Parametric generation of two-photon light in anisotropic layered media

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(Submitted 7 February 1994)
Zh. Eksp. Teor. Fiz. **105**, 1574–1582 (June 1994)

Spontaneous parametric light scattering in a system of plane-parallel linear and nonlinear anisotropic layers is analyzed. It is shown that regular layered systems (superlattices) permit generation of photon pairs (biphotons) with a broad spectrum, i.e., with a high degree of localization of photons belonging to one pair. Scattering in a three-layer nonlinear system provides a way of preparing biphotons with arbitrary polarization given by four parameters.

1. INTRODUCTION

Spontaneous parametric light scattering in transparent birefringent piezocrystals is an efficient method for generating optical radiation consisting of pairs of correlated photons.[1,2] As noted in Ref. 1, this radiation can be used for measuring group delays. However, in direct measurements of photon detection times, the precision of the method is restricted by the time response of existing photodetectors and electron circuits, which is typically ~ 1 ns.

An ingenious method for overcoming this restriction was demonstrated in Ref. 3 (see also Refs. 4–7). As a result, the limiting measurement accuracy is determined by the scatter of the emission times of photons belonging to one pair, i.e., the second-order coherence time (Ref. 8). For a monochromatic pump wave in the absence of additional filtering, this time is determined by dispersion of the refractive index of the piezocrystal and its thickness l. When a linear approximation is used for the dispersion law $\omega(k)$, the distribution function $f(\tau)$ for the difference $\tau=t_1-t_2$ in the photon emission times from a crystal has a rectangular shape. It is constant within the interval

$$|\tau|<T=|l/u_1-l/u_2|=2\pi/\Delta\Omega, \qquad (1^*)$$

and is equal to zero outside of this interval. Here, the group velocities of the signal and idle photons inside the crystal (which are assumed different) and the effective radiation spectrum width are denoted by $u=d\omega/dk$ and $\Delta\Omega$, respectively. Note that the radiation spectrum has the shape $|f(\Omega)|^2=\mathrm{sinc}^2(\Omega T/2)$ where $f(\Omega)$ is the Fourier transform of the distribution function $f(\tau)$. For $l=1$ cm and other typical experimental conditions, Eq. (1) gives $T\sim 1$ ps [in the degenerate case, we have $u_1=u_2$, and quadratic terms should be taken into account in the expansion $\omega(k)$].

An obvious way of decreasing T and hence increasing $\Delta\Omega$ and the accuracy of measuring small group delays by the method of Ref. 3 is to decrease the crystal thickness l. However, in this case, the intensity of photon pair emission ("biphoton flux") decreases too.

In the present work, we examine a method for reducing T which makes use of multilayered crystals (superlattices) and is not accompanied by an intensity drop. The essence of the method is to cancel the dispersion $\omega(k)$ by means of the neighboring layers.

A general treatment of parametric scattering in a layered anisotropic medium with neglect of wave reflection from the layer boundaries is given in Sec. 2. Two types of superlattices are examined in Secs. 3 and 4. The use of a two-layer medium to symmetrize the function $f(\tau)$ is analyzed in Sec. 5. The feasibility of using a three-layer medium for preparing biphotons with arbitrary polarization states is studied in Sec. 6.

2. PARAMETRIC SCATTERING IN A LAYERED MEDIUM

We start from an effective interaction Hamiltonian of the following form

$$H=-\int d^3r \sum_{\alpha\beta\gamma} \chi_{\alpha\beta\gamma}(z)(\mathscr{E}_\alpha^* E_\beta E_\gamma+\mathscr{E}_\alpha E_\beta^+ E_\gamma^+), \qquad (1)$$

where χ is a real quadratic susceptibility tensor, $\mathscr{E}_\alpha$ is the classical positive-frequency pump field, and E_β is the operator of the positive-frequency scattered radiation field. We are interested in collinear scattering parallel to the pump beam (the z-axis) which is nearly degenerate in frequency. In this case, $\alpha, \beta, \gamma=x, y$. We neglect the longitudinal field components and other factors introducing small corrections.

