,x_1), \tag{4.20}$$

in which the time t_2 is arbitrary. When Eqs. (4.2) and (4.12) are substituted into this relation, one finds that $G^{(1,1)}$ has the form

$$G^{(1,1)}(x_1',x_1)=r_1^{-2}\int d\omega_1 e^{i\omega_1(\bar{t}_1'-\bar{t}_1)}\times I(\omega_1;\hat{r}_1',\hat{r}_1)c/2\omega_1^2, \tag{4.21}$$

where the function I is given as

$$I(\omega_1;\hat{r}_1',\hat{r}_1)=\frac{2\hbar\omega_1^4\omega_2|E_0|^2}{\pi(4\pi c)^4}\int d^3k_2\chi^*\chi v_2'\times F(\theta_2)n_2^{-2}\delta(\omega(\vec{k}_2)+\omega_1-\omega_0)\times\zeta^*(\vec{k}_2+\vec{k}_1'-\vec{k}_0)\zeta(\vec{k}_2+\vec{k}_1-\vec{k}_0). \tag{4.22}$$

The function $F(\theta)$ in this relation is the ratio between external and internal solid angles, and n_2 is the index of refraction k_2c/ω_2.

In the limit $R\gg l$, for the Gaussian beam described in Eq. (4.17), it can be shown that the function $I(\omega_1;\hat{r}_1',\hat{r}_1)$ is well approximated in the isotropic case by the relation

$$I(\omega_1;\hat{r}_1',\hat{r}_1)=K\frac{\sin^2(\frac{1}{2}\rho l\delta\omega_1)}{(\frac{1}{2}\rho l\delta\omega_1)^2}\exp\left(-\frac{R^2\omega_1^2}{4c^2}[(\delta\theta_{1e})^2\cos^2\theta_{1e}+(\delta\varphi_1)^2\sin^2\theta_{1e}]\right)\quad(R\gg l) \tag{4.23a}$$

in which $\delta\omega_1\equiv\omega_1-\omega_1^*$, $\delta\theta_{1e}\equiv\theta_{1e}'-\theta_{1e}$, and $\delta\varphi_1\equiv\varphi_1'-\varphi_1$. The parameters K and ρ are defined by the relations

$$K=\frac{\hbar\omega_1^4\omega_2\chi^*\cdot\chi F_2 v_2' P_0 l^2}{(4\pi)^3c^5n_0n_2^2|v_{2z}|} \tag{4.23b}$$

and

$$\rho=v_{2z}^{-1}(\vec{v}_1-\vec{v}_2)\cdot\frac{d\vec{k}_1}{d\omega_1}, \tag{4.23c}$$

where P_0 is the pump-field power. The cross-spectral correlation function $G^{(1,1)}$ is thus characterized in this limit by angular widths proportional to R^{-1} in both the θ and φ directions, and by a spectral width proportional to l^{-1}.

The nonzero spectral width of the function I is, in general, due both to the finite correlation time and to the angular uncertainty in detecting the complementary photon at frequency ω_2. The spectral intensity $I(\omega_1;\hat{r}_1,\hat{r}_1)\equiv I(\omega_1,\hat{r}_1)$ may be expressed in terms of the function $\psi(\tau;\hat{r}_2,\hat{r}_1)$ by means of the relation

$$I(\omega_1,\hat{r}_1)=\frac{\hbar\omega_1^4\omega_2^3|E_0|^2\chi^*\cdot\chi}{64\pi^4c^6}\times\int d\Omega_{2e}|\tilde{\psi}(\omega_1-\omega_1^*;\hat{r}_2,\hat{r}_1)|^2, \tag{4.24}$$

where the integration is over the external solid angle, and the function $\tilde{\psi}$ is defined as

$$\tilde{\psi}(\omega;\hat{r}_2,\hat{r}_1)\equiv(2\pi)^{-1/2}\int d\tau e^{-i\omega\tau}\psi(\tau;\hat{r}_2,\hat{r}_1). \tag{4.25}$$

