Theory of parametric frequency down conversion of light

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A theoretical study is made of the process in which incident pump photons that interact with a nonlinear medium (such as a crystal lacking inversion symmetry) are spontaneously split into lower-frequency signal and idler photons. The down-converted fields are quantized and described by a continuum of modes, a subset of which interacts with each photodetector. It is shown that when two ideal photodetectors are appropriately located so that they receive the conjugate signal and idler photons, then the joint probability of two-photon detection by the two detectors can equal the single-photon detection probability. The time correlation between the two detected photons is shown to be limited either by the resolving time of the detectors, or by the bandwidth of the down-converted light, and to be independent of the coherence time of the pump field or of the length of the nonlinear medium. These conclusions are compared with the results of recent experiments.

I. INTRODUCTION

The process of parametric amplification and oscillation, in which two or more modes of the electromagnetic field are coupled through a nonlinear medium, has already been the subject of numerous studies in the past.[1–18] The focus in these treatments has ranged over a variety of topics, such as the creation of nonclassical photon statistics[3,7,8,10–15] and of squeezed quantum states.[4,17] Of particular interest is the process of parametric frequency down conversion, in which an incident pump photon effectively splits up into two lower-frequency (signal and idler) photons, which constitute a highly correlated photon pair.[6,16] This phenomenon was first observed in photon coincidence counting experiments,[19] and more recent time-resolved correlation measurements of the same process have been reported.[20]

Except for the less-than-perfect detection efficiencies of the photoelectric detectors, the joint probability for the detection of the two down-converted photons by two appropriately located detectors can be as large as the probability for the detection of one photon by one detector. This remarkable result readily follows from the parametric Hamiltonian that couples the pump mode to the signal and idler modes in the interaction, and can be obtained with the help of the general approach of Graham,[16] for example. However, the effect is not nearly so obvious when the field is treated more realistically by a multimode expansion, and when the geometry of the nonlinear medium and of the detectors is taken into account. Moreover, treatments based on just a few discrete modes are unable to determine a meaningful correlation time for the two-photon correlation function.

This turns out to be a particularly interesting problem from the experimental point of view. In their pioneering experiments, Burnham and Weinberg[19] found that the measured value of the correlation time T_c between the two down-converted photons was limited by the time resolution of the electronics, but it was of order or less than 4 nsec. They speculated that T_c might be related to the coherence time (2×10^{-10} sec) of the incident light beam, which was, however, far below the instrumental resolution limit. On the other hand, more recent experiments with a single-mode laser having a coherence time >40 nsec indicate that T_c can be several orders of magnitude smaller than the coherence time of the pump light.[20] However, a theoretical determination of the photon correlation time T_c requires a more realistic treatment of the down-conversion problem, with a continuum of modes, rather than the two- and three-mode calculations that have generally been given. The one exception to this is the previous treatment by Mollow,[6] who expressed the correlation function of the down-converted light in terms of the susceptibilities of the nonlinear medium. However, that treatment does not start from an explicit Hamiltonian, and it does not clearly bring out which physical parameters determine the correlation time T_c.

In the following we use a simple model Hamiltonian to describe the coupling of the incident pump field to the down-converted signal and idler fields over a region that coincides with the volume of the nonlinear medium. The down-converted fields are decomposed into an infinite set of modes, which is eventually treated as a continuum. The Heisenberg equations of motion for the field operators are integrated over a short-time interval corresponding to the propagation time through the medium, and certain expectation values are calculated. We show that when the directions, the frequencies, and the detection time intervals are appropriately chosen, the joint probability of two photon detections at two ideal detectors equals the single-photon detection probability. The two-photon correlation function is examined, and is found to have a range of the order of the resolving time of the photoelectric detectors, with a lower limit set by the reciprocal bandwidth of the down-converted photons. This means that the time interval between the signal and idler photons could, in principle, be in the subpicosecond range. These conclusions are then compared with the results of some recent experiments.[20]

II. EQUATIONS OF MOTION

It is well known that in a nonlinear dielectric an incident electric field **E** will create a polarization **P** having contributions that are not only linear in **E**, but also bilinear, trilinear, etc.[21] The lowest order nonlinearity can be written

$$P_i \to \widetilde{\chi}_{ijk} E_j E_k \ , \quad (1)$$

where $\widetilde{\chi}_{ijk}$ is the bilinear susceptibility. This makes a contribution to the energy of the electromagnetic field of the form

$$H_I = \tfrac{1}{2} \int_{\mathscr{V}} P_i(\mathbf{r},t) E_i(\mathbf{r},t) d^3x$$

$$= \tfrac{1}{2} \int_{\mathscr{V}} \widetilde{\chi}_{ijk} E_i(\mathbf{r},t) E_j(\mathbf{r},t) E_k(\mathbf{r},t) d^3x \ , \quad (2)$$

where the integration extends over the volume $\mathscr{V}$ of the nonlinear medium. However, in a typical parametric process all three fields may oscillate at different frequencies, and the susceptibility may also depend on these frequencies. We then need to decompose the electric field into its Fourier components $\mathscr{E}(\mathbf{r},\omega)$ and write, in place of Eq. (2),[21]

$$H_I = \tfrac{1}{2} \int_{\mathscr{V}} d^3x \int\int\int d\omega\, d\omega'\, d\omega'' \widetilde{\chi}_{ijk}(\mathbf{r};\omega,\omega',\omega'') \times \mathscr{E}_i(\mathbf{r},\omega) \mathscr{E}_j(\mathbf{r},\omega') \mathscr{E}_k(\mathbf{r},\omega'') \quad (3)$$

for the interaction energy.

When the field is quantized, $\mathbf{E}(\mathbf{r},t)$ becomes a Hilbert-space operator $\hat{\mathbf{E}}(\mathbf{r},t)$.[22] This can be decomposed into its positive-frequency and negative-frequency parts $\hat{\mathbf{E}}^{(+)}(\mathbf{r},t)$ and $\hat{\mathbf{E}}^{(-)}(\mathbf{r},t)$, and given a mode expansion in plane-wave modes of the form

$$\hat{\mathbf{E}}^{(+)}(\mathbf{r},t) = \frac{1}{L^{3/2}} \sum_{\mathbf{k},s} i \left[\frac{\hbar\omega(\mathbf{k})}{2\epsilon_0} \right]^{1/2} \hat{a}_{\mathbf{k}s}(t) \boldsymbol{\varepsilon}_{\mathbf{k}s} e^{i\mathbf{k}\cdot\mathbf{r}} \ . \quad (4)$$

L^3 is the quantization volume which is later allowed to become infinite. $\boldsymbol{\varepsilon}_{\mathbf{k}s}$ is a unit polarization vector depending on the wave vector **k** and the polarization index s ($s=1,2$), and $\hat{a}_{\mathbf{k}s}(t)$ is the photon annihilation operator for the mode $\mathbf{k},s$ of frequency $\omega(\mathbf{k})$. In the following we shall find it a little more convenient to work not with $\hat{\mathbf{E}}^{(+)}(\mathbf{r},t)$ but with another positive-frequency field operator[23]

$$\hat{\mathbf{V}}(\mathbf{r},t) \equiv \frac{1}{L^{3/2}} \sum_{\mathbf{k},s} \hat{a}_{\mathbf{k}s}(t) \boldsymbol{\varepsilon}_{\mathbf{k}s} e^{i\mathbf{k}\cdot\mathbf{r}} \ , \quad (5)$$

whose square $\hat{\mathbf{V}}^\dagger \cdot \hat{\mathbf{V}}$ has the dimensions of photon density. The integral of $\langle \hat{\mathbf{V}}^\dagger(\mathbf{r},t) \cdot \hat{\mathbf{V}}(\mathbf{r},t) \rangle$ over a volume whose linear dimensions are large compared with any optical wavelength provides a measure of the probability for detecting a photon within that volume.[23] We then write for the total energy of the field, including the parametric interaction,

$$\hat{H} = \sum_{\mathbf{k},s} \hbar\omega(\mathbf{k}) \hat{n}_{\mathbf{k}s} + \left[\int_{\mathscr{V}} d^3x\, \chi_{ijl}(\mathbf{r}) \hat{V}_i^\dagger(\mathbf{r},t) \hat{V}_j^\dagger(\mathbf{r},t) V_l(\mathbf{r},t) + \text{H.c.} \right] , \quad (6)$$

where $\chi_{ijl}(\mathbf{r})$ is another kind of "susceptibility" with different dimensions, or more generally, when the susceptibility is frequency dependent,

$$\hat{H} = \sum_{\mathbf{k},s} \hbar\omega(\mathbf{k}) \hat{n}_{\mathbf{k}s} + \int_{\mathscr{V}} d^3x \frac{1}{L^3} \sum_{\mathbf{k}',s'} \sum_{\mathbf{k}'',s''} \chi_{ijl}(\mathbf{r};\omega(\mathbf{k}_0),\omega(\mathbf{k}'),\omega(\mathbf{k}'')) \hat{a}^\dagger_{\mathbf{k}'s'}(t) \hat{a}^\dagger_{\mathbf{k}''s''}(t) V_l \times (\boldsymbol{\varepsilon}^*_{\mathbf{k}'s'})_i (\boldsymbol{\varepsilon}^*_{\mathbf{k}''s''})_j e^{i[(\mathbf{k}_0-\mathbf{k}'-\mathbf{k}'')\cdot\mathbf{r} - \omega(\mathbf{k}_0)t]} \ . \quad (7)$$