In the diffractionless approximation, the field does not depend on the transverse coordinates x and y, so (1) takes the form

$$H=\sum_{m=1}^{M} H_m+\text{H.c.}, \qquad (2)$$

where

$$H_m \propto \sum_{\alpha\beta\gamma} \chi_{\alpha\beta\gamma}^{(m)} \int_{z_m}^{z_{m-1}} dz \mathscr{E}_\alpha(z,t) E_\beta^+(z,t) E_\gamma^+(z,t). \qquad (3)$$

Here m is the layer number, counting to the left from the "exit" plane $z=0$, z_m and z_{m-1} are the boundaries of the layer m, and $\chi^{(m)}$ is its nonlinearity. Some of the layers may be linear with $\chi^{(m)}=0$. The indices α, β, and γ refer to the laboratory frame rather than the crystallographic one.

 1063-7761/94/060848-05$10.00

We restrict ourselves to the case of optically uniaxial layers with axes belonging either to the xz-plane or to the yz-plane. The waves polarized in the x or y direction are thus normal waves (ordinary or extraordinary) for the whole system of layers. As a result, the field in layer m may be represented in the form

$$E_\alpha^+(z,t) \propto \int_0^{\omega_0} d\omega D_\alpha^{(m)}(z,\omega) e^{i\omega t} a_\alpha^+(\omega), \tag{4}$$

where $a_\alpha^+(\omega)$ is the operator for photon creation in the longitudinal mode with a frequency ω and polarized parallel to the axis $\alpha=x,y$, $D_\alpha^{(m)}(z,\omega)$ is the propagator determining the phase change in the path from the z-plane belonging to the layer m to the $z=0$ plane. The factor $\sqrt{\omega}$ multiplying $a(\omega)$ is omitted here. It can be allowed for in finite normally ordered expressions through the inclusion in the propagators $D(\omega)$ or the detector efficiency $\eta(\omega)$.

Neglecting reflected waves, we have

$$D_\alpha^{(m)}(z,\omega) = \exp\left[i \sum_{n=1}^{m-1} k_\alpha^{(n)} l_n + i k_\alpha^{(m)} (z_{m-1} - z) \right]. \tag{5}$$

Here we have written $l_n = z_n - z_{n-1}$, and $k_\alpha^{(n)}(\omega)$ is the coupling resulting from the dispersion relation for an ordinary or an extraordinary wave polarized in the α direction in the layer n.

We assume the pumping field to be monochromatic:

$$\mathscr{E}_\alpha(z,t) = i D_\alpha^{(m)*}(z,\omega) e^{-i\omega_0 t} A_\alpha, \quad z_m < z < z_{m-1}, \tag{6}$$

where iA_α are the pumping field components in the plane $z=0$ and $t=0$. The phase i is added for convenience.

From (3)–(6), we get

$$H_m \propto \iint d\omega d\omega' \chi_m A_m a_{\beta_m}^+(\omega) a_{\gamma_m}^+(\omega') e^{i(-\omega_0+\omega+\omega')t} \times \int_{z_m}^{z_{m-1}} dz D_{\alpha_m}^{(m)*}(z,\omega_0) D_{\beta_m}^{(m)}(z,\omega) D_{\gamma_m}^{(m)}(z,\omega'), \tag{7}$$

where

$$\chi_m = \chi_{\alpha_m \beta_m \gamma_m}^{(m)}(\omega_0;\omega,\omega'), \quad A_m = A_{\alpha_m},$$

and we have allowed for a possible dispersion χ. There is no summation over the α, β, and γ indices here because we assumed that phase velocity matching necessary for efficient interaction occurs in each nonlinear layer only for a certain combination of indices, α_m, β_m, and γ_m.

