Limitation on two-photon temporal correlation

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ABSTRACT

We show theoretically that two-photon correlation time for a biphoton, a pair of entangled photons generated in the process of parametric down conversion, is limited by the inverse frequency width of the single-photon spectrum, which is limited by the dispersion of the nonlinear material used for the pair generation. The dispersion is determined by the transparency window of the material, which provides the ultimate limitation for the two-photon correlation time. We discuss several experimental approaches enabling realization of such a limiting case.

Keywords: Entanglement, Biphoton, Parametric Down Conversion, Correlation Function

1. INTRODUCTION

Quantum correlation between photons, or entanglement, is in the heart of quantum optics. It is this property that enables quantum communications,[1–3] cryptography,[4,5] and computing.[6–8] Sometimes, however, classical optics proposes robust techniques able to compete with quantum ones. One of the best examples here is quantum lithography.[9,10] The idea of quantum lithography is based on curious nonclassical feature of an entangled photon pair, or a biphoton.[11] Spatial characteristics of a biphoton important for the lithography can be described with the biphoton amplitude ("photonic de Broglie wavelength"[12]), having twice as short wavelength than each individual entangled photon. Therefore, biphotons could generate diffraction patterns with step twice shorter than the step of similar patterns generated with uncorrelated photons, promising serious benefits to the lithographic methods. To understand advantages as well as limitations of the quantum methods compared with similar classical techniques, the fundamental restrictions that are imposed on the quantum correlations should be realized.

In this paper, we discuss fundamental limitations for longitudinal correlation of a biphoton. Those limitations are particularly important for quantum metrology[13,14] as well as quantum lithography.[9,15] It was shown, for instance, that the group and phase dispersion as well as the polarization mode dispersion of a material can be measured with subfemtosecond precision using biphotons.[14,16–18] The measurement sensitivity in these types of measurements is given by the longitudinal correlation of the biphoton. Likewise, the choice of the material that could be used in quantum lithography depends on the longitudinal correlation of the biphotons.

We show that biphoton longitudinal correlation, that can be characterized with the correlation time of the two entangled photons, is absolutely limited by the inverse spectral width of the individual photon. This width is ultimately determined by the transparency window of the nonlinear material used for the generation of the photon pair. We discuss several experimental approaches enabling realization of such a limiting case. First, we study the approach based on usage of perfectly transparent nonlinear optical crystals (e.g. BBO) for the parametric down-conversion. We consider Type-I degenerate parametric down conversion in collinear and non-collinear configurations and show that the two-photon correlation time is dispersion limited, for macroscopic crystals. Next, we consider microscopic systems, ultimately a single atom, capable of generating photon pairs via two-photon decay, and show that the two-photon correlation time is limited by an optical cycle, due to material absorption.

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2. BASIC DEFINITIONS AND RELATIONS

Consider two optical fields with given polarization, $E_1(t)$ and $E_2(t)$, observed at the same spatial location $\vec{r}$. According to Glauber,[19,20] the fields can be described with the first- and the second order correlation functions

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

$$G^{(2)}(\tau) \equiv \langle E_1^{(-)}(t) E_2^{(-)}(t+\tau) E_1^{(+)}(t) E_2^{(+)}(t+\tau)\rangle, \tag{2}$$

where quantized electromagnetic field is presented as a sum $E(t) = E^{(+)}(t) + E^{(-)}(t)$, with the positive-frequency part of the electric field $E^{(+)}(t)$ given by the standard expression

$$E^{(+)}(t) = \int_0^\infty \sqrt{\frac{2\pi\hbar\omega}{\mathcal{A}c}}\; a_\omega e^{-i\omega t} \frac{d\omega}{2\pi}\;, \tag{3}$$

$\mathcal{A}$ is the effective cross sectional area of the beam and a_ω is the annihilation operator, whose commutation relations are

$$[a_\omega, a_{\omega'}] = 0\;, \quad [a_\omega, a_{\omega'}^\dagger] = 2\pi\delta(\omega - \omega')\;, \tag{4}$$

$\langle \ldots \rangle$ stands for the ensemble averaging.