The wave vectors appearing in this equation are those within the nonlinear medium. In writing $\hat{H}$ in this way we have assumed that the incident field is so intense that it can be treated classically as a monochromatic plane wave with wave vector $\mathbf{k}_0$ and frequency $\omega(\mathbf{k}_0)$. This is an approximation, but one that is usually acceptable for a laser beam, so long as the beam is only weakly attenuated in passing through the nonlinear medium (or crystal). We have therefore put

$$\mathbf{V}(\mathbf{r},t) = \mathbf{V} e^{i[\mathbf{k}_0\cdot\mathbf{r} - \omega(\mathbf{k}_0)t]} \quad (8)$$

for the incident field. If the crystal is spatially uniform and in the form of a rectangular parallelepiped centered at $\mathbf{r}_0$ with sides l_1, l_2, l_3, the space integration is easily carried out. We then obtain for the energy

$$\hat{H} = \sum_{\mathbf{k},s} \hbar\omega(\mathbf{k}) \hat{n}_{\mathbf{k}s} + \frac{1}{L^3} \sum_{\mathbf{k}',s'} \sum_{\mathbf{k}'',s''} \chi_{ijl}(\omega(\mathbf{k}_0),\omega(\mathbf{k}'),\omega(\mathbf{k}'')) \hat{a}^\dagger_{\mathbf{k}'s'}(t) \hat{a}^\dagger_{\mathbf{k}''s''}(t) (\boldsymbol{\varepsilon}^*_{\mathbf{k}'s''})_i (\boldsymbol{\varepsilon}^*_{\mathbf{k}''s''})_j V_l \times e^{i[(\mathbf{k}_0-\mathbf{k}'-\mathbf{k}'')\cdot\mathbf{r}_0 - \omega(\mathbf{k}_0)t]} \prod_{m=1}^{3} \left[\frac{\sin[\frac{1}{2}(\mathbf{k}_0-\mathbf{k}'-\mathbf{k}'')_m l_m]}{\frac{1}{2}(\mathbf{k}_0-\mathbf{k}'-\mathbf{k}'')_m} \right] . \quad (9)$$

This Hamiltonian can be used to derive the Heisenberg equations of motion for the annihilation operators $\hat{a}_{\mathbf{k}s}(t)$. It is a little more convenient to work with slowly varying dynamical variables

$$\hat{A}_{\mathbf{k}s}(t) \equiv \hat{a}_{\mathbf{k}s}(t) e^{i\omega(\mathbf{k})t} \quad (10)$$

that have the highly oscillatory behavior of $\hat{a}_{\mathbf{k}s}(t)$ canceled out. We then find

$$\dot{\hat{A}}_{\mathbf{k}s}(t)=\frac{1}{i\hbar}[\hat{a}_{\mathbf{k}s},\hat{H}]e^{i\omega(\mathbf{k})t}+i\omega(\mathbf{k})\hat{A}_{\mathbf{k}s}(t)$$

$$=\frac{1}{i\hbar L^3}\sum_{\mathbf{k}',s'}\sum_{\mathbf{k}'',s''}\chi_{ijl}(\omega(\mathbf{k}_0),\omega(\mathbf{k}'),\omega(\mathbf{k}''))[\hat{a}_{\mathbf{k}s},\hat{a}^{\dagger}_{\mathbf{k}'s'}\hat{a}^{\dagger}_{\mathbf{k}''s''}]e^{i\omega(\mathbf{k})t}(\boldsymbol{\varepsilon}^*_{\mathbf{k}'s'})_i(\boldsymbol{\varepsilon}^*_{\mathbf{k}''s''})_j V_l$$

$$\times e^{i[(\mathbf{k}_0-\mathbf{k}'-\mathbf{k}'')\cdot\mathbf{r}_0-\omega(\mathbf{k}_0)t]}\prod_{m=1}^{3}\left[\frac{\sin[\frac{1}{2}(\mathbf{k}_0-\mathbf{k}'-\mathbf{k}'')_m l_m]}{\frac{1}{2}(\mathbf{k}_0-\mathbf{k}'-\mathbf{k}'')_m}\right]$$

$$=\frac{1}{i\hbar L^3}\sum_{\mathbf{k}',s'}\chi_{ijl}(\omega(\mathbf{k}_0),\omega(\mathbf{k}),\omega(\mathbf{k}'))\hat{A}^{\dagger}_{\mathbf{k}'s'}(t)[(\boldsymbol{\varepsilon}^*_{\mathbf{k}s})_i(\boldsymbol{\varepsilon}^*_{\mathbf{k}'s'})_j+(\boldsymbol{\varepsilon}^*_{\mathbf{k}'s'})_i(\boldsymbol{\varepsilon}^*_{\mathbf{k}s})_j]V_l$$

$$\times e^{i(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}_0}e^{-i[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')]t}\prod_{m=1}^{3}\left[\frac{\sin[\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')_m l_m]}{\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')_m}\right], \tag{11}$$

after making use of the symmetry property with respect to the two down-converted waves $\chi_{ijl}(\omega(\mathbf{k}_0),\omega(\mathbf{k}'),\omega(\mathbf{k}))=\chi_{ijl}(\omega(\mathbf{k}_0),\omega(\mathbf{k}),\omega(\mathbf{k}'))$.

We now integrate the equation of motion over the short interaction time Δt for which the modes are coupled through the nonlinear medium. This time Δt may be taken to be the effective propagation time of the light through the crystal. In practice Δt is typically a fraction of a nanosecond, and the intensities of the down-converted light beams are generally so low that we may replace, to a good approximation, $\hat{A}^{\dagger}_{\mathbf{k}'s'}(t)$ under the time integral by $\hat{A}^{\dagger}_{\mathbf{k}'s'}(0)$. The integration over time is then trivial, and we obtain

$$\hat{A}_{\mathbf{k}s}(\Delta t)=\hat{A}_{\mathbf{k}s}(0)+\frac{1}{i\hbar L^3}\sum_{\mathbf{k}',s'}\chi_{ijl}(\omega(\mathbf{k}_0),\omega(\mathbf{k}),\omega(\mathbf{k}'))\hat{A}^{\dagger}_{\mathbf{k}'s'}(0)[(\boldsymbol{\varepsilon}^*_{\mathbf{k}s})_i(\boldsymbol{\varepsilon}^*_{\mathbf{k}'s'})_j+(\boldsymbol{\varepsilon}^*_{\mathbf{k}'s'})_i(\boldsymbol{\varepsilon}^*_{\mathbf{k}s})_j]V_l$$

$$\times e^{i(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}_0}e^{-(i/2)[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')]\Delta t}\frac{\sin\{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')]\Delta t\}}{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')]}$$

$$\times\prod_{m=1}^{3}\left[\frac{\sin[\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')_m l_m]}{\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')_m}\right]. \tag{12}$$

After the time Δt when the interaction is over, $\hat{A}_{\mathbf{k}s}(t)$ evolves essentially as a free-field mode, with

$$\hat{A}_{\mathbf{k}s}(t)=\hat{A}_{\mathbf{k}s}(\Delta t)\quad\text{for } t\geq\Delta t\ . \tag{13}$$

We can use this result to construct the configuration space operator $\hat{\mathbf{V}}(\mathbf{r},t)$ given by Eq. (5). In order to avoid complications introduced by the boundaries of the nonlinear medium (or crystal), which normally generate refracted and reflected waves, we shall suppose that the crystal is embedded in a linear medium with exactly the same refractive index. The wave vector $\mathbf{k}$ then has the same value inside and outside the crystal.

