Ramsey interference in two-photon parametric scattering

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The properties of a two-photon optical field radiated under the action of a coherent pump by two or more flat nonlinear crystals without inversion centers, separated by a linear medium, in particular, a vacuum, are analyzed. It is shown that interference between the spontaneous fields of the crystals leads to the appearance of additional structure in the spectrum of the field, with a scale inversely proportional to the distance l' between the crystals. The probability for emission of two photons at the times t_a and t_a+t has two or more maxima in its dependence on t with the distance between them proportional to l'.

1. INTRODUCTION

The Ramsey method of separated fields is widely used in beam spectroscopes and masers.[1] In this method the region of interaction between the molecule and the resonance field inducing a transition between two levels of the molecule divides spatially into two parts with an interval between them in which the field is absent. As a result, the observed spectrum $g(\omega)$ (ω is the field frequency), which has a total transit-time width of order $1/2\tau$ (τ is the time it takes a molecule to cross the field region), acquires fine interference structure with period $1/\tau'$, where τ' is the time it takes a molecule to cross the gap between the fields, which is much greater than τ.

In the present paper it is shown that an analogous phenomenon takes place in the emission of a photon pair by a macroscopic body of nonlinear material (usually a transparent piezocrystal) according to the scheme $\mathbf{k}_0 \rightarrow \mathbf{k}_a + \mathbf{k}_b$, where $\mathbf{k}_0$ is the pump wave vector (the spontaneous parametric scattering (SPS) effect; see, for example, Ref. 2). The design of this experiment is shown in Fig. 1. Here the field and the material exchange roles: the roles of the two field regions and the "dark" gap between them are played by two nonlinear layers of thickness $2l$ separated by a linear layer of thickness l'; the role of the molecule is played by the signal ($\mathbf{k}_a$) and idler ($\mathbf{k}_b$) modes of the electromagnetic field; and the role of the transit time of the molecule $\tau(\tau')$ is played by the difference in the time it takes the signal and idler photons to cross the nonlinear (linear) layers, which is determined by the dispersion of the group velocity.

We assume the pump to be classical and prescribed, which makes the problem linear and corresponds to the usual experimental conditions. In the molecular model this approximation corresponds to the case of a weak field. At the same time, our model allows an additional possibility: the parameters τ and/or τ' can be negative, in which case opposite signs on τ and τ' imply cancellation of dispersion in the linear and nonlinear layers.

Below we will consider in more detail (but in a preliminary fashion) the more general problem of n identical nonlinear layers separated by $n-1$ identical linear gaps. Here the width of the central maximum in the SPS spectrum in a fixed direction is narrowed as a result of the gaps by a factor of $1+\tau'/\tau$ (in the case $\tau'/\tau>0$).

Note that the SPS effect in two or more crystals, but with different geometry, was considered in Refs. 3–6, where the possibility was considered of formulating new EPR–Bell-type experiments. Interesting interference effects, arising as a result of mixing of the signal and/or idler beams radiated by two crystals with a common coherent pump, were observed by Mandel *et al.*[7–9]

2. PARAMETRIC SCATTERING SPECTRUM IN A MULTILAYER MEDIUM

The SPS effect presents the unique possibility of preparing an almost pure two-photon field with macroscopic coherence scales and easily varied space-time structure.[2,10,11] In the first approximation in the pump amplitude E_0 the field state vector has the form

$$|\psi\rangle = \left(1+\frac{1}{2}\sum_{\mathbf{kk}'} F_{\mathbf{kk}'} a_{\mathbf{k}}^{+} a_{\mathbf{k}'}^{+}\right)|0\rangle, \tag{1}$$

where $a_{\mathbf{k}}^{+}$ is the photon creation operator in the $\mathbf{k}$ mode, $|0\rangle$ is the vacuum state and the function $F_{\mathbf{kk}'}$ has the meaning of an effective field describing the shape of the two-photon wave packet ("biphoton") in the spectral representation. Its Fourier transform defines the structure of the biphoton in 8-dimensional space-time, which can be measured by the coincidence method with two broadband point detectors.

