

Spatiotemporal grouping of photons in spontaneous parametric scattering of light

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The results of an experimental investigation of the spatial (transverse) and temporal (longitudinal) grouping of photons in the process of three-photon parametric scattering of light (PSL) are described in the paper.

PSL (see, e.g., Refs. 1-3) is the process of interaction of photons of monochromatic radiation (pumping) with a medium having no center of symmetry. As a result, at the exit from the medium radiation is formed occupying a broad section of the spectrum, in the Stokes region relative to the pumping frequency, in the entire range of transparency of the scattering medium - a piezoelectric crystal. An elementary act of scattering consists of the decomposition of a pumping photon with frequency ω_0 and wave vector k_0 into a pair of photons with frequencies ω_1 and ω_2 and wave vectors k_1 and k_2 .

The parameters of the photons satisfy the conditions

$$\omega_1 + \omega_2 = \omega_0; \quad k_1 + k_2 = k_0. \quad (1)$$

It has been shown⁴ and confirmed experimentally⁵ that photons related by the condition (1) (parametrically coupled photons) are characterized by grouping ("superclassical" in comparison with Brown-Twiss grouping⁶) in the time of transit through a given spatial plane $\{z\}$ perpendicular to the direction $k_0/|k_0|$. Besides temporal grouping, parametrically coupled photons possess spatial grouping in the $\{z\}$ plane.⁷

Spatiotemporal grouping of photons implies an increased probability of simultaneously finding two photons at the space-time points x_1 and x_2 ; $x_i \equiv \{z_i, \rho_i, t_i\}$, $i = 1, 2$; z_i is the coordinate on the axis $z \parallel k_0$ with the origin on the exit face of the scattering medium (k_0 is usually perpendicular to this face); ρ_i is the radius vector defining the position of the point x_i in the $\{z_i\}$ plane having the coordinate z_i and perpendicular to z ; t_i are the moments (times) of observation.

From the experimental conditions, $z_1 = z_2 = z$. In this case the probability of detecting a parametrically coupled

pair of photons with frequencies ω_1 and ω_2 at the points x_1 and x_2 is determined, according to Ref. 7, by the expression

$$f_{12}(z, \rho, t) \equiv f_{12}(\rho, t) = \frac{1}{1 + (\tilde{z} + \tilde{r})^2} \exp \left[\frac{-\rho^2 q_m^2}{1 + (\tilde{z} + \tilde{r})^2} \right]. \quad (2)$$

Here q_m is the transverse component, in the $\{z\}$ plane, of the wave vector of the scattering, and determines the angular spectrum of the scattering;

$$\tilde{z} = 2zz_m^{-1}; \quad \tilde{r} = 2l \cdot z_m^{-1}; \quad \tilde{r} = 2t(dx)^{-1}, \quad \alpha \equiv (u_1^{-1} - u_2^{-1}); \quad t = |t_2 - t_1|;$$

$$u_i = d\omega_i/dk_i; \quad k_i = \frac{n_i \omega_i}{c}; \quad k_1 \approx k_2 = k; \quad \rho = |\rho_2 - \rho_1|;$$

$z_m \equiv k/q_m^2$ is a parameter defining the size of the near zone with respect to z in accordance with the condition $\rho < a_0 - \delta_m(z + l)$; a_0 is the diameter of the pumping beam; l is the length of the scattering layer along z ; $\delta_m \equiv k/q_m$ is the scattering angle.

The biphoton length $l^{(2)}$ (the average distance in the direction $z/|z|$ between parametrically coupled photons) is determined by the second-order coherence time⁷ $t^{(2)}$, and for the case of almost collinear scattering ($\rho \approx 0$) it equals

$$t^{(2)} = 2f_{11}^{-1}(0, 0) \int_0^\infty dt f_{12}(0, t). \quad (3)$$

From (3) it follows that the biphoton length varies in the range from $l_{\min}^{(2)} \equiv c(2\Delta\omega_{sc})^{-1}$ ($\Delta\omega_{sc}$ is the frequency width of the PSL spectrum) in the scattering near zone ($z \ll z_m$) to $l_{\max}^{(2)} \equiv 2c(\Delta\omega_s^{-1})$ ($\Delta\omega_s$ is the frequency width of wave synchronism) in the far zone ($z \gg z_m$). The coherence time⁽²⁾ and the biphoton length $l^{(2)}$ are determined by the relative delay of the photons in the medium on account of the dispersion of its dielectric permittivity. In both the near and the far zone the biphoton length $l^{(2)}$ and the photon lengths $l^{(1)}$ determined by the first-order coherence functions are both approximately equal⁷

