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Relationship between quantum two-photon correlation and classical spectrum of light

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We show that the two-photon temporal (longitudinal) correlation of light is limited by its classical optical spectral width. We present examples of the sources of two-photon light approaching this limit.

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The discovery of two-photon quantum correlation has greatly contributed to fundamental physics and, more recently, to the potentially applied areas of quantum information and technology. Interest in these fields brings up a question about the ultimate resolution that can be achieved with two-photon correlation measurements, i.e., the question about the fundamental properties of two-photon correlation function. Study of the biphoton *transverse* correlation function [1–4] shows that it is possible to mutually localize two photons two times better than each photon can be localized taken separately. For such areas as quantum metrology (e.g., [5,6]) and quantum lithography [2,7], it is important to understand if the superclassical photon correlation in the *longitudinal* direction, or temporal correlation, is also possible.

We discuss the connection between the quantum two-photon temporal correlation and the classical spectrum of light. We show that universal integral relations between these functions impose very general limitations on how short a two-photon correlation time could be achieved. This limitation is set approximately by the inverse spectral width of light. This implies that light with a strong two-photon correlation cannot be narrow band, and therefore the use of narrow-band photonic devices, such as those proposed in [8], for generation or handling of such light, is not allowed.

In the absence of bandwidth-limiting structures, optical linewidth and hence the two-photon correlation time is limited by the transparency window of the nonlinear material used for generation of the photon pair. We consider parametric down conversion and two-photon decay of an atom as examples of a photon pair source, and show that in both cases this limitation holds. We also found specific cases of parametric down conversion that provide the shortest possible correlation time, which is a few femtoseconds. This result suggests a relatively simple technique for achieving femtosecond resolution in continuous-wave optical measurements.

Let us consider two optical fields, $E_1(t)$ and $E_2(t)$, observed at the same spatial location. According to Glauber [9,10], the fields can be described with the first- and the second-order correlation functions

$$G^{(1)}(\tau) \equiv \langle E_1^{(-)}(t) E_2^{(+)}(t+\tau)\rangle,$$

$$G^{(2)}(\tau) \equiv \langle E_1^{(-)}(t) E_2^{(-)}(t+\tau) E_1^{(+)}(t) E_2^{(+)}(t+\tau)\rangle. \quad (1)$$

The first-order correlation function $G^{(1)}(t)$ is the autocorrelation function for $E_1(t)=E_2(t)$.

Let us consider a generic two-photon state which can be entangled or separable, and degenerate or nondegenerate, depending on the weighting function $h(\nu)$,

$$|\Psi\rangle = \int_{-\infty}^{\infty} d\nu h(\nu) a_1^{\dagger}(\omega_0+\nu) a_2^{\dagger}(\omega_0-\nu)|0\rangle, \quad (2)$$

For this state, one can substitute (2) into (1) and show [11] that the correlation functions $G^{(1)}(t)$ and $G^{(2)}(t)$ are closely related,

$$G^{(1)}(t) = \int_{-\infty}^{\infty} d\nu |h(\nu)|^2 e^{-i\nu t} = \int_{-\infty}^{\infty} f(t') f^*(t'-t) dt', \quad (3)$$

$$G^{(2)}(t) = \left| \int_{-\infty}^{\infty} d\nu h(\nu) e^{-i\nu t} \right|^2 = |f(t)|^2, \quad (4)$$

where we introduced

$$f(t) \equiv \int_{-\infty}^{\infty} d\nu h(\nu) e^{-i\nu t}. \quad (5)$$

From Eq. (3) and the Wiener-Khintchine theorem we find the relation between the weighting function and the power spectrum of the field: $S(\omega)=|h(\omega)|^2$. Since $S(\omega)$ always has a finite frequency width, $G^{(1)}(t)$ has the finite duration limited by the inverse spectral width. This duration cannot be shorter than the optical cycle, because otherwise the optical-field spectrum would extend beyond the zero frequency. Such an argument cannot be directly applied to the $G^{(2)}(t)$ which characterizes the two-photon correlation time. In what follows we discuss the questions about the fundamental limitations on the $G^{(2)}(t)$ and practical approaching of the fundamental limits.