Hence, we find the state vector of the scattered field in first-order perturbation theory

$$|\psi\rangle = |0\rangle + \frac{1}{i\hbar} \int_{-\infty}^{\infty} dt \sum_m H_m |0\rangle = |0\rangle + \int_{-\omega^*}^{\omega^*} d\Omega \sum_m F_m(\Omega) a_{\beta_m}^+(\Omega) a_{\gamma_m}^+(-\Omega) |0\rangle, \tag{8}$$

where the following notation is used:

$$\Omega = \omega - \omega^*, \quad \omega^* = \omega_0/2, \quad F_m(\Omega) = \Gamma_m f_m(\Omega),$$

$$\Gamma_m = 2\pi\omega^* l_m \chi_m A_m / c,$$

$$f_m(\Omega) = e^{-i\varphi_m}(1 - e^{-i\delta_m})/i\delta_m,$$

$$\varphi_m = \sum_{n=1}^{m-1} \delta_m^{(n)}, \quad m = 2,3,\ldots,M, \tag{9}$$

$$\delta_m^{(n)} = [k_{\alpha_m}^{(n)}(\omega_0) - k_{\beta_m}^{(n)}(\omega^* + \Omega) - k_{\gamma_m}^{(n)}(\omega^* - \Omega)] l_n,$$

$$\delta_m = \delta_m^{(m)}, \quad \varphi_1 = 0.$$

Here, Γ_m and $\delta_m^{(n)}$ represent parametric gain factor in the layer m and the phase detuning in the layer n, respectively, for the fields with polarizations α_m, β_m, and γ_m. The approximation $\omega(\omega_0 - \omega) \simeq \omega^{*2}$ is used.

Thus, the function

$$F_m(\Omega) = \Gamma_m f_m(\Omega) = \Gamma_m e^{-i(\varphi_m + \delta_m/2)} \operatorname{sinc}(\delta_m/2) \tag{10}$$

determines the contribution of the mth layer to the amplitude of the state $|1\rangle_{\Omega,\beta_m}|1\rangle_{-\Omega,\gamma_m}$ in which one photon has a frequency $\omega^* + \Omega$ and polarization β_m and another photon has a frequency $\omega^* - \Omega$ and polarization γ_m. All the photons belong to one transverse mode with an axis parallel to the z-axis. The contributions from different layers to the total biphoton field can interfere. In the case of identical nonlinear layers separated by a linear gap (vacuum, in particular), this is an analog of Ramsey interference observed in beam spectroscopy (see Ref. 9).

3. CANCELLATION OF DISPERSION BY MEANS OF LINEAR INTERMEDIATE LAYERS

We assume there are N identical nonlinear layers of thickness l with optical axes in the plane yz and a type-I interaction ($\alpha\beta\gamma = yxx = eoo$) separated by $N-1$ linear gaps of thickness l'. This model has already been treated in Ref. 9, and only a part of the results are presented here.

We adopt the notation $\delta_{2n+1} = \delta$, $\delta_{2n} = \delta'$. Then it follows from (9) that $\varphi_m = (m-1)(\delta + \delta')$ and

$$f(\Omega) = \sum_{m=1}^{N} f_m(\Omega) = (i\delta)^{-1}(1 - e^{-i\delta}) \sum_{m=1}^{N} e^{-i\varphi_m} = \frac{\sin(\delta/2)}{\delta/2} \frac{\sin[N(\delta+\delta')/2]}{\sin[(\delta+\delta')/2]} \exp\{-i[N\delta + (N-1)\delta']/2\}. \tag{11}$$

Here, the first factor describes ordinary parametric scattering in a layer of thickness l and the second describes interference between layers. Assume $\delta(\Omega) + \delta'(\Omega) = \text{const}$, i.e. the dispersion is cancelled out. Then the spectral width is determined solely by the nonlinear layer thickness. As a result, for small l a wide spectrum results, and the photons in a pair can be localized precisely, [see (1*)]. In this case the total biphoton flux intensity is determined by the total thickness Nl.

The Fourier transform $f(\tau)$ of the function $f(\Omega)$ determines the probability amplitude for emission of a pair of photons from the plane $z=0$ at times t and $t+\tau$. As shown in Ref. 9, the function $f(\tau)$ consists of square pulses of length T [see (1*)]. These pulses overlap and interfere if dispersion is cancelled.