Ultimately we shall be interested in calculating the response of a photodetector that receives the down-converted light at some distant point $\mathbf{r}$ at some time t. For this purpose we need the part of the total field to which the detector responds. In practice, it is often convenient to locate the detector at the focal plane of a lens,[20] possibly with a filter in front, so that it responds only to wave vectors of the field within some small range of directions and within some limited frequency range. However, we prefer to avoid the complications that the lens would introduce into the calculation, although its effects appear implicitly below. Let $[\mathbf{k}]_1$ denote the limited set of wave vectors to which the detector at $\mathbf{r}_1$ responds, where the set $[\mathbf{k}]_1$ is centered on some wave vector $\mathbf{k}_1$ and contains contributions within some range Δk_1, which is not necessarily small. We then define the response operator $\hat{\mathbf{W}}(\mathbf{r}_1,t)$ to be given by a sum like that in Eq. (5), but with $\mathbf{k}$ limited to the set $[\mathbf{k}]_1$. From Eqs. (10), (12), and (13) we then have for $t\geq\Delta t$,

$$\hat{\mathbf{W}}(\mathbf{r}_1,t)=\frac{1}{L^{3/2}}\sum_{[\mathbf{k}]_1,s}\hat{A}_{\mathbf{k}s}(0)\boldsymbol{\varepsilon}_{\mathbf{k}s}e^{i[\mathbf{k}\cdot\mathbf{r}_1-\omega(\mathbf{k})t]}$$

$$+\frac{1}{L^{3/2}}\sum_{\mathbf{k},s}\hat{A}^{\dagger}_{\mathbf{k}s}(0)\mathbf{f}(\mathbf{k},s;\mathbf{r}_1,t)$$

$$=\hat{\mathbf{W}}_{\text{free}}(\mathbf{r}_1,t)+\frac{1}{L^{3/2}}\sum_{\mathbf{k},s}\hat{A}^{\dagger}_{\mathbf{k}s}(0)\mathbf{f}(\mathbf{k},s;\mathbf{r}_1,t)\ , \tag{14}$$

where $\hat{\mathbf{W}}_{\text{free}}(\mathbf{r}_1,t)$ is the field operator in the absence of any interaction, and

$$\mathbf{f}(\mathbf{k},s;\mathbf{r}_1,t)\equiv\frac{1}{i\hbar L^3}\sum_{[\mathbf{k}']_1,s'}\chi_{ijl}(\omega(\mathbf{k}_0),\omega(\mathbf{k}),\omega(\mathbf{k}'))\boldsymbol{\varepsilon}_{\mathbf{k}'s'}T_{ij}{}^*(\mathbf{k},s;\mathbf{k}',s')V_l e^{i(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}_0}$$
$$\times e^{i[\mathbf{k}'\cdot\mathbf{r}_1-\omega(\mathbf{k}')t]}e^{-(i/2)[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')]\Delta t}\frac{\sin\{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')]\Delta t\}}{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')]}$$
$$\times\prod_{m=1}^{3}\left[\frac{\sin[\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')_m l_m]}{\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')_m}\right], \tag{15}$$

with

$$T_{ij}(\mathbf{k},s;\mathbf{k}',s')\equiv(\boldsymbol{\varepsilon}_{\mathbf{k}s})_i(\boldsymbol{\varepsilon}_{\mathbf{k}'s'})_j+(\boldsymbol{\varepsilon}_{\mathbf{k}'s'})_i(\boldsymbol{\varepsilon}_{\mathbf{k}s})_j\ . \tag{16}$$

In expressing $\hat{\mathbf{W}}(\mathbf{r},t)$ in the form of Eq. (14) we have interchanged the variables $\mathbf{k},s$ and $\mathbf{k}',s'$.

III. PROBABILITY OF PHOTON DETECTION

Once the quantum state of the field has been specified, we can use Eq. (14) to calculate expectation values of any function of the field operators. In the special case of parametric frequency down conversion, in which the electromagnetic field of interest is that generated by the down conversion of the incident pump light, we take the initial quantum state to be the vacuum $|\text{vac}\rangle$. By virtue of the property

$$\hat{A}_{\mathbf{k}s}(0)|\text{vac}\rangle=0=\langle\text{vac}|\hat{A}^\dagger_{\mathbf{k}s}(0)\ ,$$
$$\hat{\mathbf{W}}_{\text{free}}(\mathbf{r},t)|\text{vac}\rangle=0=\langle\text{vac}|\hat{\mathbf{W}}^\dagger_{\text{free}}(\mathbf{r},t)\ , \tag{17}$$

it then follows from Eq. (14) that the averaged measured light intensity at $\mathbf{r},t$ is given by

$$\langle\hat{\mathbf{W}}^\dagger(\mathbf{r},t)\cdot\hat{\mathbf{W}}(\mathbf{r},t)\rangle=\frac{1}{L^3}\sum_{\mathbf{k},s}\sum_{\mathbf{k}',s'}\langle\hat{A}_{\mathbf{k}s}(0)\hat{A}^\dagger_{\mathbf{k}'s'}(0)\rangle\mathbf{f}^*(\mathbf{k},s;\mathbf{r},t)\cdot\mathbf{f}(\mathbf{k}',s';\mathbf{r},t)=\frac{1}{L^3}\sum_{\mathbf{k},s}|\mathbf{f}(\mathbf{k},s;\mathbf{r},t)|^2\ , \tag{18}$$

when we apply the commutation rule

$$[\hat{A}_{\mathbf{k}s}(0),\hat{A}^\dagger_{\mathbf{k}'s'}(0)]=\delta^3_{\mathbf{k}\mathbf{k}'}\delta_{ss'}\ . \tag{19}$$

In practice, the light intensity cannot actually be measured at a point and at an instant of time, but we have to integrate Eq. (18) in order to arrive at measurable quantities. If the detector has a small illuminated surface area δS normal to the direction of the incident light in the far field of the source, and if the measurement time is a short interval δt, then the detector is effectively counting the photons in a volume $\delta\mathscr{V}=\delta S u\delta t$ (but with $\delta\mathscr{V}\gg\lambda^3$), where u is the velocity of light in the medium. We shall therefore take the volume integral over $\delta\mathscr{V}$ of the average intensity given by Eq. (18) to be a measure of the probability $P_1(t)$ of photon detection by the detector at time t, and we write

$$P_1(t)=\int_{\delta\mathscr{V}}d^3x\frac{1}{L^3}\sum_{\mathbf{k},s}|\mathbf{f}(\mathbf{k},s;\mathbf{r},t)|^2\ . \tag{20}$$

In the limit $L\to\infty$, the sum over $\mathbf{k}$ can be replaced by an integral in the usual way, and with the help of Eq. (15) we obtain

$$P_1(t)=\frac{1}{\hbar^2(2\pi)^9}\int_{\delta\mathscr{V}_1}d^3x_1\int d^3k\int_{[\mathbf{k}']_1}d^3k'\int_{[\mathbf{k}'']_1}d^3k''$$
$$\times\sum_{s,s',s''}\chi^*_{ijl}(\omega(\mathbf{k}_0),\omega(\mathbf{k}),\omega(\mathbf{k}'))\chi_{uvw}(\omega(\mathbf{k}_0),\omega(\mathbf{k}),\omega(\mathbf{k}''))(\boldsymbol{\varepsilon}^*_{\mathbf{k}'s'}\cdot\boldsymbol{\varepsilon}_{\mathbf{k}''s''})$$
$$\times T_{ij}(\mathbf{k},s;\mathbf{k}',s')T^*_{uv}(\mathbf{k},s;\mathbf{k}'',s'')V^*_l V_w e^{i[(\mathbf{k}''-\mathbf{k}')\cdot(\mathbf{r}_1-\mathbf{r}_0)-\omega(\mathbf{k}'')t+\omega(\mathbf{k}')t]}e^{-(1/2)i[\omega(\mathbf{k}')-\omega(\mathbf{k}'')]\Delta t}$$
$$\times\frac{\sin\{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')]\Delta t\}}{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')]}\frac{\sin\{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}'')]\Delta t\}}{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}'')]}$$
$$\times\prod_{m=1}^{3}\left[\frac{\sin[\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')_m l_m]}{\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')_m}\right]\prod_{m=1}^{3}\left[\frac{\sin[\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}'')_m l_m]}{\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}'')_m}\right]. \tag{21}$$

It is worth observing that because of the sinc factors [sinc denotes $(\sin x)/x$], and because l_1,l_2,l_3 and Δt are typically many thousands of optical wavelengths and optical periods long, the dominant contribution to the three $\mathbf{k}$ integrals come from those wave vectors that are close to satisfying the index-matching conditions, or energy- and momentum-conserving conditions,

$$\mathbf{k}_0-\mathbf{k}-\mathbf{k}'\approx\mathbf{0}\approx\mathbf{k}_0-\mathbf{k}-\mathbf{k}'' , \tag{22a}$$

$$\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')\approx 0\approx\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}'') . \tag{22b}$$