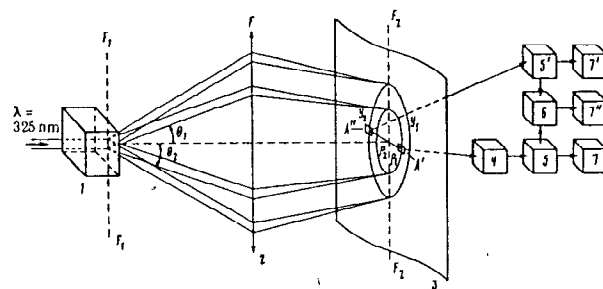


FIG. 1. Scheme of the experimental apparatus for observing the phenomenon of spatial and temporal grouping of photons in spontaneous parametric scattering: 1) Nonlinear crystal; 2) thin focusing lens; 3) focal plane of the lens; 4) monochromator for channel 1; 5, 5') photoelectron receivers for channels 1 and 2; 6) coincidence circuit; 7, 7', 7'') pulse counters.

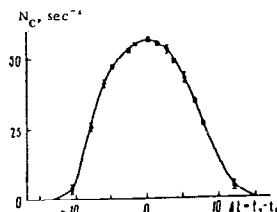


FIG. 2. Curve of coincidences of pulses in channels 1 and 2 as a function of the electronic delay between the channels; 0 corresponds to the introduction of a fixed delay $\Delta t_f = 7$ nsec, $\lambda_1 = 630$ nm, $\lambda_2 = 670$ nm, $T_{\text{obs}} = 100$ sec; there were five measurements at each point.

$$\rho^{(2)} = 1/q_m [1 + (\tilde{r} + \tilde{r}')^2]^{1/2}. \quad (4)$$

From (4) it follows that at $t_1 \approx t_2 \approx 0$, i.e., at the time of pair creation, in the scattering near zone $\rho^{(2)} = q_m^{-1}$ and is determined by the angular width of the spectrum of scattered radiation. In the far zone $\rho^{(2)} \approx 2\theta z$ and is determined by the distance traveled by the photons emitted from one biphoton, and can reach considered values.

In the present work we use a biphoton-field detector consisting of two photon counters, linear with respect to intensity, and a correlator — a coincidence circuit (Fig. 1). A lithium-iodate crystal excited by coherent monochromatic radiation with a wavelength of 325 nm and a power of $5 \cdot 10^{-3}$ W served as the source of the biphoton field. The crystal had a length $l = 1.5$ cm. Variation of the orientation of the crystal relative to the pumping beam enabled us to vary the frequency-spatial distribution $\phi_i(\omega_i)$ of the scattered radiation (see the reconstructed PSL curves in Ref. 9).

To simplify the experimental scheme, the angular distribution ϕ_i was converted to a distribution $\rho_i(\omega_i)$ in a plane using a thin lens with a focal length $F = 10$ cm. The focal plane $[F_1]$ of the lens was matched with the exit face of the crystal while its axis was matched with the axis of the pumping beam. Then in the second focal plane $[F_2]$ of the lens the radiation of each color formed a ring, so that a system of concentric rings of different colors, the radii of which are connected with the scattering angle through the focal length ($\rho_i(\omega_i) = F \tan \theta_i$), was ultimately formed in the $[F_2]$ plane. The dispersion of the radii of the rings was determined by the dispersion of the index of refraction of the crystal and by the orientation. The width (in ρ_i) of the ring formed by radiation of frequency ω_i was determined by the first-order coherence length $\rho^{(1)}$, and in the experiment had the order of magnitude $\rho^{(1)} \approx 0.01$ – 0.02 cm. In this case the biphoton length was of the order of $l^{(2)} \approx 0.2$ cm, while the coherence time was of the order of $t^{(2)} \approx 6 \cdot 10^{-12}$ sec. As a consequence of the condition (1), parametrically coupled points lay in diametrically opposite regions of the ring system. We used a nearly collinear and degenerate regime of scattering ($\lambda_1 \approx \lambda_2 \approx 650$ nm, $\theta_i < 1$).