We address the following fundamental questions: (i) What is the minimum characteristic duration $\tau^{(2)}$ of $G^{(2)}(t)$, and (ii) what is the minimum ratio $\tau^{(2)}/\tau^{(1)}$ of characteristic durations of $G^{(2)}(t)$ and $G^{(1)}(t)$, if the spectrum $S(\omega)$ is localized in the frequency interval $[\omega_0-\Omega/2, \omega_0+\Omega/2]$, given by, e.g., the transparency window of a nonlinear material? Question (ii) is important for optical applications where precision ranging or timing is required, but using broadband light due to, e.g., strong dispersion is precluded.

From the properties of Fourier transforms we find that both minima of $\tau^{(2)}$ and $\tau^{(2)}/\tau^{(1)}$ are achieved in the strongly nondegenerate case, where the single-photon spectrum con-

sists of two vanishingly narrow (e.g., rectangular) frequency regions of width $\delta\omega$, centered at $\omega_0-\Omega/2$ and $\omega_0+\Omega/2$. In this case Eqs. (3) and (4) yield

$$G^{(1)}(t) = -\frac{1}{2\delta\omega}\mathrm{sinc}(\delta\omega t)\cos\left(\frac{\Omega t}{2}\right), \tag{6}$$

$$G^{(2)}(t) = \mathrm{sinc}^2(\delta\omega t)\cos^2\left(\frac{\Omega t}{2}\right). \tag{7}$$

We see that both correlation functions have wide envelopes filled with narrow fringes. Considering the fringe quarter period as the characteristic correlation time, we see that $\tilde{\tau}^{(2)} = \pi/2\Omega = \tilde{\tau}^{(1)}/2$ (tilde is to emphasize that we compare periodic functions).

In the following we would like to apply the general relations we have derived to a specific case of a biphoton generated in the process of spontaneous parametric down conversion (SPDC). In SPDC, the phase-matching conditions result in $\omega_0=\omega_p/2$, where ω_p is the frequency of the pump radiation and ω_0 is the degenerate frequency of the signal and idler photons. We will consider collinear type-I SPDC because, as we will see below, it is this case that can yield the shortest $\tau^{(2)}$. The spectrum of SPDC radiation is limited from above by ω_p, therefore even in transparent media $\tilde{\tau}^{(2)}_{min} = \pi/2\omega_p$. This means that it is impossible to mutually localize two photons of a biphoton better than it is classically possible for the pump photon. This limit is the same as the limit obtained for transverse correlation function of the biphoton [1–4].

For collinear SPDC, we have

$$h(\nu) = \frac{1-e^{iL\Delta_k(\nu)}}{iL\Delta_k(\nu)}, \tag{8}$$

$$\Delta_k(\nu) = k_p - k_s(\omega_0+\nu) - k_i(\omega_0-\nu), \tag{9}$$

where L is the SPDC crystal length, $\Delta_k(\nu)$ is the wave detuning between the pump and the signal and idler components (labeled "s" and "i", respectively), and ν is the frequency detuning from the central frequency ω_0. The spectral density of these components is peaked where the phase-matching condition $\Delta_k(\nu)=0$ is satisfied, which usually occurs in the vicinity of a unique pair of frequencies ω_1 and ω_2 such that $\omega_1+\omega_2=\omega_p=2\omega_0$. If a nondegenerate, $\omega_1\neq\omega_2$, two-photon state is sent through an interferometer with varying arm lengths, one observes the quantum beat corresponding to the frequency $\omega_1-\omega_2$ [12]. The original interpretation of this experiment is interference of two biphoton amplitudes. Alternatively, one can say that the interferometer prepares a nondegenerate two-photon state with the "symmetrized" $h(\nu)$, which results in fringed $G^{(1)}$ and $G^{(2)}$, as predicted in Eqs. (6) and (7). Of course, the two explanations are equivalent.

Quasiperiodic functions such as (7) are not always practical for measurement, as they give the result modulo the period. They are an even worse option for gathering two photons at the same spatial location (e.g., for two-photon lithography or fluorescence microscopy), since the joint detection probability is normalized with respect to the entire envelope which contains many fringes. Therefore it is important to study questions (i) and (ii) for the degenerate case when the spectrum $S(\omega)$ has a single maximum at ω_0. It is clear from relations (3) and (4), that both $G^{(2)}(t)$ and $G^{(1)}(t)$ have the narrowest widths when the power spectrum $|h(\nu)|^2$ is the broadest one. The broadest function that can be defined on a given transparency window Ω is rectangular, in which case

$$G^{(1)}(t) = -\frac{1}{\Omega}\mathrm{sinc}\left(\frac{\Omega t}{2}\right), \quad G^{(2)}(t) = \mathrm{sinc}^2\left(\frac{\Omega t}{2}\right). \tag{10}$$