It follows that $\mathbf{k}'$ and $\mathbf{k}''$ are close to each other, and the difference vector

$$\mathbf{k}''-\mathbf{k}'\equiv\mathbf{k}''' \tag{23}$$

is small and of order $1/l$ even if the pass band Δk_1 of the detector is wide. The scalar product $\boldsymbol{\varepsilon}^*_{\mathbf{k}'s'}\cdot\boldsymbol{\varepsilon}_{\mathbf{k}''s''}$ in Eq. (21) can therefore be well approximated by $\boldsymbol{\varepsilon}^*_{\mathbf{k}'s'}\cdot\boldsymbol{\varepsilon}_{\mathbf{k}'s''}=\delta_{s's''}$, and for the same reason $T_{uv}(\mathbf{k},s;\mathbf{k}'',s'')$ can be replaced by $T_{uv}(\mathbf{k},s;\mathbf{k}',s')$, and $\chi_{uvw}(\omega(\mathbf{k}_0),\omega(\mathbf{k}),\omega(\mathbf{k}''))$ by $\chi_{uvw}(\omega(\mathbf{k}_0),\omega(\mathbf{k}),\omega(\mathbf{k}'))$. In the same spirit we may use a Taylor expansion to express $\omega(\mathbf{k}'')$ in terms of $\omega(\mathbf{k}')$, by writing

$$\begin{aligned}\omega(\mathbf{k}'')&=\omega(\mathbf{k}')+\mathbf{k}'''\cdot\nabla_{\mathbf{k}'}\omega(\mathbf{k}')+\cdots\\&=\omega(\mathbf{k}')+\mathbf{k}'''\cdot\mathbf{u}(\mathbf{k}') ,\end{aligned} \tag{24}$$

where $\mathbf{u}(\mathbf{k})\equiv\nabla_{\mathbf{k}}\omega(\mathbf{k})$ is the group velocity. These transformations allow us to simplify Eq. (21), after making $\mathbf{k}'''$ the new variable of integration in place of $\mathbf{k}''$. The $\mathbf{k}'''$ integration is carried out most easily, if we reexpress all the sinc factors as space-time integrals, by putting

$$\begin{aligned}\mathscr{F}\equiv&\frac{1}{(2\pi)^3}\int d^3k'''\, e^{i[\mathbf{k}'''\cdot(\mathbf{r}_1-\mathbf{r}_0)-\mathbf{k}'''\cdot\mathbf{u}(\mathbf{k}')t]}e^{(i/2)[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')]\Delta t}e^{-(i/2)[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')-\mathbf{k}'''\cdot\mathbf{u}(\mathbf{k}')]\Delta t}\\
&\times\frac{\sin\{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')]\Delta t\}}{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')]}\frac{\sin\{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')-\mathbf{k}'''\cdot\mathbf{u}(\mathbf{k}')]\Delta t\}}{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')-\mathbf{k}'''\cdot\mathbf{u}(\mathbf{k}')]}\\
&\times\prod_{m=1}^{3}\frac{\sin[\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')_m l_m]}{\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')_m}\prod_{m=1}^{3}\frac{\sin[\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}'-\mathbf{k}''')_m l_m]}{\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}'-\mathbf{k}''')_m}\\
=&\frac{1}{(2\pi)^3}\int d^3k'''\int_{\substack{-l_1/2\le x'\le l_1/2\\-l_2/2\le y'\le l_2/2\\-l_3/2\le z'\le l_3/2}}d^3r'\int_{\substack{-l_1/2\le x''\le l_1/2\\-l_2/2\le y''\le l_2/2\\-l_3/2\le z''\le l_3/2}}d^3r''\int_0^{\Delta t}dt'\int_0^{\Delta t}dt''\, e^{i(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')\cdot(\mathbf{r}'+\mathbf{r}'')}\\
&\times e^{i[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')](t'-t'')}e^{i\mathbf{k}'''\cdot[\mathbf{r}_1-\mathbf{r}_0-\mathbf{r}''-\mathbf{u}(\mathbf{k}')(t-t'')]}\\
=&\int_{\substack{-l_1/2\le x'\le l_1/2\\-l_2/2\le y'\le l_2/2\\-l_3/2\le z'\le l_3/2}}d^3r'\int_{\substack{-l_1/2\le x''\le l_1/2\\-l_2/2\le y''\le l_2/2\\-l_3/2\le z''\le l_3/2}}d^3r''\int_0^{\Delta t}dt'\int_0^{\Delta t}dt''\,\delta^3(\mathbf{r}_1-\mathbf{r}_0-\mathbf{r}''-\mathbf{u}(\mathbf{k}')(t-t''))\\
&\times e^{i(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')\cdot(\mathbf{r}'+\mathbf{r}'')}e^{i[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')](t'-t'')} .\end{aligned}$$

If dispersive effects are small, we may take the group velocity vector $\mathbf{u}(\mathbf{k})$ to be almost independent of the wave number k, but pointing in the direction of the wave vector $\mathbf{k}$. We shall therefore write $\mathbf{u}(\mathbf{k})=u\boldsymbol{\kappa}$, where $\boldsymbol{\kappa}\equiv\mathbf{k}/k$. The integration over $\mathbf{r}''$ can now be carried out, and we obtain

$$\begin{aligned}\mathscr{F}=&\int_{\substack{-l_1/2\le x'\le l_1/2\\-l_2/2\le y'\le l_2/2\\-l_3/2\le z'\le l_3/2}}d^3r'\int_0^{\Delta t}dt'\int_0^{\Delta t}dt''\, e^{i(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')\cdot[\mathbf{r}'+\mathbf{r}_1-\mathbf{r}_0-\boldsymbol{\kappa}u(t-t'')]}\\
&\times e^{i[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')](t'-t'')}U(\mathbf{r}_1-\mathbf{r}_0-\boldsymbol{\kappa}'u(t-t'')\,|\,l_1,l_2,l_3) ,\end{aligned}$$

where the step function $U(\mathbf{r}\,|\,l_1,l_2,l_3)$ is defined to be unity if $\mathbf{r}$ falls within the volume $\mathscr{V}_c$ of the crystal and zero otherwise,

$$U(\mathbf{r}\,|\,l_2,l_1,l_3)=\begin{cases}1, & \text{if } \mathbf{r}\in\mathscr{V}_c\\ 0, & \text{otherwise} .\end{cases} \tag{25}$$

In the following we shall suppose that the detector is located in the far field of the nonlinear medium, and that $|\mathbf{r}_1-\mathbf{r}_0|$ and ut are both large and almost equal. After performing the $\mathbf{r}'$ and t' integrations we have

$$\begin{aligned}\mathscr{F}=&\int_0^{\Delta t}dt''\, e^{i(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')\cdot[\mathbf{r}_1-\mathbf{r}_0-\boldsymbol{\kappa}'u(t-t'')]}e^{i[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')[(1/2)\Delta t-t'']}\\
&\times U(\mathbf{r}_1-\mathbf{r}_0-\boldsymbol{\kappa}'u(t-t'')\,|\,l_1,l_2,l_3)\prod_{m=1}^{3}\frac{\sin[\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')_m l_m]}{\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')_m}\frac{\sin\{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')]\Delta t\}}{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')]} .\end{aligned} \tag{26}$$

Use of Eq. (26) in Eq. (21) with the foregoing substitutions then leads to the following expression for the photon detection probability at time t:

$$P_1(t)=\frac{1}{\hbar^2}\int_{\delta\mathscr{V}_1} d^3x_1 \frac{1}{(2\pi)^6}\int d^3k \int_{[\mathbf{k}']_1} d^3k'$$
$$\times\sum_{s,s'}\chi^*_{ijl}(\omega(\mathbf{k}_0),\omega(\mathbf{k}),\omega(\mathbf{k}'))\chi_{uvw}(\omega(\mathbf{k}_0),\omega(\mathbf{k}),\omega(\mathbf{k}'))T_{ij}(\mathbf{k},s;k',s')T^*_{uv}(\mathbf{k},s;\mathbf{k}',s')V^*_l V_w$$
$$\times\int_0^{\Delta t} dt''\, e^{i(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')\cdot[\mathbf{r}_1-\mathbf{r}_0-\boldsymbol{\kappa}'u(t-t'')]}e^{i[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')][(1/2)\Delta t-t'']}U(\mathbf{r}_1-\mathbf{r}_0-\boldsymbol{\kappa}'u(t-t'')\,|\,l_1,l_2,l_3)$$
$$\times\prod_{m=1}^{3}\frac{\sin[\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')_m l_m]}{\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}')_m}\frac{\sin\{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')]\Delta t\}}{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}')]}\ . \tag{27}$$

In order to evaluate the $\mathbf{k}$ integrals one would of course need to know the form of the susceptibility tensor. We compare $P_1(t)$ with a certain probability for the detection of two photons below.