The total intensity (radiated power per unit solid angle) in the direction $\hat{r}_1$ is obtained simply by eliminating the frequency δ function in Eq. (4.22)

and setting $\hat{r}_1' = \hat{r}_1$. One readily finds, for sufficiently large values of R and l (though independently of their relative magnitudes) the relation

$$\frac{dP_1}{d\Omega_{1e}} = \frac{2\hbar\omega_1^4\omega_2|\chi|^2F_2P_0l}{(8\pi)^2c^5n_0n_2^2}\,\frac{v_2'}{|(\vec{v}_1-\vec{v}_2)\cdot d\vec{k}_1/d\omega_1|}\,. \quad (4.26)$$

In the absence of linear dispersion, $d\vec{k}_1/d\omega_1 = \hat{k}_1/v_1'$, and this expression reduces to the result found by Kleinman.[7,11] The modification which arises when linear dispersion is present is due to the fact that waves of different frequency which propagate in the same direction outside the target originate from waves propagating in different directions inside the target, if the index of refraction is frequency dependent.

V. HIGHER-ORDER STATISTICS AND PARAMETRIC GAIN

The approximate nature of the results found in the preceding section is evident from the fact that the expressions which were found for the coincidence counting rate fall identically to zero outside limited coherence regions. This is due, of course, to the assumption that only two photons are present in the scattered field. What was omitted is the possibility of simultaneously detecting two photons which are members of two different parametrically generated pairs. One should expect the coincidence counting rate to fall to the "accidental" counting rate at sufficiently separated space-time points, i.e., to the product of the individual one-photon rates. The accidental rate, for small P_0, is proportional to P_0^2, and thus cannot appear in the result of a lowest-order analysis.

In this section, the restriction on the pump-field intensity implicit in the analysis of the preceding section will be removed. Solutions will be found which are valid to arbitrary powers of the pump-field amplitude, and from which field-correlation functions of arbitrary order may be evaluated. The contributions due to multiple pair emissions, even those which occur frequently enough to interfere coherently with one another, will be fully represented, and thus the effect of parametric gain will be found. The pump field will still be represented by a prescribed unattenuated function, however, thus implying that the probability remains small that a single pump-field photon is affected by the target.

Let us denote by $\delta\alpha_{k_2k_1}$ the momentum-space wave function (expressed, for convenience, in terms of discrete-mode basis functions) of the photon pair which is emitted by an infinitesimal fraction of the atoms within the target. The field state vector due to these atoms alone is then

$$\left(1+\tfrac{1}{2}\sum_{k_2,k_1} a_{k_2}^\dagger a_{k_1}^\dagger \delta\alpha_{k_2k_1}\right)|0\rangle = \left(1+\tfrac{1}{2}\sum_{k_2,k_1}\left(a_{k_2}^\dagger a_{k_1}^\dagger\delta\alpha_{k_2k_1} - \delta\alpha^*{}_{k_2k_1}a_{k_2}a_{k_1}\right)\right)|0\rangle\,. \quad (5.1)$$

The term added on the right-hand side makes the operator multiplying the vacuum state unitary. It is clear that the field state vector which results from the emission of photon pairs by all of the atoms within the target can be found by multiplying the vacuum state by products of similar unitary operators for each infinitesimal fraction of the atoms in the target. When this is done in a symmetrical way, the result is

$$|\rangle = \exp\left(\tfrac{1}{2}\sum_{k_2,k_1}\left(a_{k_2}^\dagger a_{k_1}^\dagger\alpha_{k_2k_1} - \alpha^*{}_{k_2k_1}a_{k_2}a_{k_1}\right)\right)|0\rangle\,. \quad (5.2)$$

The quantity $\alpha_{k_2k_1}$ in this relation is just the full wave function for the photon pair emitted by the entire target, *as calculated in lowest order.* (It is linear in the incident field amplitude E_0 in the case under discussion.)

It is perhaps worthwhile to show that the method used here gives results in agreement with those obtained by more familiar methods, in the closely related two-mode parametric amplification process.[5] For that case, by elementary perturbation-theory arguments one is led to write, as in Eq. (5.1), the contribution made by an infinitesimal fraction of the atoms in the cavity to the (interaction picture) state vector for the system at time t as $(1+i\delta\kappa t a^\dagger b^\dagger)|0\rangle$, where $a^\dagger$ and $b^\dagger$ are the creation operators for the two modes, and $\delta\kappa$ is the contribution of the atoms in question to the nonlinear pair-emission coefficient κ. By steps similar to those used to reach Eq. (5.2), one is then led to write $|t\rangle = \exp[i\kappa t(a^\dagger b^\dagger + ab)]|0\rangle$ for the state vector due to all of the atoms. This is just $e^{-iHt/\hbar}|0\rangle$, where H is exactly the Hamiltonian which expresses the parametric coupling between the two modes, and leads to exponential increases in the energy of each.