It is interesting to point out that the results (10) can also be derived from (6) and (7) if the two rectangular areas are allowed to merge by setting $\delta\omega=\Omega/2$. In the following we will refer to this example as the limiting case, since it provides the smallest widths possible for both correlation functions. Redefining a correlation time as half of the full width at half maximum (FWHM) of the corresponding correlation function, we find that the fundamental limitation on the two-photon correlation time is $\tau^{(2)}\approx 2.78/\Omega$. Note that this time is only slightly shorter than the autocorrelation time $\tau^{(1)} \approx 3.78/\Omega$ evaluated for the same limiting case.

An important question is, how closely can we approach the limiting case with realistic SPDC sources. We rewrite Eq. (9) in the following form:

$$\Delta_k(\nu) = k_p - \frac{\omega_0+\nu}{c}n(\omega_0+\nu) - \frac{\omega_0-\nu}{c}n(\omega_0-\nu), \tag{11}$$

where n is the refraction index which can be expanded in series with a parameter $\xi\equiv\nu/\omega_0<1$,

$$n(\omega_0+\nu) = n[\omega_0(1+\xi)] = \sum_{j=0}^{\infty} d_j\xi^j. \tag{12}$$

Substituting the series (12) into (11) and grouping the terms by powers of ξ, we arrive at

$$\Delta_k(\nu) = -2\frac{\omega_0}{c}\sum_{j=1}^{\infty}(d_{2j-1}+d_{2j})\xi^{2j} \equiv -\sum_{j=1}^{\infty}\mu_j\xi^{2j}. \tag{13}$$

The zeroth-order term dropped out because we defined $\nu=0$ as the perfect phase-matching condition; the odd powers of ξ dropped out because we chose the degenerate type-I SPDC where $n_s(\omega)=n_i(\omega)$; for the same reason we dropped the indexes "s" and "i" in Eqs. (11)–(13).

The first five coefficients of the series (13) calculated for collinear degenerate type-I SPDC in a $BeSO_4$ crystal are shown in Fig. 1 as functions of the degenerate wavelength $\lambda_0=2\pi c/\omega_0$. All these functions have zeros at different wavelengths. The first term $j=1$ of the series (13) turns to zero at the "crossover" wavelength $\lambda_0=923.8$ nm. At this particular wavelength, the lowest-order term in (13) is ξ^4, which is a better approximation of the limiting case than the ξ^2. The inset of Fig. 1 represents the wave detuning $\Delta_k(\nu)$ versus relative frequency detuning ξ, calculated directly from Eq. (11) for the degenerate wavelengths of 825, 923.8, and 1000 nm. Collinear phase matching for these wavelengths is

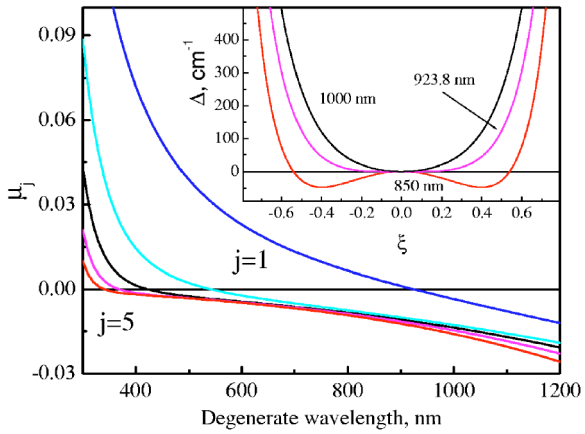

FIG. 1. The first five coefficients of the expansion (13) as functions of the degenerate wavelength $\lambda_0 = 2\pi c/\omega_0$ calculated for the n_o in $BeSO_4$ by differentiating the Sellmeier equation. Inset: The wave detuning $\Delta_k(\nu)$ vs relative frequency detuning ξ for a 5-mm-long crystal.

achieved when the angle θ_a between the optical beams and the crystal's optical axis is 43.2°, 42.0°, and 41.6°, respectively.