The first-order correlation function becomes an auto-correlation function if $E_1(t) = E_2(t)$, i.e. $E_1(t)$ and $E_2(t)$ are the fields of the same optical mode. It is well known that the width of $\mathrm{G}^{(1)}(t)$ is limited and cannot be shorter than the optical wavelength. A simplest way to see it is by recalling that $\mathrm{G}^{(1)}(t)$ is given by Fourier transform of the optical field spectral density (Wiener-Khintchine theorem), which cannot extend beyond zero frequency. As a result, it is impossible to generate an optical pulse shorter than its central wavelength. More formally, we can write

$$G^{(1)}(t) = \int_{-\infty}^{\infty} d\omega S(\omega) e^{-i\omega t}, \tag{5}$$

where $S(\omega)$ is the power spectrum of the field. Because usually $S(\omega)$ has a finite frequency width, $G^{(1)}(t)$ has the duration limited by the inverse spectral width.

The above argument cannot be directly applied to the $\mathrm{G}^{(2)}(t)$, whether it pertains to the fields in the same or in different optical modes. However, it is $G^{(2)}(t)$, not $G^{(1)}(t)$, characterizes a biphoton (however, it is $G^{(1)}(t)$, not $G^{(2)}(t)$, that is often observed in two-photon correlation experiments[21]). The questions then arise, what is the limitation on the correlation time of an entangled photon pair? Can this time be shorter than the inverse spectral width? Ultimately, can it be shorter, or perhaps much shorter, than the optical cycle of the photon field? Mathematically, all these questions are the questions about the properties of the function $G^{(2)}(t)$, particularly about its minimum possible width. Let us discuss this question in application to the most commonly used source of entangled photon pairs: spontaneous parametric down conversion (SPDC).

For a selected pair of transverse modes satisfying the phase matching condition at degenerate frequency $\omega_0 = \omega_p/2$, the two-photon state generated in SPDC with no absorption is given as (see, e.g.,[11])

$$|\Psi\rangle = \int_{-\infty}^{\infty} d\nu h(L\Delta(\nu)) a_s^\dagger(\omega_0 + \nu) a_i^\dagger(\omega_0 - \nu)|0\rangle, \tag{6}$$

where the subscripts label the signal and idler photons, ν is the frequency detuning from the exact phase matching, L is the SPDC crystal length,

$$h(L\Delta(\nu)) = h(\nu) = \frac{1 - e^{iL\Delta(\nu)}}{iL\Delta(\nu)}, \tag{7}$$

and

$$\Delta(\nu) = k_p - k_s(\omega_0 + \nu) - k_i(\omega_0 - \nu) \tag{8}$$

is the wave detuning. Usually, Eq.(8) is expanded in series, where ν is treated as a small parameter, and only the leading term is retained. For example,[22] for the Type-II and non-degenerate Type-I SPDC

$$\Delta(\nu) \approx \left(\frac{1}{u_s} - \frac{1}{u_i}\right)\nu, \tag{9}$$

while for the degenerate Type-I SPDC this term turns to zero, and

$$\Delta(\nu) \approx \left(\frac{d}{d\omega}\frac{1}{u}\right)_{\omega_0}\nu^2. \tag{10}$$

In Eqs.(9) and (10) $u_{s,i} = d\omega_{s,i}/dk_{s,i}$ is the group velocity of the signal or idler light.