In order to show how the result in Eq. (5.2) enables one to write down closed solutions for the field-correlation functions in terms of those evaluated in lowest order in Sec. IV, it is convenient to begin by introducing the characteristic or moment-generating function[8]

$$\chi(\{\eta_k\}) \equiv \left\langle\exp\left(\sum_k (a_k^\dagger\eta_k - \eta_k^* a_k)\right)\right\rangle \equiv \langle e^{a^\dagger\eta - \eta^* a}\rangle\,. \quad (5.3)$$

This function can be evaluated in the state defined by Eq. (5.2) most easily by introducing a dimensionless parameter λ into the exponential function on the right-hand side, thus defining the unitary

operator

$$U(\lambda) \equiv \exp[\tfrac{1}{2}\lambda(a^\dagger a^\dagger : \alpha - \alpha^* : aa)], \tag{5.4}$$

in which α is the symmetrical second-rank tensor with elements $\alpha_{k_1 k_2}$. The characteristic function $\chi(\eta)$ is then given as

$$\chi(\eta) = \langle 0 | e^{a^\dagger(\lambda)\eta - \eta^* a(\lambda)} | 0 \rangle |_{\lambda=1}, \tag{5.5}$$

where

$$a(\lambda) \equiv U^{-1}(\lambda) a U(\lambda). \tag{5.6}$$

By differentiating Eqs. (5.6) and (5.4), one finds

$$\frac{d}{d\lambda} a(\lambda) = a^\dagger(\lambda)\alpha; \quad \frac{d}{d\lambda} a^\dagger(\lambda) = \alpha^* a(\lambda). \tag{5.7}$$

The solution to these equations, subject to the initial condition $a(0) = a$ is

$$\begin{aligned} a(\lambda) &= (\cosh\beta\lambda) a + a^\dagger \left(\frac{\sinh\beta\lambda}{\beta}\right)\alpha, \\ a^\dagger(\lambda) &= a^\dagger \cosh\beta\lambda + \alpha^* \left(\frac{\sinh\beta\lambda}{\beta}\right) a, \end{aligned} \tag{5.8}$$

where β is the Hermitian matrix

$$\beta \equiv (\alpha\alpha^*)^{1/2}. \tag{5.9}$$

Only positive powers of $\beta^2 = \alpha\alpha^*$, it should be noted, appear in Eqs. (5.8).

By substituting Eqs. (5.8) into Eq. (5.5) and using the result to evaluate the *normally ordered* characteristic function[8]

$$\chi_N(\eta) \equiv \langle e^{a^\dagger \eta} e^{-\eta^* a} \rangle = e^{(1/2)|\eta|^2} \chi(\eta) \tag{5.10}$$

(where $|\eta|^2 \equiv \sum_k |\eta_k|^2$), one finds the relation

$$\chi_N(\eta) = \exp\left\{ -\eta^*(\sinh^2\beta)\eta + \left[\tfrac{1}{2}\eta^*\eta^* : \left(\frac{\sinh 2\beta}{2\beta}\alpha\right) + \text{c.c.} \right] \right\}. \tag{5.11}$$

The transcription of this relation into terms relating to continuous functions of space and time is straightforward. One begins by introducing the normally ordered *characteristic functional*

$$\chi_N[\mathcal{E}(x)] \equiv \langle \exp[(2i/\hbar c)\int d^3r A^{(-)}(x)\mathcal{E}(x)] \times \exp[(2i/\hbar c)\int d^3r \mathcal{E}^*(x) A^{(+)}(x)] \rangle, \tag{5.12}$$

where $A^{(+)}(x)$ and $A^{(-)}(x)$ are the positive- and negative-frequency parts of the vector-potential field operator, and $\mathcal{E}(x)$ is a freely propagating, positive-frequency c-number function. One may think of the matrix β as the momentum-space representative of an operator which acts on one-particle state vectors, and of the second-rank tensor α as the representative of a two-particle state vector. One is led to introduce the quantities

$$G^{(0,2)} \equiv \tfrac{1}{2}\hbar c^2 \omega^{-1/2}\omega^{-1/2} : \left(\frac{\sinh 2\beta}{2\beta}\alpha\right) \tag{5.13}$$