There are three main ways of controlling the structure of the biphoton: placing various optical elements (filters, diaphragms, beamsplitters, lenses, etc.) in the pump field[11] or the scattered radiation field[2,10] or varying the geometry of the nonlinear scattering region.[3] Here we will consider the last method.

We assume an interaction energy of the form χE^3, where χ is the quadratic susceptibility of the crystal. In the

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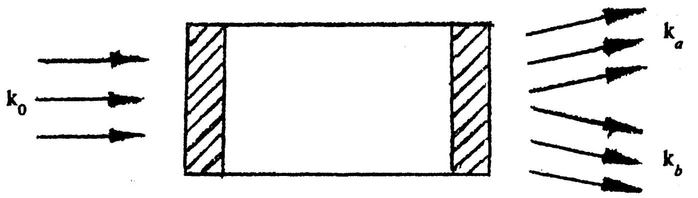

FIG. 1. Scheme for observing Ramsey interference in parametric light scattering: the pump photon $\mathbf{k}_0$ decays into two photons $\mathbf{k}_a$ and $\mathbf{k}_b$ as a result of coherent mixing in the two nonlinear crystals (hatched regions) separated by a linear medium, in particular, air.

diffraction-free approximation the modes of the scattered field are pairwise coupled by the conditions $\omega+\tilde{\omega}=\omega_0$ and $\mathbf{k}_\perp+\tilde{\mathbf{k}}_\perp=0$, as a result of which we have (for more detail, see Ref. 2)

$$F_{\mathbf{k}\mathbf{k}'}=-i\Gamma f_{\mathbf{k}}\delta_{\mathbf{k}'\tilde{\mathbf{k}}}, \tag{2}$$

where

$$\Gamma=2\pi c^{-1}\sqrt{\omega\tilde{\omega}}\chi E_0 L \tag{3}$$

is the parametric gain coefficient in a layer of thickness $L=nl$ and the function

$$f_{\mathbf{k}}=\sum_{m=1}^{n} f_{\mathbf{k}}^{(m)}, \tag{4}$$

$$f_{\mathbf{k}}^{(m)}=\frac{1}{L}\int_{z_m}^{z_m+l} dz D_m^*(\mathbf{k}_0,z)\,D_m(\mathbf{k},z)\,D_m(\tilde{\mathbf{k}},z) \tag{5}$$

describes how dispersion of the refractive index in the linear and nonlinear media affects the efficiency with which the three waves interact. Here $D_m(\mathbf{k},z)$ is the propagation function (reckoned from the plane $z=0$) and

$$z_m=-ml-(m-1)l' \tag{6}$$

is the coordinate of the left boundary of the mth nonlinear layer. According to Eq. (4) the effective field consists of n components, which can interfere as a consequence of the assumed coherence of the pump field over the entire nonlinear region.

If we neglect reflections from the boundaries of the layers, we have

$$D_m(\mathbf{k},z)=\exp[-ik_z z+(m-1)(k_z'-k_z)l'], \tag{7}$$

where k_z and k_z' are the longitudinal components of the wave vectors in the nonlinear and linear media. The boundary conditions take the form $\mathbf{k}_\perp=\mathbf{k}_\perp'$. From Eqs. (5) and (7) we find that

$$f_{\mathbf{k}}^{(m)}=\frac{1-\exp(-i\delta)}{in\delta}\exp[-i(m-1)(\delta+\delta')], \tag{8}$$

where $\delta=\Delta l$, $\delta'=\Delta' l'$,

$$\Delta(\omega,\vartheta)=k_0-k_z-\tilde{k}_z, \tag{9}$$

$$\Delta'(\omega,\vartheta)=k_0'-k_z'-\tilde{k}_z' \tag{10}$$

are the wave detunings, $\omega=\omega(k)$ is the frequency corresponding the wave vector $\mathbf{k}$, and ϑ is the angle between $\mathbf{k}$ and the z axis (the scattering angle). Note that the function $f_k^{(m)}$ should change sign under reversal of the direction of the polar axis in the mth crystal, since then $\chi\to-\chi$.