To find the $G^{(1)}(t)$ for, e.g., the signal field, we introduce the density operator for this filed, $\hat{\rho}_s = tr_i[\hat{\rho}]$, where $\hat{\rho} = |\Psi\rangle\langle\Psi|$, and find[23] that it represents a diagonal mixed state, i.e.

$$G^{(1)}(t) = tr[\hat{\rho}_s a^\dagger(\nu)e^{i\nu t_1}a(\nu)e^{-i\nu t_2}] = \int_{-\infty}^{\infty} d\nu S(\nu)e^{-i\nu t}, \tag{11}$$

with spectral density determined by

$$S(\nu) = |h(\nu)|^2 = \frac{1}{2}\mathrm{sinc}^2\left[\frac{L\Delta(\nu)}{2}\right]. \tag{12}$$

It is easy to show,[11] that for the state described with (6),

$$G^{(2)}(t) = |\langle 0|E_s^{(+)}(t_1)E_i^{(+)}(t_2)|\Psi\rangle|^2 = \left|\int_{-\infty}^{\infty} d\nu h(\nu)e^{-i\nu t}\right|^2, \tag{13}$$

where $t = t_2 - t_1$.

Comparing Eqs. (13) and (11) we see that the correlation functions $\mathrm{G}^{(1)}(t)$ and $\mathrm{G}^{(2)}(t)$ both are defined via $h(\nu)$ and, hence, are closely related to each other.[21] To make this relation more transparent, we introduce

$$f(t) \equiv \int_{-\infty}^{\infty} d\nu h(\nu)e^{-i\nu t}, \tag{14}$$

and, applying the convolution theorem, arrive at the following time-domain relations:

$$G^{(1)}(t) = \int_{-\infty}^{\infty} f(t')f^*(t'-t)\,dt', \tag{15}$$

$$G^{(2)}(t) = |f(t)|^2. \tag{16}$$

In the following Section, we study the correlation times determined by $\mathrm{G}^{(1)}(t)$ and $\mathrm{G}^{(2)}(t)$ for various systems.

3. TWO-PHOTON CORRELATION TIME

Using Eqs. (15) and (16) we can compare the autocorrelation time and two-photon correlation time for several particular cases. For example, applying relations (15) to Gaussian function

$$f(t) = e^{-\Omega^2 t^2}, \tag{17}$$

we see that $G^{(2)}(t) = \exp(-2\Omega^2 t^2)$ is twice narrower than $G^{(1)}(t) \propto \exp(-\Omega^2 t^2/2)/\Omega$. On the other hand, making $f(t)$ rectangular (such as in the Type-II SPDC), we obtain both correlation functions of the same full width at half maximum (FWHM).

From Eqs.(15) and (16), it is clear that for the narrowest $G^{(2)}(t)$, $G^{(1)}(t)$ is also the narrowest. The latter condition is achieved when the spectrum $|h(\nu)|^2$ of $G^{(1)}(t)$ (c.f. Eq.(11)) is maximally broad, i.e. has a rectangular shape covering the entire transparency window Ω:

$$h(\nu) = \begin{cases} 1/\Omega, & -\Omega/2 \leq \nu \leq \Omega/2, \\ 0, & \text{elsewhere.} \end{cases} \tag{18}$$

Substituting this $h(\nu)$ into Eqs. (13) and (11), we find

$$G^{(1)}(t) = \frac{1}{\Omega}\mathrm{sinc}\left(\frac{\Omega t}{2}\right), \tag{19}$$

$$G^{(2)}(t) = \mathrm{sinc}^2\left(\frac{\Omega t}{2}\right). \tag{20}$$

In this case $G^{(2)}(t)$ is only slightly narrower than $G^{(1)}(t)$.

We have just shown that the smallest achievable width of the two-photon correlation function $G^{(2)}(t)$ is close to the width of the $G^{(1)}(t)$, or the inverse spectral width of light π/Ω. Let us now ask a different question: in principle, how much narrower can $G^{(2)}(t)$ be compared with $G^{(1)}(t)$? This question may be important for optical applications requiring precision ranging or timing, but precluding using broadband light due to, e.g., strong dispersion. Another important question is, how closely can we approach the discussed ideal case with realistic Type-I SPDC sources.