and

$$G^{(1,1)*} \equiv \tfrac{1}{2}\hbar c^2 \omega^{-1/2}(\sinh^2\beta)\omega^{-1/2},$$

where the "frequency operator" ω is defined to have eigenvalue $\omega = kc$ when operating on a state with configuration-space wave function $e^{i\mathbf{k}\cdot\mathbf{r}}$. The characteristic functional defined by Eq. (5.12) is found from Eq. (5.11) to have the value

$$\chi_N[\mathcal{E}(x)] = \exp\left[-\left(\frac{2}{\hbar c}\right)^2 \int\int d^3r d^3r' \mathcal{E}^*(x) G^{(1,1)*}(x,x') \mathcal{E}(x') - \frac{1}{2}\left(\frac{2}{\hbar c}\right)^2 \left[\int\int d^3r_2 d^3r_1 \mathcal{E}^*(x_2)\mathcal{E}^*(x_1) G^{(0,2)}(x_2,x_1) + \text{c.c.} \right] \right], \tag{5.14}$$

where the functions $G^{(0,2)}(x_2, x_1)$ and $G^{(1,1)*}(x, x') \equiv G^{(1,1)}(x', x)$ are the configuration-space representatives of the objects defined by Eqs. (5.13). That these functions are in fact the field-correlation functions defined by Eqs. (2.2) and (2.11) may be shown simply by expanding the exponential in Eq. (5.14) and comparing the result to the definition in Eq. (5.12).

In the limit of weak pump fields, α and β approach zero, and Eqs. (5.13) and (5.9) reduce to the expressions

$$G_0^{(0,2)} = \tfrac{1}{2}\hbar c^2 \omega^{-1/2}\omega^{-1/2} : \alpha, \tag{5.15a}$$

$$\begin{aligned} G_0^{(1,1)} &= \tfrac{1}{2}\hbar c^2 (\omega^{-1/2}\alpha\alpha^*\omega^{-1/2})^* \\ &= (2/\hbar c^2) G_0^{(2,0)} \omega G_0^{(0,2)}, \end{aligned} \tag{5.15b}$$

where the latter relation follows from Eq. (5.15a) and its complex conjugate. Equation (5.15b) is in fact equivalent to the relation already noted in Eq. (4.20) as existing, in lowest order, between the functions $G^{(1,1)}$ and $G^{(0,2)}$.

By using Eq. (5.15a) to express α in terms of $G_0^{(0,2)}$, one finds with the aid of Eqs. (5.13), (5.9), and (5.15b), that $G^{(0,2)}$ and $G^{(1,1)}$ are given by the relations

$$G^{(0,2)} = \omega^{-1/2} f_1\left(\frac{2}{\hbar c^2}\omega^{1/2} G_0^{(1,1)*} \omega^{1/2}\right) \times \omega^{1/2} G_0^{(0,2)}, \tag{5.16a}$$

$$G^{(1,1)*} = \tfrac{1}{2}\hbar c^2 \omega^{-1/2} \times f_2\left(\frac{2}{\hbar c^2}\omega^{1/2} G_0^{(1,1)*} \omega^{1/2}\right)\omega^{-1/2}, \tag{5.16b}$$

in which $G_0^{(0,2)}$ and $G_0^{(1,1)}$ are the corresponding

lowest-order solutions, and the (analytic) functions $f_1(x)$ and $f_2(x)$ are defined as

$$f_1(x) \equiv (\sinh 2\sqrt{x})/2\sqrt{x}\ ,$$
$$f_2(x) \equiv \sinh^2\sqrt{x}\ . \tag{5.17}$$

Together with the results of Sec. IV (in which $G_0^{(0,2)}$ and $G_0^{(1,1)}$ are evaluated), then Eqs. (5.16), (5.17), and (5.14) represent a complete solution, valid to all orders in the pump-field strength, for the statistical properties of the radiated field. The solution so represented involves integrations over the limited regions of space in which the lowest-order field-correlation functions are nonvanishing, and thus involves the values of these functions only at points far from the target.