Substituting the expression (8) in Eq. (4), we find the total effective field radiated by the n layers:

$$f_{\mathbf{k}}=\frac{1-\exp(-i\delta)}{in\delta}\,\frac{\exp[-in(\delta+\delta')]-1}{\exp[-i(\delta+\delta')]-1}$$

$$=\frac{\sin(\delta/2)}{\delta/2}\,\frac{\sin[n(\delta+\delta')/2]}{n\sin[(\delta+\delta')/2]}\exp(-i\alpha), \tag{11}$$

where $2\alpha=n\delta+(n-1)\delta'$. The first factor in the last line of Eq. (11) describes the ordinary SPS spectrum in the lth layer, and the second, the effect of interference between the spontaneous fields radiated by the n crystals.

If ordinary instead of homodyne detectors are used, the scattered field will appear to be stationary, and in photons per second out to one transverse mode, i.e., within a solid angle of λ^2/a^2, where a is the transverse diameter of the radiating region (practically the diameter of the pump beam) in the direction ϑ for small ϑ according to Eqs. (2) and (11) its intensity $\dot{N}(\vartheta)$ will be equal to

$$\dot{N}(\vartheta)=\int_0^{\omega_0} N(\omega,\vartheta)\,\frac{d\omega}{2}\,\pi\equiv\Gamma^2\Delta\nu(\vartheta), \tag{12}$$

where

$$N(\omega,\vartheta)=|F_{\mathbf{k}\tilde{\mathbf{k}}}|^2=\Gamma^2 g(\omega,\vartheta), \tag{13}$$

$$g(\omega,\vartheta)=|f_{\mathbf{k}}|^2=\left\{\frac{\sin(\delta/2)}{\delta/2}\,\frac{\sin[n(\delta+\delta')/2]}{n\sin[(\delta+\delta')/2]}\right\}^2 \tag{14}$$

(here we have neglected the slow dependence of Γ on ω, ϑ, and ϕ). If the condition $g_{\max}=1$ is fulfilled, the parameter $\Delta\nu(\vartheta)$ has the meaning of an effective spectral width (in hertz) for observing in the given direction.

In the absence of gaps ($\delta'=0$), Eq. (14) gives the usual spectrum for the layer $L=nl$

$$g(\omega,\vartheta)=(2/\Delta L)^2\sin^2(\Delta L/2), \tag{15}$$

whose effective width is inversely proportional to L so that $\dot{N}\propto L^2\Delta\nu\propto L$. When the dispersion cancels ($\delta+\delta'=0$) it is necessary to replace L in Eq. (15) by l, resulting in an increase in the spectral width and the total intensity by a factor of n in comparison with a solid crystal of length L.

Let the synchronism condition be fulfilled for some $\mathbf{k}_a$: $\Delta(\omega_a,\vartheta_a)=0$. Frequently a linear expansion of the function $\Delta(\omega,\vartheta)$ in the vicinity of ω_a, ϑ_a suffices:

$$\delta=\Delta l=\tau\Omega+\mu(\vartheta-\vartheta_a), \tag{16}$$

$$\tau=l[u_b^{-1}-u_a^{-1}\cos(\vartheta_a+\vartheta_b)]/\cos\vartheta_b, \tag{17}$$

$$\mu=k_0\mathrm{tg}(\vartheta_b) \tag{18}$$

(we assume the polarization of the scattered field to be ordinary). Here $\Omega=\omega-\omega_a$, $\vartheta_{a,b}$ are the scattering angles within the crystal for the signal ($\mathbf{k}_a$) and induced