IV. TWO-PHOTON DETECTION PROBABILITY

Next we consider the joint probability $P_2(t,t+\tau)$ of detecting two photons with two detectors located in the far field of the crystal, in the neighborhood of points $\mathbf{r}_1$ and $\mathbf{r}_2$ at two different times t within δt and $t+\tau$ within δt. We therefore need to calculate the expectation of the normally ordered and time ordered operator product

$$\hat{W}^\dagger_p(\mathbf{r}_1,t)\hat{W}^\dagger_q(\mathbf{r}_2,t+\tau)\hat{W}_q(\mathbf{r}_2,t+\tau)\hat{W}_p(\mathbf{r}_1,t)\ ,$$

and to integrate this over the small surfaces of the two detectors and over the short measurement time intervals δt. As before, we replace the surface-time integral by an integral over the equivalent volume $\delta\mathscr{V}$. Let us suppose that the detector at $\mathbf{r}_1$ responds to the set of wave vectors $[\mathbf{k}]_1$ and the detector at $\mathbf{r}_2$ to the set $[\mathbf{k}]_2$, where $[\mathbf{k}]_1$ and $[\mathbf{k}]_2$ in general differ both in direction and frequency. $[\mathbf{k}]_1$ is centered on $\mathbf{k}_1$ and $[\mathbf{k}]_2$ on $\mathbf{k}_2$. Because of the particular form of $\hat{\mathbf{W}}(\mathbf{r},t)$ given by Eq. (14), in which all operators are effectively zero-time operators, the time order of the operator product is not significant, but the normal order is important. We therefore have for the joint probability of two-photon detection at times t and $t+\tau$,

$$P_2(t,t+\tau)=\int_{\delta\mathscr{V}_1} d^3x_1\int_{\delta\mathscr{V}_2} d^3x_2\langle\text{vac}\,|\,\hat{W}^\dagger_p(\mathbf{r}_1,t)\hat{W}^\dagger_q(\mathbf{r}_2,t+\tau)\hat{W}_q(\mathbf{r}_2,t+\tau)\hat{W}_p(\mathbf{r}_1,t)\,|\,\text{vac}\rangle$$
$$=\int_{\delta\mathscr{V}_1} d^3x_1\int_{\delta\mathscr{V}_2} d^3x_2\left\langle\text{vac}\right|\left[\frac{1}{L^{3/2}}\sum_{\mathbf{k},s}\hat{A}_{\mathbf{k}s}(0)f^*_p(\mathbf{k},s;\mathbf{r}_1,t))\right]$$
$$\times\left[\hat{W}^\dagger_{q\,\text{free}}(\mathbf{r}_2,t+\tau)+\frac{1}{L^{3/2}}\sum_{\mathbf{k}',s'}\hat{A}_{\mathbf{k}'s'}(0)f^*_q(\mathbf{k}',s';\mathbf{r}_2,t+\tau)\right]$$
$$\times\left[\hat{W}_{q\,\text{free}}(\mathbf{r}_2,t+\tau)+\frac{1}{L^{3/2}}\sum_{\mathbf{k}'',s''}\hat{A}^\dagger_{\mathbf{k}''s''}(0)f_q(\mathbf{k}'',s'';\mathbf{r}_2,t+\tau)\right]$$
$$\times\left[\frac{1}{L^{3/2}}\sum_{\mathbf{k}''',s'''}\hat{A}^\dagger_{\mathbf{k}'''s'''}(0)f_p(\mathbf{k}''',s''';\mathbf{r}_1,t)\right]\left|\text{vac}\right\rangle\ . \tag{28}$$

We now use the following commutation rule:

$$[\hat{A}_{\mathbf{k}s}(0),\hat{W}^\dagger_{q\,\text{free}}(\mathbf{r}_2,t+\tau)]=\begin{cases}\frac{1}{L^{3/2}}(\boldsymbol{\varepsilon}^*_{\mathbf{k}s})_q\,e^{-i[\mathbf{k}\cdot\mathbf{r}_2-\omega(\mathbf{k})(t+\tau)]} & \text{if } \mathbf{k}\in[\mathbf{k}]_2\\ 0\ \ \text{otherwise}\ ,\end{cases} \tag{29}$$

to rearrange the operator product in Eq. (28) and eventually to put it into normal order. We obtain

$$P_2(t,t+\tau)=\int_{\delta\mathscr{V}_1} d^3x_1\int_{\delta\mathscr{V}_2} d^3x_2\left[\frac{1}{L^6}\sum_{[\mathbf{k}]_2,s}\ \sum_{[\mathbf{k}''']_2,s'''}(\boldsymbol{\varepsilon}^*_{\mathbf{k}s}\cdot\boldsymbol{\varepsilon}_{\mathbf{k}'''s'''})e^{i\{(\mathbf{k}'''-\mathbf{k})\cdot\mathbf{r}_2-[\omega(\mathbf{k}''')-\omega(\mathbf{k})](t+\tau)\}}\mathbf{f}^*(\mathbf{k},s,\mathbf{r}_1,t)\cdot\mathbf{f}(\mathbf{k}''',s''',\mathbf{r}_1,t)\right.$$
$$+\frac{1}{L^6}\sum_{\mathbf{k},s}\sum_{\mathbf{k}',s'}\sum_{\mathbf{k}'',s''}\sum_{\mathbf{k}''',s'''}\langle\hat{A}_{\mathbf{k}s}(0)\hat{A}_{\mathbf{k}'s'}(0)\hat{A}^\dagger_{\mathbf{k}''s''}(0)\hat{A}^\dagger_{\mathbf{k}'''s'''}(0)\rangle$$
$$\left.\times f^*_p(\mathbf{k},s;\mathbf{r}_1,t)f^*_q(\mathbf{k}',s';\mathbf{r}_2,t+\tau)f_q(\mathbf{k}'',s'';\mathbf{r}_2,t+\tau)f_p(\mathbf{k}''',s''';\mathbf{r}_1,t)\right]\ . \tag{30}$$

Because the state of the field is the vacuum state, we have from the commutation relations

$$\langle \hat{A}_{ks}(0)\hat{A}_{\mathbf{k}'s'}(0)\hat{A}^{\dagger}_{\mathbf{k}''s'}(0)\hat{A}^{\dagger}_{\mathbf{k}'''s'''}(0)\rangle = \delta^3_{\mathbf{k}\mathbf{k}''}\delta_{ss''}\delta^3_{\mathbf{k}'\mathbf{k}'''}\delta_{s's'''}+\delta_{\mathbf{k}\mathbf{k}'''}\delta_{ss'''}\delta_{\mathbf{k}'\mathbf{k}''}\delta_{s's''} , \tag{31}$$

and this allows us to rewrite Eq. (30)

$$\begin{aligned} P_2(t,t+\tau) = \int_{\delta\mathscr{V}_1} d^3x_1 \int_{\delta\mathscr{V}_2} d^3x_2 \frac{1}{L^6} \sum_{[\mathbf{k}]_2,s}\sum_{[\mathbf{k}']_2,s'} [(\boldsymbol{\varepsilon}^*_{\mathbf{k}s}\cdot\boldsymbol{\varepsilon}_{\mathbf{k}'s'})e^{i\{(\mathbf{k}'-\mathbf{k})\cdot\mathbf{r}_2-[\omega(\mathbf{k}')-\omega(\mathbf{k})](t+\tau)\}}\mathbf{f}^*(\mathbf{k},s;\mathbf{r}_1,t)\cdot\mathbf{f}(\mathbf{k}',s';\mathbf{r}_1,t) \\ +f^*_p(\mathbf{k},s;\mathbf{r}_1,t)f^*_q(\mathbf{k}',s';\mathbf{r}_2,t+\tau)f_q(\mathbf{k},s;\mathbf{r}_2,t+\tau)f_p(\mathbf{k}',s';\mathbf{r}_1,t) \\ +|\mathbf{f}(\mathbf{k},s;\mathbf{r}_1,t)|^2|\mathbf{f}(\mathbf{k}',s';\mathbf{r}_2,t+\tau)|^2] . \end{aligned} \tag{32}$$