The entrance openings of the receivers were placed in the $[F_2]$ plane and their positions could be varied both along the radius vector ρ_i and in directions transverse to it (along a tangent to a ring of radius Y).

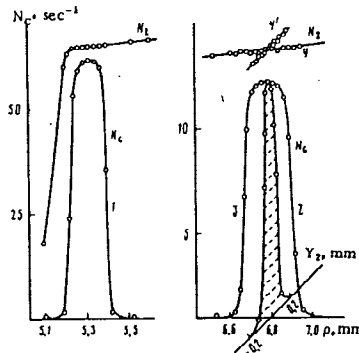


FIG. 3. Spatial distribution of the numbers of readings and coincidences in channels 1 and 2 (in the $[F_2]$ plane). 1) $N_c = N_c(\rho_1)$ ($\rho_1 = 5.3$ mm, $\lambda_1 = 650$ nm); 2) $N_c = N_c(\rho_2)$ ($\rho_2 = 6.8$ mm, $\lambda_1 = 630$ nm); 3) $N_c = N_c(Y_2)$ ($\rho = 6.8$ mm); 4, 4') $N_2 = N_2(\rho_2, Y_2)$.

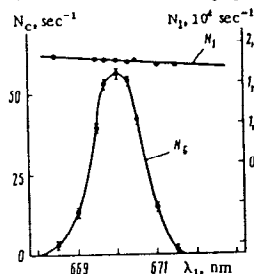


FIG. 4. Dependence of N_c and N_1 on the transmission wavelength of channel 1 for a fixed position of the spatial filter of channel 2. $\lambda_1 = 630$, $\Delta \lambda_1 = 1.5$ nm, $\lambda_2 = 670$, $\Delta \lambda_2 = 1.5$ nm.

The photoreceiver channels differed somewhat from one another. Channel 1 recorded photons of frequency ω_1 , spectral width $\Delta \omega_1$, assigned direction $k_1/|k_1|$, and angular width $\Delta \theta_1$ in the $[F_2]$ plane. A square opening, the area of which could be varied in the range from 0 to 0.16 mm², served as the spatial filter. A spectrograph of the ISP-51 type in the monochromator regime served as the spectral filter. Channel 2 included only a spatial filter, similar to the filter of the first channel.

Photomultipliers of the FEU-79 type with following electrical digital circuits served as the photon counters in both channels. The circuits put out current pulses of duration 10–15 nsec at half-height. The number of pulses was recorded with instruments of the ChZ-34 type. A coincidence circuit of the summing type, yielding a pulse at the output if the time delay between pulses at the input did not exceed 12 nsec, served as the correlator. The dead time was no more than 2–3 nsec. The number of coincidence pulses was recorded by a ChZ-34 instrument. All three pulse counters were strictly synchronized in operating time.

The dependences of the count rate N_c of the coincidence pulses on the position of the spatial filter in channel 2 along the ρ_2 and Y_2 coordinates (for a fixed position of the spatial and frequency filter of channel 1), on the transmission frequency ω_1 of channel 1 for a fixed position of the spatial filters of channel 2, and on the time delay in the electrical circuit of channel 2 were investigated experimentally.

Temporal grouping of photons. In Fig. 2 we present a characteristic dependence of N_C on the introduction of a delay in the time of measurement in channel 2 with respect to that in channel 1. To obtain this curve channel 1 was tuned to receive radiation with a wavelength $\lambda_1 = 630$ nm while channel 2 was tuned to $\lambda_2 = 670$ nm. The spatial filters were set at points of the $[F_2]$ plane with coordinates calculated from the dispersion characteristics of the crystal. The maximum of the N_C curve was observed with the introduction of a delay $\Delta t \approx 7$ nsec into channel 2, which was due to the need to compensate for the difference in the optical paths of the channels. With the introduction of a more or less compensating delay of ~ 20 nsec, the coincidence-count rate fell to the count level for random coincidences, viz., $N'_C \approx 5 \cdot 10^{-1} \text{ sec}^{-1}$. We note that in Ref. 5, where temporal grouping was observed, the count rate N_C for coincidence pulses at the maximum of the curve did not exceed 2 sec^{-1} and differed little from N'_C .

Spatial grouping of photons. The investigation of the spatial grouping of photons comes down to an investigation of the dependence on the mutual arrangement of the spatial filters of the photoreceiver channels and on their positions in the $[F_2]$ plane.