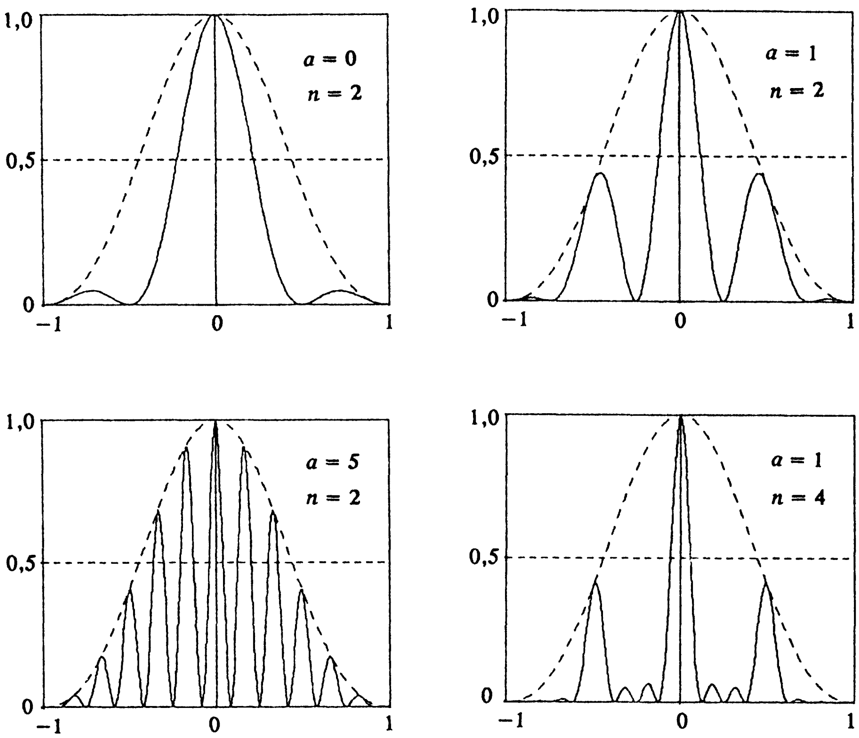

FIG. 2. Spectrum of parametric scattering by n sequentially arranged crystals. Dimensionless frequency $(\omega-\omega_a)\tau/2\pi$ is plotted along the abscissa, the parameter $a=\tau/\tau'$ is proportional to the ratio l'/l of the length of the gap between the crystals to their length. The dashed curve (the envelope of the spectrum) is the spectrum for the case of dispersion cancellation, i.e., for $a=-1$.

$(\mathbf{k}_b=\tilde{\mathbf{k}}_a=\mathbf{k}_0-\mathbf{k}_a)$ waves, and $u_{a,b}$ are the group velocities. The expansion of $\delta'(\omega,\vartheta)$, which, however, contains a constant component $\delta'_a = \Delta'(\mathbf{k}_a)$, has an analogous form. In the case of vacuum gaps and small scattering angles we have $\delta'=0$. Note that for $\vartheta_{a,b}\ll 1$ the parameter τ has the meaning of a difference in the transit times of the induced and signal photons through the nonlinear layer (and analogously for τ').

Let $\vartheta=\vartheta_a$. Then by virtue of relation (16) the spectral function (14) takes the form

$$g(\Omega)=\left\{\frac{\sin(\Omega\tau/2)}{\Omega\tau/2}\,\frac{\sin[n\Omega(\tau+\tau')/2+n\delta'_a/2]}{n\sin[\Omega(\tau+\tau')/2+\delta'_a/2]}\right\}^2. \tag{19}$$

Plots of this function for selected values of the parameters are shown in Fig. 2 for $\delta'_a = 0$. Note that for $\tau+\tau'=0$, $\delta'_a = \pm\pi$, and n even, it follows from Eq. (19) that $g(\Omega)=0$, i.e., the radiation is completely suppressed (the same result follows from Eq. (8) for $\delta+\delta'=\pm\pi$).

3. PARAMETRIC SCATTERING BY TWO LAYERS

Let us consider the case $n=2$ in more detail. According to Eq. (19)

$$g(\Omega)=\left\{\frac{2}{\Omega\tau}\sin\frac{\Omega\tau}{2}\cos\left[\frac{\Omega(\tau+\tau')}{2}+\frac{\delta'_a}{2}\right]\right\}^2. \tag{20}$$

This function describes the observed spectrum in the Ramsey method in the case of a weak field if by Ω we understand the difference between the field frequency and the frequency of the molecular transition, by τ and τ' the transit times of a molecule through the field regions and the gap between them, and by δ'_a the phase difference of the two fields.[1] In essence, expression (20) is the response of a linear oscillator to two pulses with frequency $\omega_0-\Omega$, duration τ, interval τ', and phase difference δ'_a.