Our previous assumption that the interaction time Δt, or the propagation time through the nonlinear medium is short, and that the down-converted light is of low intensity, makes the last two terms, which are of the fourth order in $\mathbf{f}$, much smaller than the first term, which is of the second order in $\mathbf{f}$. Accordingly we now discared these smaller terms. After replacing sums by integrals in the usual manner, and using Eq. (15) to substitute for $\mathbf{f}(\mathbf{k},s;\mathbf{r},t)$, we obtain

$$\begin{aligned} P_2(t,t+\tau) = \frac{1}{\hbar^2}\int_{\delta\mathscr{V}_1} d^3x_1 \int_{\delta\mathscr{V}_2} d^3x_2 \frac{1}{(2\pi)^{12}} \int_{[\mathbf{k}]_2} d^3k \int_{[\mathbf{k}']_2} d^3k' \int_{[\mathbf{k}'']_1} d^3k'' \int_{[\mathbf{k}''']_1} d^3k''' \\ \times \sum_{s,s',s'',s'''} (\boldsymbol{\varepsilon}^*_{\mathbf{k}s}\cdot\boldsymbol{\varepsilon}_{\mathbf{k}'s'})(\boldsymbol{\varepsilon}^*_{\mathbf{k}''s''}\cdot\boldsymbol{\varepsilon}_{\mathbf{k}'''s'''})\chi^*_{ijl}(\omega(\mathbf{k}_0),\omega(\mathbf{k}''),\omega(\mathbf{k}))\chi_{uvw}(\omega(\mathbf{k}_0),\omega(\mathbf{k}'''),\omega(\mathbf{k}')) \\ \times T_{ij}(\mathbf{k},s;\mathbf{k}'',s'')T^*_{uv}(\mathbf{k}',s';\mathbf{k}''',s''')V^*_l V_w e^{i[(\mathbf{k}'-\mathbf{k})\cdot(\mathbf{r}_2-\mathbf{r}_0)+(\mathbf{k}'''-\mathbf{k}'')\cdot(\mathbf{r}_1-\mathbf{r}_0)]}e^{i[\omega(\mathbf{k}'')-\omega(\mathbf{k}''')-\omega(\mathbf{k}')+\omega(\mathbf{k})]t}e^{-i[\omega(\mathbf{k}')-\omega(\mathbf{k})]\tau} \\ \times e^{(i/2)[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}'')]\Delta t}e^{-(i/2)[\omega(\mathbf{k}_0)-\omega(\mathbf{k}')-\omega(\mathbf{k}''')]\Delta t}\frac{\sin\{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}'')]\Delta t\}}{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}'')]} \\ \times\frac{\sin\{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k}')-\omega(\mathbf{k}''')]\Delta t\}}{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k}')-\omega(k''')]}\prod_{m=1}^{3}\frac{\sin[\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}'')_m l_m]}{\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}'')_m}\prod_{m=1}^{3}\frac{\sin[\frac{1}{2}(\mathbf{k}_0-\mathbf{k}'-\mathbf{k}''')_m l_m]}{\frac{1}{2}(\mathbf{k}_0-\mathbf{k}'-\mathbf{k}''')_m} . \end{aligned} \tag{33}$$

Once again we note that, because of the various sinc factors, the dominant contributions to the four $\mathbf{k}$ integrals come from those wave vectors $\mathbf{k}$, $\mathbf{k}'$, $\mathbf{k}''$, and $\mathbf{k}'''$ that are close to satisfying the index-matching or energy- and momentum-conserving conditions

$$\mathbf{k}+\mathbf{k}''\approx\mathbf{k}_0\approx\mathbf{k}'+\mathbf{k}'' , \tag{34a}$$

$$\omega(\mathbf{k})+\omega(\mathbf{k}'')\approx\omega(\mathbf{k}_0)\approx\omega(\mathbf{k}')+\omega(\mathbf{k}''') . \tag{34b}$$

Now let us suppose that the two detectors 1 and 2 are so arranged that the wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ to which they respond, and on which the sets $[\mathbf{k}'']_1$,$[\mathbf{k}''']_1$ and $[\mathbf{k}]_2$,$[\mathbf{k}']_2$ are centered, are conjugate, in the sense that $\mathbf{k}_1$ and $\mathbf{k}_2$ satisfy the index-matching conditions

$$\mathbf{k}_1+\mathbf{k}_2=\mathbf{k}_0 , \tag{35a}$$

$$\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)=\omega(\mathbf{k}_0) . \tag{35b}$$

Then $\mathbf{k}$,$\mathbf{k}'$ under the integral in Eq. (33) differ from $\mathbf{k}_2$ only by terms of order of the response band of detector 2, and $\mathbf{k}''$,$\mathbf{k}'''$ differ from $\mathbf{k}_1$ only by terms of the same order. We may therefore replace $\mathbf{k}'$ by $\mathbf{k}$ and $\mathbf{k}'''$ by $\mathbf{k}''$ in those factors in Eq. (33) like $\boldsymbol{\varepsilon}^*_{\mathbf{k}s}\cdot\boldsymbol{\varepsilon}_{\mathbf{k}'s'}$, χ_{uvw}, and T^*_{uv}, that vary only slowly with $\mathbf{k}'$,$\mathbf{k}'''$. In the remaining factors we make the substitution

$$\mathbf{k}'-\mathbf{k}\equiv\mathbf{q}_2 , \tag{36a}$$

$$\mathbf{k}'''-\mathbf{k}''\equiv\mathbf{q}_1 , \tag{36b}$$

where q_1,q_2 are small compared with any optical wave number, and we write

$$\omega(\mathbf{k}')\approx\omega(\mathbf{k})+\mathbf{q}_2\cdot\mathbf{u}(\mathbf{k}) , \tag{37a}$$

$$\omega(\mathbf{k}''')\approx\omega(\mathbf{k}'')+\mathbf{q}_1\cdot\mathbf{u}(\mathbf{k}'') , \tag{37b}$$

in terms of the group velocities $\mathbf{u}(\mathbf{k})$ and $\mathbf{u}(\mathbf{k}'')$, as before.

After reexpressing the sinc factors as space and time integrals as in Eq. (25) above, we obtain from Eq. (33),

$$\begin{aligned} P_2(t,t+\tau) = \frac{1}{\hbar^2}\int_{\delta\mathscr{V}_1} d^3x_1 \int_{\delta\mathscr{V}_2} d^3x_2 \frac{1}{(2\pi)^{12}} \int_{[\mathbf{k}]_2} d^3k \int_{[\mathbf{k}'']_1} d^3k'' \int d^3q_1 \int d^3q_2 \\ \times\sum_{s,s''}\chi^*_{ijl}(\omega(\mathbf{k}_0),\omega(\mathbf{k}),\omega(\mathbf{k}''))\chi_{uvw}(\omega(\mathbf{k}_0),\omega(\mathbf{k}),\omega(\mathbf{k}''))T_{ij}(\mathbf{k},s;\mathbf{k}'',s'')T^*_{uv}(\mathbf{k},s;\mathbf{k}'',s'')V^*_l V_w \\ \times\int_{\substack{-l_1/2\le x'\le l_1/2\\ -l_2/2\le y'\le l_2/2\\ -l_3/3\le z'\le l_3/2}} d^3r' \int_{\substack{-l_1/2\le x''\le l_1/2\\ -l_2/2\le y''\le l_2/2\\ -l_3/2\le z''\le l_3/2}} d^3r'' \int_0^{\Delta t}dt' \int_0^{\Delta t}dt'' e^{i\mathbf{q}_1\cdot[\mathbf{r}_1-\mathbf{r}_0-\mathbf{r}''-\mathbf{u}(\mathbf{k}'')(t-t'')]} \\ \times e^{i\mathbf{q}_2\cdot[\mathbf{r}_2-\mathbf{r}_0-\mathbf{r}''-\mathbf{u}(\mathbf{k})(t+\tau-t'')]}e^{i[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}'')](t'-t'')}e^{i(\mathbf{k}_0-\mathbf{k}-\mathbf{k}'')\cdot(\mathbf{r}'+\mathbf{r}'')} . \end{aligned} \tag{38}$$