In Fig. 3 we present the dependences $N_C(\rho_2)$ for two values of ρ_1 and ω_1 (curves 1 and 2) and the dependence $N_C(\mathbf{Y})$ (curve 3, with the hatched plane of the figure). Values of the count rate $N_2(\rho_2)$ in channel 2 are also given there. The contrast of the curve, i.e., the ratio of $N_C \text{ max}$ to N'_C exceeded 10^2 in order of magnitude. The decrease in $N_2(\rho_2)$ at small ρ^2 is connected with the characteristic distribution of the scattering in space on account of the dispersion of the dielectric permittivity of the medium. A change in the frequency ω_1 leads to a shift in the maximum of the $N_C(\rho_2)$ curve (curve 2) as a consequence of the nonzero slope of the reconstructed PS curve $\theta_1 = \theta_1(\omega_1)$. Curve 3 was obtained by moving the spatial filter along a tangent to a circle of radius $\rho_2 = 6.8$ mm at the maximum of curve 2. Since channel 2 did not contain any spectral filters and the spectral curve of sensitivity of the FEU-79 is a sufficiently smooth curve in this range, the observed maximum of the dependence of the count rate of coincidence pulses on the position of the spatial filter of the photoreceiver channel indicates the presence of spatial grouping of parametrically coupled photons with a coherence length $\rho^{(1)} \approx 0.1\text{--}0.2$ mm.

The dependence, presented in Fig. 4, of N_C on the transmission wavelength λ_1 of channel 1 for fixed positions of the spatial filters of channel 2 corresponding to the position of the maximum of curve 2 in Fig. 3 confirms the presence of grouping only for parametrically connected photons.

Degree of grouping. To estimate the degree of grouping — the ratio $g^{(2)} = N_C^{ph}/N_b$ of the intensity N_C^{ph} of the photon flux in channel 1 to the intensity N_b of the bi-photon flux, we estimated the quantum efficiency of channel 2 for two values of the wavelength, $\lambda = 630$ and 670 nm. To isolate the signal at just this wavelength we introduced frequency selectors — interference light filters — into the channel. With allowance for their transmission, the quantum efficiency η_2 of channels 2, measured using a secondary light standard of type SI-8, proved to be

$$\eta_2/630 \text{ nm} = 2.5 \pm 0.4\% \text{ and } \eta_2/670 \text{ nm} = 1.8 \pm 0.5\%.$$

The determination of the quantity $g^{(2)}$ is based on the fact that, owing to the independence of the processes of measurement by the channels, the count rate N_C of the number of coincidence pulses is $N_C = \eta_1 \eta_2 N_b$; η_1 is the quantum efficiency of channel 1. Then $g^{(2)} = \eta_2 N_1 N_C^{-1}$. With allowance for the values of η_2 and the influence of Fresnel reflection at the faces of the crystal and at the surfaces of the thin lens on the pairing, it was found that $g^{(2)} \approx 0.94$ on the average. The accuracy of the measurements of the degree of grouping was determined mainly by the accuracy of the determination of the quantum efficiency by the standard photometric method, and equalled $\Delta g^{(2)} \approx 0.2$. Thus, to within the experimental accuracy, the parametrically scattered light is entirely two-photon light.

The two-photon character of the parametrically scattered light allows us to solve the problem of the absolute determination of the quantum efficiency of the photoreceivers. The values of the quantum efficiency can be easily determined from a measurement of the count rates N_1 , N_2 , and N_C of pulses in channels 1 and 2 and of coincidence pulses, using the formulas $\eta_1 = N_C/N_2$ and $\eta_2 = N_C/N_1$.

We note the possibility of one-channel calibration of a photomultiplier,^{10,11} when one directs first photons of frequency ω_1 , then photons of frequency ω_2 , and then biphotons consisting of photons of these frequencies to the one receiver being calibrated. The quantum efficiency is determined from the relation $\eta_{2(1)} = 1 - \frac{M_b - N_{1(2)}}{N_{2(1)}}$, where

M_b is the count rate of the number of pulses (both one- and two-electron pulses) in the recording of the biphotons, while N_1 and N_2 are the alternate-counting rates for the one-photon components of the biphoton field. Experiments conducted on one-channel calibration showed¹² the possibility of attaining high accuracy (as good as 2–3%) in the measurement of η .

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