Let us find the total intensity $\dot{N}(\vartheta)$, defined according to Eq. (12) by the parameter $\Delta\nu$. From Eq. (20) it follows that if the condition $|1+\tau'/\tau|\geqslant 1$ holds, $\Delta\nu$ does not depend on the presence of the gap, i.e., Ramsey interference only redistributes the energy within the spectrum:

$$\Delta\nu=\int_{-\infty}^{\infty} g(\Omega)\,\frac{d\Omega}{2\pi}=\frac{1}{2|\tau|}\equiv\Delta\nu_0\left(|1+\frac{\tau'}{\tau}|\geqslant 1\right). \tag{21}$$

However, for the reverse inequality Eq. (20) gives

$$\Delta\nu=\Delta\nu_0[1+(1-|1+\tau'/\tau|)\cos\delta'_a](-2\leqslant\tau'/\tau\leqslant 0). \tag{22}$$

Thus, for $\tau+\tau'=0$ (dispersion cancellation) and $\delta'_a = 2m\pi$, $m=0, \pm 1,...,$ the spectral width and the intensity double, and for $\delta'_a = (2m+1)\pi$, they vanish.

Let us now consider another method for detecting the effect—with the help of two sensors and a scheme of delayed coincidences. Let two broadband sensors detect the photons in two conjugate directions $\mathbf{k}_a$ and $\mathbf{k}_b$. We assume the distances to the $z=0$ plane along these directions to be identical. According to Eq. (2), the probability of registering the signal photon at the time t_a and the induced photon at the time $t_b=t_a+t$ is proportional to the Fourier transform of the function $f_{\mathbf{k}}=f(\omega)$ defined by Eq. (11):

$$f(t)=\frac{1}{2\pi}\int_0^{\omega_0} d\omega e^{i\omega t}f(\omega). \tag{23}$$

The function $f(t)$ has the sense of the shape of the two-photon wave packet, which can be conveniently described in terms of a leading wave emitted by one of the detectors

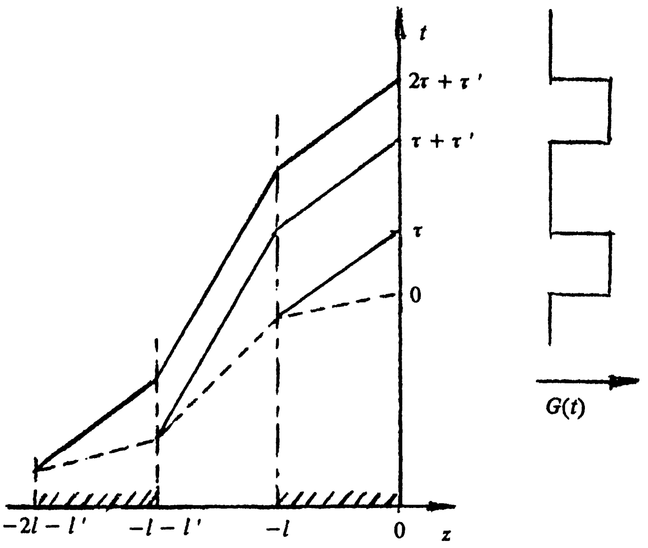

FIG. 3. Scheme for forming a two-pulse photon correlation function $G(t)$: the world line of the signal photon that intersects the $z=0$ plane at the fixed time $t_a=0$ is depicted by the dashed line. Possible world lines of the idler photon, which presumably has a lower velocity, are depicted by the solid lines.

back toward the scattering region, where it is effectively reflected and from which it propagates toward the second detector.[2,3,10,12]

Setting $n=2$ in Eq. (11), extending the limits of integration in expression (23) to infinity, and using expansion (16), we obtain

$$f(t)=\frac{1}{4\pi l}\int_{-l}^{0}dz\int_{-\infty}^{\infty}d\omega\exp(i\Delta z)[1+\exp(-i\delta-i\delta')]$$

$$=\frac{1}{2l}\int_{-l}^{0}dz\Big[\delta(t+\tau z/l)+\exp(-i\delta_a')$$

$$\times\delta\Big(t+\frac{\tau z}{l}-\tau-\tau'\Big)\Big]$$

$$=\frac{1}{2|\tau|}\Big[\Pi\Big(\frac{t}{\tau}\Big)+\exp(-i\delta_a')\Pi\Big(\frac{t-\tau-\tau'}{\tau}\Big)\Big]. \quad (24)$$