Coefficients μ_j have the same qualitative behavior for a number of nonlinear crystals. We chose $BeSO_4$ as an example because its crossover wavelength is conveniently near the peak of sensitivity of commercial photon counting detectors, and therefore this might be the optimal SPDC source for the two-photon correlation measurements with enhanced temporal resolution. To estimate this resolution we substitute $\Delta(\nu,\lambda_0)$ into the spectral density for the crossover $\lambda_0 = 923.8$ nm and note (see Fig. 2) that the result closely approximates a rectangle. Therefore we can evaluate the correlation time using the result obtained for the limiting case considered above: $\tau^{(2)} \approx 2.78/\Omega$. From Fig. 2 we find that in our case $\Omega \approx 1.0 \times 10^{15}$ s^{-1} for the 5-mm sample and 1.7×10^{15} s^{-1} for the 0.5-mm sample. This yields the correlation time $\tau^{(2)} \approx 2.8$ and 1.6 fs, accordingly. For shorter crystals, the spectrum is broader. Eventually, for a very short crystal, the material dispersion is not going to play any significant role and the spectral width will be determined by the material absorption. The $BeSO_4$ has a transparency window [13] between 170 and 1580 nm, which gives us the ultimate correlation time of $\tau^{(2)} \approx 0.3$ fs. An ultimate example of a two-photon source whose small size makes dispersion unimportant (so the two-photon correlation time is solely determined by the spectral width of the emitted light) is a single two-level atom. Studies of this system have shown [14,15] that the spectral width is determined by the energy (frequency) of the atomic transition and that the two-photon spectrum has nearly a rectangular shape closely matching the limiting case.

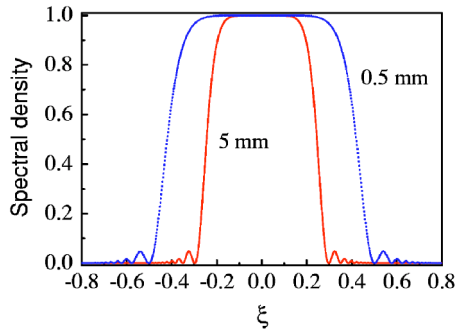

FIG. 2. Power spectrum of SPDC light vs frequency detuning for a 5-mm (inner curve) and 0.5-mm (outer curve) $BeSO_4$ crystal at the crossover degenerate wavelength 923.8 nm.

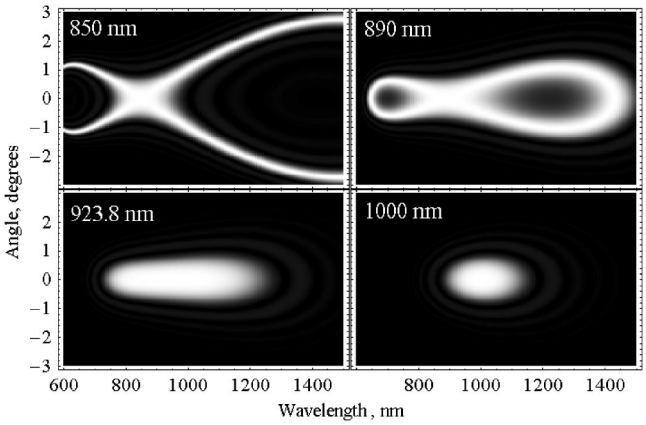

FIG. 3. SPDC spectra of a 5-mm $BeSO_4$ crystal for different degenerate wavelengths. As expected, the "crossover" wavelength $\lambda_0 = 923.8$ nm corresponds to the broadest spectrum of SPDC light irradiated in the forward (collinear with the pump) direction.

SPDC spectra calculated for a 5-mm $BeSO_4$ crystal are shown in Fig. 3 for four different degenerate wavelengths (and the crystal axis orientations allowing collinear solution in each case), including the crossover wavelength. In these calculations, we used the "exact" material dispersion relations as per [13] rather than their series expansions. We see that with changing the sign of μ_1 from positive to negative, the noncollinear solution disappears and only the collinear one remains. The physics of this phenomenon will be discussed elsewhere. At the crossover wavelength the SPDC becomes a broadband source of collimated two-photon light, whose spectrum can easily cover an octave. If one chooses to depart from the essentially quantum regime of spontaneous down conversion, our system can be considered as a parametric amplifier whose normalized gain curve (for the collinear direction) is represented in Fig. 2. Its central wavelength depends on the choice of the crystal; for example, the commonly used BBO crystal has a crossover wavelength of 1431.4 nm, so the flat region of its gain curve covers both optical communication bands, 1.3 and 1.5 μm. As a next step from a parametric amplifier, one can consider building a superbroadband optical parametric oscillator (OPO). Continuous-wave-pumped OPOs with noncollinear configuration have been shown to operate in the mode-locked regime [16]. Implementing a similar technique in our case, one should be able to generate the self-referenced optical combs [17].