The integrations over $\mathbf{q}_1$ and $\mathbf{q}_2$ would yield $\delta^{(3)}(\mathbf{r}_1-\mathbf{r}_0-\mathbf{r}''-\mathbf{u}(\mathbf{k}'')(t-t''))$ and $\delta^3(\mathbf{r}_2-\mathbf{r}_0-\mathbf{r}''-\mathbf{u}(\mathbf{k})(t+\tau-t''))$, respectively, if the ranges of $\mathbf{q}_1,\mathbf{q}_2$ were infinite. Actually the range of $\mathbf{q}_m$ is only of order Δk_m $(m=1,2)$, because of the limited ranges of $\mathbf{k}$, $\mathbf{k}'$, $\mathbf{k}''$, and $\mathbf{k}'''$. Nevertheless, the integration may be treated as effectively yielding a δ function, if, for almost all values of the arguments,

$$|\,\mathbf{r}_1-\mathbf{r}_0-\mathbf{r}''-\mathbf{u}(\mathbf{k}'')(t-t'')\,| \gg 1/\Delta k_1 ,$$

$$|\,\mathbf{r}_2-\mathbf{r}_0-\mathbf{r}''-\mathbf{u}(\mathbf{k})(t+\tau-t'')\,| \gg 1/\Delta k_2 ,$$

and if the remainder of the integrand behaves as a slowly varying test function with respect to $\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}'',\mathbf{k}'',t''$ when $\mathbf{r}_1$, $\mathbf{r}_2$, $\mathbf{r}''$, $\mathbf{u}(\mathbf{k}'')t''$, and $\mathbf{u}(\mathbf{k})t''$ vary by an amount of order $1/\Delta k$. These conditions will be satisfied if the passband Δk is large enough, as it usually is in practice. We are then justified in writing δ functions for the $\mathbf{q}_1$ and $\mathbf{q}_2$ integrals. Also we have the general δ-function product relation

$$\int_{\substack{-l_1/2\le x''\le l_1/2\\ -l_2/2\le y''\le l_2/2\\ -l_3/2\le z''\le l_3/2}} d^3r''\, g(\mathbf{r}'')\delta^{(3)}(\mathbf{r}''-\mathbf{a})\delta^{(3)}(\mathbf{r}''-\mathbf{b})=\delta^{(3)}(\mathbf{a}-\mathbf{b})g(\mathbf{a})U(\mathbf{a}\,|\,l_1,l_2,l_3) , \tag{39}$$

where $U(\mathbf{a}\,|\,l_1,l_2,l_3)$ is the unit step function defined by Eq. (25). When this is applied to Eq. (38) we obtain

$$\begin{aligned} P_2(t,t+\tau)=&\frac{1}{\hbar^2}\int_{\delta\mathscr{V}_1} d^3x_1 \int_{\delta\mathscr{V}_2} d^3x_2 \frac{1}{(2\pi)^6}\int_{[\mathbf{k}]_2} d^3k \int_{[\mathbf{k}'']_1} d^3k'' \\ &\times\sum_{s,s''}\chi^*_{ijl}(\omega(\mathbf{k}_0),\omega(\mathbf{k}),\omega(\mathbf{k}''))\chi_{uvw}(\omega(\mathbf{k}_0),\omega(\mathbf{k}),\omega(\mathbf{k}''))T_{ij}(\mathbf{k},s;\mathbf{k}'',s'')T^*_{uv}(\mathbf{k},s;\mathbf{k}'',s'')V^*_l V_w \\ &\times\int_0^{\Delta t} dt' \int_0^{\Delta t} dt'' \int_{\substack{-l_1/2\le x'\le l_1/2\\ -l_2/2\le y'\le l_2/2\\ -l_3/2\le z'\le l_3/2}} d^3r'\, \delta^{(3)}(\mathbf{r}_2-\mathbf{r}_1-\mathbf{u}(\mathbf{k})(t+\tau-t'')+\mathbf{u}(\mathbf{k}'')(t-t'')) \\ &\times U(\mathbf{r}_1-\mathbf{r}_0-\mathbf{u}(\mathbf{k}'')(t-t'')\,|\,l_1,l_2,l_3)e^{i[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}'')](t'-t'')}e^{i(\mathbf{k}_0-\mathbf{k}-\mathbf{k}'')\cdot[\mathbf{r}_1-\mathbf{r}_0+\mathbf{r}'-\mathbf{u}(\mathbf{k}'')(t-t'')]} . \end{aligned} \tag{40}$$

The three-dimensional $\mathbf{r}'$ integration and the t' integration yield a product of four sine functions, which were the basis for the approximate relations (34) above. Recalling that $\mathbf{u}(\mathbf{k})=\boldsymbol{\kappa}u$ and $\mathbf{u}(\mathbf{k}'')=\boldsymbol{\kappa}''u$, let us suppose that the locations of the two detectors and the time t are so chosen that $\mathbf{r}_{20}-\mathbf{r}_0=\boldsymbol{\kappa}_2ut$, $\mathbf{r}_{10}-\mathbf{r}_0=\boldsymbol{\kappa}_1ut$, where $\mathbf{k}_1,\mathbf{k}_2$ satisfy the index-matching conditions (35), and $\mathbf{r}_{10},\mathbf{r}_{20}$ are the midpoints of the volumes $\delta\mathscr{V}_1,\delta\mathscr{V}_2$. In other words, the detectors are placed so as to pick out conjugate photon pairs from the signal and idler. The integration of the δ function over $\mathbf{r}_2$ will yield unity only if the volume of integration $\delta\mathscr{V}_2$ is large enough to accommodate all possible values of the vector

$$\mathbf{r}_1+\mathbf{u}(\mathbf{k})(t+\tau-t'')-\mathbf{u}(\mathbf{k}'')(t-t'') .$$

If $\mathbf{r}_1-\mathbf{r}_{10}\equiv\delta\mathbf{r}_1$, $\mathbf{r}_2-\mathbf{r}_{20}\equiv\delta\mathbf{r}_2$, the necessary condition can be written

$$\delta\mathbf{r}_2-\delta\mathbf{r}_1-(\boldsymbol{\kappa}-\boldsymbol{\kappa}_2)ut+(\boldsymbol{\kappa}''-\boldsymbol{\kappa}_1)ut+(\boldsymbol{\kappa}-\boldsymbol{\kappa}'')ut''-\boldsymbol{\kappa}u\tau=0. \tag{41}$$

Now $|\,\boldsymbol{\kappa}-\boldsymbol{\kappa}_2\,|\lesssim\frac{1}{2}\Delta\theta_2$, $|\,\boldsymbol{\kappa}''-\boldsymbol{\kappa}_1\,|\lesssim\frac{1}{2}\Delta\theta_1$, where $\Delta\theta_1$ and $\Delta\theta_2$ are the angular ranges of the wave vectors to which detectors 2 and 1 respond, and $|\,\boldsymbol{\kappa}-\boldsymbol{\kappa}''\,|\approx\theta$, the angle between the conjugate $\mathbf{k}_1$ and $\mathbf{k}_2$ vectors. Hence

$$\begin{aligned} &|\,(\boldsymbol{\kappa}''-\boldsymbol{\kappa}_1)ut\,|\lesssim\tfrac{1}{2}\Delta\theta_1\,ut , \\ &|\,(\boldsymbol{\kappa}-\boldsymbol{\kappa}_2)ut\,|\lesssim\tfrac{1}{2}\Delta\theta_2\,ut , \\ &|\,(\boldsymbol{\kappa}-\boldsymbol{\kappa}'')ut''\,|\lesssim\theta u\,\Delta t=\theta l_3 . \end{aligned} \tag{42}$$

Because of the finite size of the nonlinear medium, for a given signal wave vector $\mathbf{k}''$ there is a whole range of conjugate idler wave vectors $\mathbf{k}$ lying within an angle $\delta\theta_2\sim 1/k_2l_1$. If Eq. (41) is to hold, and if detector 1 has an aperture of linear dimensions $\Delta\theta_1\,ut$ in the plane containing the vectors $\mathbf{k}_1,\mathbf{k}_2$, then detector 2 needs an aperture of size $(\Delta\theta_1+\delta\theta_2)ut+\theta l_3$ in the same plane.