Here we have introduced the function $\Pi(x)$, equal to 1 for $0<x<1$ and 0 outside this interval. According to Eq. (24), the effective field $f(t)$, whose square determines the probability of delayed coincidences, consists of two rectangular pulses of duration $|\tau|$, separated for $|1+\tau'/\tau|>1$ by some interval (equal to τ' for $\tau, \tau'>0$). Such a shape of the biphoton is readily explained (Fig. 3), assuming that the photons are emitted in pairs by each cross section of the two nonlinear layers with equal probability and propagate with group velocities $u_{a,b}$ and $u'_{a,b}$ (see Eq. (17)).

For $|1+\tau'/\tau|<1$, the impulses overlap, which handily explains any increase or decrease of the total intensity according to Eq. (22).

The differential probability of delayed coincidences (with dimensions of $\sec^{-2}$) in the case of broadband detectors with 100% efficiency and aperture equal to the coherence area is equal to the correlation function for the photon fluxes:

$$G(t)\equiv\langle n_a(t_a)n_b(t_a+t)\rangle$$

$$=\Gamma^2|f(t)|^2=\Gamma^2\Delta\nu_0^2\Big[\Pi\Big(\frac{t}{\tau}\Big)+\Pi\Big(\frac{t-\tau-\tau'}{\tau}\Big)$$

$$+2\cos\delta_a'\Pi\Big(\frac{t}{\tau}\Big)\Pi\Big(\frac{t-\tau-\tau'}{\tau}\Big)\Big] \quad (25)$$

[here we have used Eqs. (21) and (24)].

If $|1+\tau'/\tau|>1$ holds, then the last term in expression (25) is equal to zero, i.e., the pulses overlap (see Fig. 3). In this case observation in an isolated run of the difference in readout times t belonging, for example, to the interval $[0,\tau]$ allows one to say that the photons were created in the first crystal. If t is registered exactly, it is possible to identify the z coordinate of pair creation (this, of course, is simply the obvious interpretation of the quantum formalism).

For the reverse inequality, the pulses of the effective field (24) overlap either completely or partially. In this case, according to Eq. (25), for $\delta_a'=\pm\pi$ in the overlap region $G(t)=0$, and for $\delta_a'=0$, $G(t)$ is doubled, i.e., beams of radiation from two given segments Δz_1 and Δz_2 of the first and second crystals either suppress or amplify each other [compare Eq. (22)]. Thus the indeterminacy of the region of pair creation leads to a unique sort of interference, exhibited in the way the probability of delayed coincidences depends on the dispersion properties of the gap. Note that these latter properties can be inhomogeneous in z, in which case it is necessary to replace $\delta'=\Delta'l'$ by the corresponding integral phase (compare the influence of a magnetic field inhomogeneity in the Ramsey method[1]).

The scheme of coincidences with resolution time much greater than the coherence times $|\tau|$ and $|\tau'|$ gives a total rate of coincidences per unit time equal to

$$\dot{N}_c=\int_{-\infty}^{\infty}G(t)dt=\Gamma^2\int_{-\infty}^{\infty}g(\Omega)\frac{d\Omega}{2\pi}. \quad (26)$$

Comparing with Eq. (12), we find that $\dot{N}_c=\dot{N}_a$. This equality is in agreement with the obvious picture of photons emitted only in pairs and is used for etalon-free calibration of photodetectors.[13,14]

In conclusion, we note that the present effect, like other two-photon parametric effects (including even the hypothetical two-photon "accelerated" radiation, which should be registered by two detectors accelerated in a vacuum in opposite directions[9]), has a close classical analog, differing only in its additional "background." This follows from a general phenomenological description of linear transformations in quantum optics.[4] To realize the corresponding classical experiment, it is necessary that, in addition to the pump beam, intense signal and induced "starter" radiation with brightness exceeding that of the zero fluctuations of the vacuum fall upon the nonlinear crystals.

It should be possible to use this effect to measure dispersion and to shape the superluminescence spectrum. Note also that a similar kind of interference should also be observed in four-wave parametric mixing (see Ref. 3).

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