Returning to the quantum aspects of the discussed SPDC source, we would like to point out that at a wavelength slightly shorter than the crossover wavelength (see the upper-right quarter of Fig. 3) a peculiar "double" collinear phase

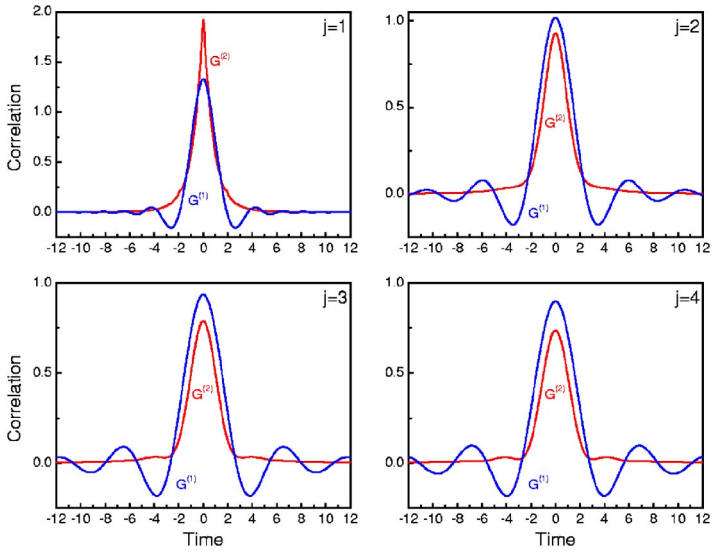

FIG. 4. $G^{(1)}(t)$ and $G^{(2)}(t)$ calculated for individual terms of the series (13) $j=1$ through 4. While the time scales for the plots are different, we see that on each plot the ratio between the correlation functions' widths remains approximately the same.

matching becomes possible. In this case, the entangled states of the form $|1\rangle_{\lambda_0}|1\rangle_{\lambda_0}+|1\rangle_{\lambda^+}|1\rangle_{\lambda^-}$ will be emitted collinearly with the pump. These states are analogous to polarization entangled states arising in type-II SPDC and may be of interest in quantum interferometry, especially in fiber-optic and other applications where frequency encoding of information is more robust than polarization encoding.

A rectangular power spectrum is generally unachievable. To understand how the answer to question (ii) changes for the degenerate SPDC case occurring in a real crystal, we consider a hypothetical $\Delta_k(\xi)$ depending on a single term of the series (13), proportional to an even power of ξ. For this model, we can carry out simple numerical calculations, with results shown in Fig. 4 for $j=1$, 2, 3, and 4 (i.e., the second, fourth, sixth, and eighth powers of ξ). The first case describes the collinear degenerate SPDC of type-I far away (and at the blue side) from the crossover wavelength. The second case was already discussed above, and was shown to be a reasonably good approximation of the limiting case. Higher-order cases approximate it even better. We therefore see that in all these examples the relation between the $G^{(1)}\times(t)$ and $G^{(2)}(t)$ widths is almost universal. Therefore we conclude that, in a realistic case when all power terms are present, this relation holds as well: $G^{(2)}(t)$ is only incrementally narrower than $G^{(1)}(t)$.

To conclude, we have carried out a theoretical study of the limitations imposed on the two-photon correlation time of a biphoton, and demonstrated that for the optimal case of strongly nondegenerate SPDC this time is determined by the single period of the optical pump oscillation. In all realistic cases of degenerate SPDC this time is only slightly shorter than the inverse spectral width of an individual signal or idler photon. The strongest possible temporal correlation of a photon pair, desired for a variety of two-photon applications, is ultimately limited by the inverse of the transparency window of the nonlinear optical material generating the biphotons. This ultimate case can only be achieved in the limit of zero interaction length, that is for a single-atom system. For crystals, the limitation is more severe and is determined by the material dispersion rather than by the transparency window. We have found a solution eliminating the wave detuning dispersion up to the fourth order of the frequency detuning. As a result, very short (of the order of a femtosecond) two-photon correlation times can be achieved, providing access to a femtosecond time domain with the simple technical means of a photon-correlation technique.

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