4. CANCELLATION OF DISPERSION IN TYPE-II INTERACTION

Let us consider a system of identical nonlinear layers. Assume the axes of the odd and even layers lie in the planes yz and xy, respectively. The optical axes make the same angle with the z-axis in all the layers, providing phase matching $\delta(0)=0$ at the degenerate frequency $\Omega=0$ for type-II interactions: $yxy=eoe$ in odd layers and $xxy=eeo$ in even layers. Thus, the orientations of crystallographic axes in adjacent layers differ from one another by a 90° rotation about the z-axis.

According to (8),

$$|\psi\rangle=|0\rangle+\int d\Omega F(\Omega)a_x^+(\Omega)a_y^+(-\Omega)|0\rangle, \qquad (12)$$

where $F=\Sigma_m F_m$. In the group-velocity approximation, we have

$$\delta(\Omega)\equiv[k_e(\omega_0)-k_0(\omega^*+\Omega)-k_e(\omega^*-\Omega)]l\approx\Omega T,$$

$$\delta'(\Omega)\equiv[k_0(\omega_0)-k_e(\omega^*+\Omega)-k_0(\omega^*-\Omega)]l\approx\varepsilon-\Omega T,$$

where $T=l(u_e^{-1}-u_0^{-1})$ and $\varepsilon=\delta'(0)$.

For $m=2n+1$, we find from (9)

$$\begin{aligned}&\Gamma_m\propto\chi_{yxy}A_y,\\ &f_m(\Omega)=e^{-in\varepsilon}f_1(\Omega),\\ &\varphi_m=n(\delta+\delta')\approx n\varepsilon,\\ &f_1(\Omega)=(1-e^{-i\Omega T})/i\Omega T=e^{-i\Omega T/2}\operatorname{sinc}(\Omega T/2).\end{aligned} \qquad (13)$$

For $m=2n$, we have

$$\begin{aligned}&\Gamma_m\propto\chi_{xxy}A_x,\\ &f_m(\Omega)=e^{(-in\varepsilon+i\Omega T)}f_1(\Omega),\\ &\varphi_m=n(\delta+\delta')-\delta\approx n\varepsilon-\Omega T.\end{aligned} \qquad (14)$$

Here, $\chi_{\alpha\beta\gamma}=\chi_{\alpha\beta\gamma}(\omega_0,\omega^*+\Omega,\omega^*-\Omega)\equiv\chi_{\alpha\beta\gamma}(\omega_0,\omega^*-\Omega,\omega^*+\Omega)$ are the components of the tensor χ in the laboratory frame. Neglecting the dispersion of χ on the interval $\Delta\Omega$ we get $\chi_{xxy}=\chi_{xyx}$.

Let x' and y' be unit vectors linked to the crystal in the even (rotated by an angle of 90°) layers, then $x'=y$, $y'=-x$. Consequently, for even layers, we have $\chi_{xyx}=\chi_{y'x'y'}$ and the parameters Γ_{2n+1} and Γ_{2n} differ only in the components A_y and A_x of the pump field.

We require that $|\Gamma_{2n+1}|=|\Gamma_{2n}|$. This necessitates that the condition $A_x/A_y=e^{i\vartheta}$ be fulfilled, i.e., the pump polarization in the plane $z=0$ should correspond to a point on the Poincaré sphere with coordinates $(90°-\vartheta,\varphi=90°)$. For $\vartheta=0$ and $\vartheta=90°$, these correspond to linear polarization at an angle of 45° and circular polarization, respectively. The necessary pump polarization at the input to the system (at $z=z_M=-\Sigma l_m$) is determined from (5) and (6).