The geometric significance of this requirement can be understood by reference to Fig. 1. If a signal wave making an angle θ_1 with the pump beam is conjugate to an idler wave making an angle $-\theta_2$ (with $\theta_1\sim\theta_2$), then signal waves within angles $\theta_1\pm\frac{1}{2}(\Delta\theta_1+\theta_1l_3/ut)$ are conjugate to idler waves within $\theta_2\pm\frac{1}{2}(\delta\theta_2+\Delta\theta_1+\theta_1l_3/ut)$. Because of the crystal length l_3 these waves at distance ut spread to cover a transverse width

$$(\delta\theta_2+\Delta\theta_1+\theta_1l_3/ut)ut+\theta_2l_3=(\Delta\theta_1+\delta\theta_2)ut+\theta l_3 , \tag{43}$$

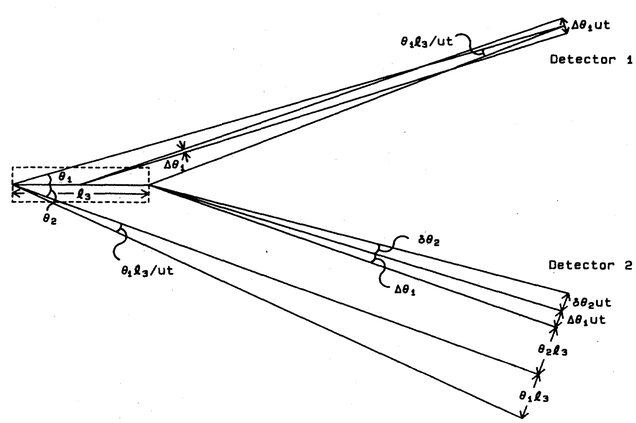

FIG. 1. The geometry of the two-photon detection experiment.

where $\theta=\theta_1+\theta_2$. The condition $\delta x_2 \gtrsim (\Delta\theta_1+\delta\theta_2)ut+\theta l_3$ is therefore necessary to ensure that detector 2 responds to all wave vectors which are conjugate to wave vectors to which detector 1 responds. In addition, for Eq. (41) to be satisfied it is necessary that the longitudinal range of $\delta \mathbf{r}_2$ is no less than $\delta \mathbf{r}_1-\boldsymbol{\kappa} u\tau$, or the resolving time interval δt_2 of detector 2 is no less than $\delta t_1+\tau$. When $\tau=0$ this merely requires δt_2 to be no less than δt_1. If these conditions are satisfied, then we obtain from Eq. (40) after integration over $\mathbf{r}'$, t', and $\mathbf{r}_2$, with $\tau=0$,

$$P_2(t,t)=\frac{1}{\hbar^2}\int_{\delta\mathscr{V}_1}d^3x_1\frac{1}{(2\pi)^6}\int_{[\mathbf{k}]_2}d^3k\int_{[\mathbf{k}'']_1}d^3k''\sum_{s,s''}\chi^*_{ijl}(\omega(\mathbf{k}_0),\omega(\mathbf{k}),\omega(\mathbf{k}''))$$
$$\times\chi_{uvw}(\omega(\mathbf{k}_0),\omega(\mathbf{k}),\omega(\mathbf{k}''))T_{ij}(\mathbf{k},s;\mathbf{k}'',s'')T^*_{uv}(\mathbf{k},s;\mathbf{k}'',s'')V^*_l V_w$$
$$\times\int_0^{\Delta t}dt''\,e^{i(\mathbf{k}_0-\mathbf{k}-\mathbf{k}'')\cdot[\mathbf{r}_1-\mathbf{r}_0-\boldsymbol{\kappa}''u(t-t'')]}e^{i[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}'')][(1/2)\Delta t-t'']}U(\mathbf{r}_1-\mathbf{r}_0-\boldsymbol{\kappa}''u(t-t'')\,|\,l_1,l_2,l_3)$$
$$\times\prod_{m=1}^{3}\frac{\sin[\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}'')_m l_m]}{\frac{1}{2}(\mathbf{k}_0-\mathbf{k}-\mathbf{k}'')_m}\frac{\sin\{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}'')]\Delta t\}}{\frac{1}{2}[\omega(\mathbf{k}_0)-\omega(\mathbf{k})-\omega(\mathbf{k}'')]}\,. \tag{44}$$

It is interesting to compare $P_2(t,t)$ given above with the one-photon detection probability $P_1(t)$ given by Eq. (27). Although the $\mathbf{k}$ integration in Eq. (27) appears to be unrestricted in range, whereas the $\mathbf{k}$ integration in Eq. (44) is limited to the set $[\mathbf{k}]_2$, i.e., to a small range Δk about the vector $\mathbf{k}_2$ that characterizes the photodetector 2, the distinction is really illusory. For, because of the sinc factors, the vectors $\mathbf{k},\mathbf{k}'$ in Eq. (27) are connected so that

$$\mathbf{k}=\mathbf{k}_0-\mathbf{k}'+O\left[\frac{1}{l}\right], \tag{45a}$$

$$\omega(\mathbf{k})=\omega(\mathbf{k}_0)-\omega(\mathbf{k}')+O\left[\frac{1}{\Delta t}\right]. \tag{45b}$$

If the passband Δk is as large as or larger than $1/l$, then $\mathbf{k}$ differs from $\mathbf{k}_0-\mathbf{k}_1$ by a vector whose length is no greater than Δk, and $\omega(\mathbf{k})$ differs from $\omega(\mathbf{k}_0)-\omega(\mathbf{k}_1)$ only by a term of order $\Delta k/u$. The integral over $\mathbf{k}$ therefore has an effective range Δk which is centered on $\mathbf{k}_2\equiv\mathbf{k}_0-\mathbf{k}_1$, and we could have imposed the restriction $[\mathbf{k}]_2$ on the $\mathbf{k}$ integral in Eq. (27) With this substitution we have

$$P_2(t,t)=P_1(t)\,, \tag{46}$$

or the two-photon detection probability is exactly equal to the one-photon probability. In other words, every signal photon emitted in one direction in the process of down conversion is accompanied by an idler photon emitted in the conjugate direction, with the relevant two wave vectors connected by the index-matching conditions (35). The frequencies of the two conjugate photons are determined partly by the two directions of propagation, and they may differ substantially. The effective equality of $P_1(t)$ and $P_2(t,t)$ has been confirmed in photon counting experiments,[19,20] when due allowance is made for the quantum and collection efficiencies of the detectors.

V. TWO-TIME CORRELATIONS

In connection with the derivation of Eq. (41) and the subsequent discussion we have already seen that $P_2(t,t+\tau)$ begins to fall from its maximum value once τ exceeds the resolution limit δt of detectors 1 and 2. This suggests that the two-photon correlation time T_c may be governed by practical considerations involving the photodetectors, rather than by fundamental properties of the pumping light or the nonlinear medium.

For another approach to the variation of $P_2(t,t+\tau)$ with τ we return to Eq. (38), and we observe that the τ dependence is carried entirely by the factor

$$e^{i\mathbf{q}_2\cdot\mathbf{u}(\mathbf{k})\tau}=e^{i\mathbf{q}_2\cdot\boldsymbol{\kappa}u\tau}$$

under the integral. As we have taken u to be virtually constant with frequency, the correlation time is governed by the effective range of $\mathbf{q}_2\cdot\boldsymbol{\kappa}$ under the integral. Now $\mathbf{q}_2\equiv\mathbf{k}-\mathbf{k}'$, and from Eq. (33) $\mathbf{k}$ and $\mathbf{k}'$ both belong to the set $[\mathbf{k}]_2$ and therefore differ only by Δk. Hence the range of $P_2(t,t+\tau)$, or the two-photon correlation time T_c, has a lower limit of order $1/u\Delta k=1/\Delta\omega$, where $\Delta\omega$ is the bandwidth of the detector field. So long as the resolving time δt of the detectors exceeds $1/\Delta\omega$, T_c is governed by δt. But if δt falls below $1/\Delta\omega$ then $1/\Delta\omega$ provides the range on the time interval within which the signal and idler photons can be detected. We see that it is really an instrumental limit, rather than one determined by the nature of the incident pumping light or of the nonlinear medium.

VI. DISCUSSION

We have shown that when the positions and apertures of the two photodetectors are properly chosen, so that the detectors capture the conjugate signal and idler photons, then the joint two-photon detection probability equals the one-photon detection probability, provided that the quantum detection efficiencies are both 100%. If the detection efficiencies α_1,α_2 are less than 100%, then $P_1(t)$ has to be multiplied by α_1, and $P_2(t,t+\tau)$ by $\alpha_1\alpha_2$, so that

$$P_2(t,t+\tau)=\alpha_2 P_1(t)\,. \tag{47}$$

This has been confirmed in the original,[19] and also in the more recent, experiments,[20] and is consistent with the idea that the signal and idler photons are always created together from a single-pump photon. It suggests the possibility of using the two down-converted photons in order to discriminate against background in an optical communication channel.[24]

The correlation time T_c within which the two down-converted photons show up is obviously an important quantity. We have seen that T_c has nothing to do with the bandwidth of the incident pump field, which was actually taken to be zero in our calculation. While the pump bandwidth determines the time uncertainty of an incoming (pump) photon, it does not affect the relative time separation between the signal and idler photons. T_c is ultimately limited by the acceptance bandwidth of the down-converted light, which is generally governed by practical considerations, but can be exceedingly short. In particular, T_c can be shorter than the propagation time Δt through the nonlinear medium. This conclusion is also consistent with recent measurements, in which T_c was found to be below 150 psec, and limited by the transit time spread of the photodetector, when the propagation time Δt was close to 400 psec.[20] However, there appears to be no fundamental reason why T_c cannot be in the subpicosecond range. The process of parametric down conversion therefore provides us with a highly correlated photon pair with extremely small time separation.

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