The multiple integrations represented formally in Eqs. (5.16) are not difficult to carry out in many limiting cases of interest. In the case described in the weak-pump-field limit by Eqs. (4.23), for example, one finds by making use of Eqs. (5.16b), (4.21), and (4.23a) a relation expressing the spectral function $I(\omega_1; \hat{r}_1', \hat{r}_1)$ as a power series consisting of terms similar to the one given by Eq. (4.23a), but with ever-increasing angular widths. The series is governed by the dimensionless parameter

$$u = I_0(\omega_1, \hat{r}_1)\, 8\pi^2 c^2/\hbar\omega_1{}^3 R^2 \cos\theta_{1e}\,, \tag{5.18}$$

where $I_0(\omega_1, \hat{r}_1)$ is the intensity as given in the weak-pump-field limit by Eqs. (4.23) with $\hat{r}_1' = \hat{r}_1$,[7]

$$I_0(\omega_1, \hat{r}_1) = \frac{\hbar\omega_1^4\omega_2|\chi|^2 F_2 v_2' P_0 l^2}{(4\pi)^3 c^5 n_0 n_2^2}\ \frac{\sin^2(\frac{1}{2}\rho l\delta\omega_1)}{(\frac{1}{2}\rho l\delta\omega_1)^2}\,. \tag{5.19}$$

For pump fields of arbitrary strength, the spectral intensity at frequency ω_1 is found to be given, in the case under discussion, by the relation

$$I(\omega_1, \hat{r}_1) = I_0(\omega_1, \hat{r}_1)\, \mathfrak{F}(u), \tag{5.20}$$

where $\mathfrak{F}(u)$ is the function

$$\mathfrak{F}(u) = \frac{2}{u}\int_0^{\sqrt{u}} \frac{\sinh^2 y}{y}\, dy = \sum_{n=1}^{\infty} \frac{2^{2n-1}u^{n-1}}{n(2n)!}\,. \tag{5.21}$$

The parameter u, as given by Eq. (5.18), is roughly equal, in lowest order, to the number of photons with frequency between ω_1 and $\omega_1 + d\omega_1$ which pass by a given point in time $1/d\omega_1$, within the solid-angular width $\Delta\Omega_1 \sim c^2/R^2\omega_1{}^2$ specified by Eq. (4.23a). When this number becomes comparable to unity, the lowest-order expression $I_0(\omega_1)$ for the spectral density ceases to be applicable, and it becomes necessary to use instead the relation (5.20), which shows the effect of parametric gain. Since the gain is greater where I_0 is greater, the relative intensity is increased at the center of the emission line, and the spectral width is consequently reduced by the parametric-gain process.

The solutions for the field-correlation functions $G^{(1,1)}$ and $G^{(0,2)}$ enable one to construct the characteristic functional given by Eq. (5.14), and hence to evaluate field-correlation functions of arbitrarily high order. The four-point function defined by Eq. (2.1), for example, is given by Eqs. (5.12) and (5.14) as

$$\begin{aligned} G^{(2,2)}(x_1', x_2'; x_2, x_1) = {} & G^{(2,0)}(x_1', x_2')G^{(0,2)}(x_2, x_1) \\ & + G^{(1,1)}(x_1', x_1)G^{(1,1)}(x_2', x_2) \\ & + G^{(1,1)}(x_1', x_2)G^{(1,1)}(x_2', x_1), \end{aligned} \tag{5.22}$$

to all orders in the pump-field power P_0. For weak pump fields, the first term on the right-hand side of this relation is proportional to P_0, while the second and third terms are proportional to P_0^2. The latter two terms were omitted in the approximate relation (2.5) and in the analysis of Sec. IV. They are necessary even in lowest order, however, to evaluate the coincidence counting rate at points x_1 and x_2 so far separated from one another as to make the function $G^{(0,2)}$ vanishingly small.

The function given by Eq. (5.22), it may be noted, has the form characteristic of Gaussian fields when $G^{(0,2)} = 0$. Indeed, it is clear from the solution in Eq. (5.14) that the field statistics of arbitrarily high order are Gaussian, provided that the detection points lie outside the two-photon coherence regions. The transition from the familiar classically understandable Gaussian statistics to the highly correlated intrinsically quantum-mechanical statistics which takes place as the detection points are brought within the (mutual) two-photon coherence regions is one of the most interesting features of the parametric frequency-splitting process. It is one of the simplest physically realizable electromagnetic processes for which solutions can be obtained in closed form, and which exhibits such specifically quantum-mechanical features.

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