As a result we find

$$\begin{aligned}&F_m(\Omega)=\Gamma_1e^{-i\alpha_m}f_1(\Omega),\\ &\alpha_{2n+1}=n\varepsilon,\\ &\alpha_{2n}=n\varepsilon+\vartheta-\Omega T.\end{aligned} \qquad (15)$$

The spectrum of a biphoton wave packet is described by a function

$$F(\Omega)=\sum_{m=1}^{M}F_m(\Omega)$$
$$=\Gamma_1F_1(\Omega)[1+e^{-i(\varepsilon+\vartheta-\Omega T)}+e^{-i\varepsilon}+\dots]. \qquad (16)$$

Let $\vartheta=0$ and $\varepsilon=2k\pi$. Then for an even number of layers M, we get

$$F(\Omega)=\frac{M}{2}\Gamma_1f_1(\Omega)(1+e^{i\Omega T})=M\Gamma_1\operatorname{sinc}(\Omega T). \qquad (17)$$

The bandwidth π/T and the intensity are thus determined by the thickness $2l$ of a pair of layers and the total thickness Ml of the material, respectively. Note that $F(\Omega)$ does not contain a frequency-dependent phase factor, so the distribution function $f(t_x-t_y)$ is even (t_α are the times at which photons with polarization α are detected).

Let the pump be polarized parallel to the y-axis. Then we have $\Gamma_{2n}=0$, i.e., even layers play a passive part compensating the dispersion in odd layers, as in the model presented in Sec. 3. The even terms in the sum (16) should now be set equal to zero:

$$F=\Gamma f_1(\Omega)(e^{-iN\varepsilon}-1)/(e^{-i\varepsilon}-1). \qquad (18)$$

Here $N=(M-1)/2$ is the number of active layers with axis in the yz-plane. For $\varepsilon=2k\pi$, we get (compare with (11) for $\delta=-\delta'=\Omega T$)

$$F=N\Gamma e^{-i\Omega T/2}\operatorname{sinc}(\Omega T/2). \qquad (19)$$

5. SYMMETRIZATION OF TYPE-II BIPHOTONS

Let us consider two anisotropic layers, linear $(m=1)$ and nonlinear $(m=2)$, with axes in the yz-plane. We assume that the interaction $yxy=eoe$ takes place. According to (8) and (9),

$$|\psi\rangle=\left|0\rangle+\int d\Omega F(\Omega)a_x^+(\Omega)a_y^+(-\Omega)\right|0\rangle, \qquad (20)$$

where

$$\begin{aligned}&F=2\pi\omega^*l_2c^{-1}\chi_{yxy}A_yf(\Omega),\\ &f=\operatorname{sinc}(\delta_2/2)\exp[-i(\delta_1+\delta_2)/2],\\ &\delta_m=[k_e^{(m)}-k_0^{(m)}(\Omega)-k_e^{(m)}(-\Omega)]l_m.\end{aligned} \qquad (21)$$

In the linear approximation for the dispersion, we have

$$\delta_1(\Omega)=\varepsilon+T_1\Omega,\quad \delta_2(\Omega)=T_2\Omega, \qquad (22)$$

where $\varepsilon=\delta_1(0)$ and

$$T_m=l_m/u_e^{(m)}-l_m/u_0^{(m)}. \qquad (23)$$

The Fourier transform of the function $f(\Omega)$ determines the probability amplitude for detecting photons at the times t_x and $t_x+\tau$ (when the distances from the detectors to the exit plane $z=0$ are equal). According to (21) and (22),

$$f(\tau)\equiv\int d\Omega e^{i\Omega\tau}f(\Omega)/2\pi=\frac{1}{T_2}e^{-i\varepsilon}\Pi\left(\frac{\tau-T_1-T_2/2}{T_2}\right), \tag{24}$$

where $\Pi(x)$ is a square-wave function (the Fourier transform of the function $\mathrm{sinc}(\Omega)$): $\Pi(x)=1$ for $-1/2<x<1/2$, and $\Pi(x)=0$ otherwise. Consequently, for $T_1=-T_2/2$, the distribution function becomes even: $f(\tau)\propto\Pi(\tau/T_2)$, i.e. x-photons are detected earlier or later than y-photons with equal probabilities. This symmetry is essential when observing polarization interference effects and ultrashort delays.[6]

6. PREPARATION OF BIPHOTONS WITH ARBITRARY POLARIZATION

In classical optics, it is usual to specify the polarization state, i.e., the transverse structure of a plane monochromatic wave, by means of three numbers, for instance, the Stokes parameters. However, such a description does not completely determine the results of experiments in which not only the intensity is recorded, but also its fluctuations and correlations or still higher-order moments as well.[7] Thus, in the general case, the transverse structure of a plane wave is given by more than three parameters.

For instance, in the case of a plane wave with a certain number of photons N, the photons may be distributed over two orthogonal modes in $N+1$ ways, so the field state is given by $N+1$ complex numbers $(c_0,c_1,...,c_N)$ constituting the polarization vector in a $(2N+2)$-dimensional space[10]. If we are not interested in the overall phase of the state, then $2N$ independent real numbers remain in view of the normalization condition $\Sigma|c_i|^2=1$. These numbers give a point on the sphere S^{2N} in a $(2N+1)$-dimensional space.

For $N=1$, the space of states can be mapped onto the Poincaré sphere as in the classical case. For $N=2$, we have four polarization parameters and an S^4 sphere as the projective space. The polarization transformers are characterized by two parameters, so they do not provide a way to go between two arbitrary points on S^4. The missing degrees of freedom (in the case when a two-photon state is prepared using parametric scattering) can be supplied by using two or three nonlinear layers with different types of interaction.

Let us consider a sequence of three layers with interactions $yxx=eoo$, $yxy=eoe$, $xyy=eoo$. The optical axes belong to the planes yz, yz, and xz, respectively. According to (8), we have a state

$$|\psi\rangle=|0\rangle+\int d\Omega(F_0a_x^+a_x^{+\prime}+F_1a_x^+a_y^{+\prime}+F_2a_y^+a_y^{+\prime})|0\rangle, \tag{25}$$

where the primed operators are related to modes with frequency $-\Omega$ and the functions $F_m(\Omega)$ are determined from Eqs. (9).

By adjusting the parameters, we can obtain arbitrary relations between the polarization components $c_m\propto F_m$, i.e., prepare a biphoton with arbitrary polarization at a given pair of frequencies $\omega^*+\Omega$ and $\omega^*-\Omega$. The frequency dependence of F_m after Fourier transformation determines the polarization-time properties of the biphotons. For wave packets emitted from different layers to overlap in time, narrow-band frequency filters which "stretch" the packets can be used (see Ref. 6, where this method was used to observe interference in the case of type-II interaction).

This method for preparing biphotons with predetermined polarization properties may be called sequential. Note that only two crystals can be used when transforming a two-component beam with ordinary polarization transformers. In this case, the c_m components are mutually transformed using $SU(2)$ matrices of dimensionality 3×3, the analogs of the Jones matrices.[7,10]

We can use also a parallel method with two or three crystals located close to each other and giving spatially separated beams. When these beams are combined using semitransparent mirrors, a single beam with predetermined polarization properties is obtained.

7. CONCLUSION

Our analysis was restricted to the first order of perturbation theory, i.e., spontaneous scattering. If pumping is sufficiently intense, the probabilities of emitting four, six, etc., photons at one time become noticeable. In this case, the parameter Γ becomes of order or greater than 1 (parametric superfluorescence or squeezed-vacuum generation). The use of layered media to control the spectrum-time and the polarization structure of radiation is possible in this case as well.

The implementation of this method will involve a number of technological and other problems. To obtain a second-order coherence time less than 10^{-14} s, one needs to produce a superlattice of oriented single-crystal layers with a period of order 0.1 mm and a number of layers of order 10–100. The linear dispersion approximation and the neglect of reflections from layer interfaces can prove to be invalid in this case.

Note that we can use multiple beam transmission through a single thin plate consisting of one or two layers, instead of a superlattice. Polydomain single crystals may also be of interest (see Ref. 11 in which experiments on observing angular-frequency spectrum of parametric scattering in such crystals are described).

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Translated by A. M. Mozharovskii

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