3.1. Type-I SPDC: collinear case

Let us start by re-writing equation (8) in the following form:

$$\Delta(\nu) = k_p - \frac{\omega_0 + \nu}{c} n(\omega_0 + \nu) - \frac{\omega_0 - \nu}{c} n(\omega_0 - \nu). \tag{21}$$

Here n is the refraction index. We dropped the "signal" and "idler" label because we will be interested in the degenerate Type-I SPDC. We can expand Eq. (21) in series with a parameter $\xi \equiv \nu/\omega_0 < 1$:

$$n(\omega_0 + \nu) = n(\omega_0(1 + \xi)) = \sum_{j=0}^{\infty} d_j \xi^j, \tag{22}$$

and group the terms by powers of ξ, arriving at

$$\Delta(\nu) = -2\frac{\omega_0}{c} \sum_{j=1}^{\infty} (d_{2j-1} + d_{2j}) \xi^{2j} \equiv \sum_{j=1}^{\infty} \mu_j \xi^{2j}. \tag{23}$$

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 Type-I SPDC where $n_s(\omega) = n_i(\omega)$.

The first five coefficients of the series (23) calculated for collinear degenerate Type-I SPDC in a BBO crystal are shown in the left part of Fig.1 as functions of the degenerate wavelength $\lambda_0 = 2\pi c/\omega_0$. All these functions have roots at different wavelengths. Lower order terms turn to zero at longer wavelengths. The first term $j = 1$ of the series (23) turns to zero at $\lambda_0 = 1431.4$ nm. At this particular wavelength, the lowest order term in (23) is ξ^4, which is a better approximation of the ultimate case of rectangular spectrum than the ξ^2. The right part of Fig.1 represents the wave detuning $\Delta(\nu)$ vs. relative frequency detuning ξ, calculated directly from Eq.(21) for the degenerate wavelengths of 1000, 1200, 1431.4, 1600 and 1800 nm. Collinear phase matching for these wavelengths is achieved when the angle θ_a between the optical beams and the crystal's optical axis is 23.9, 21.2, 20.0, 19.9 and 20.4 degrees, respectively.

Substituting $\Delta(\nu, \lambda_0)$ into $h(\nu)$ we see that for the "critical" 1431.4 nm wavelength, the spectral distribution of SPDC light closely approximates a rectangle, and the above result for the width of $G^{(2)}(t)$ being approximately

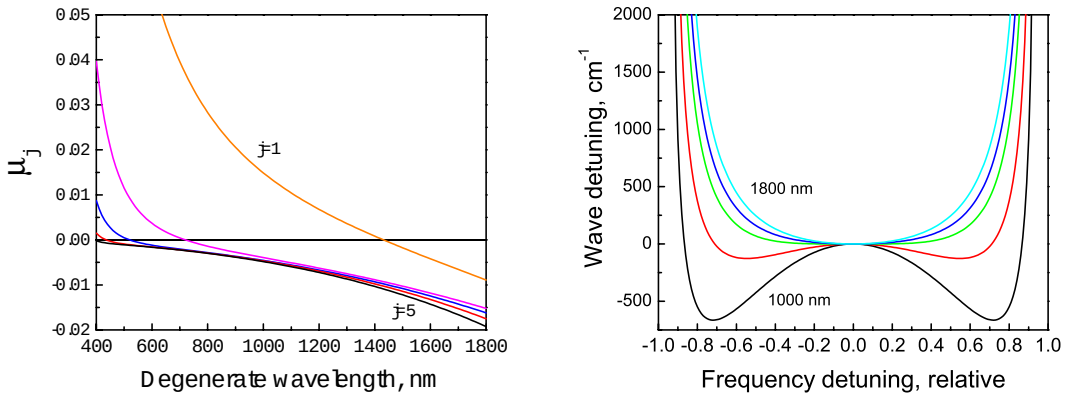

Figure 1. Left: The first five coefficients of the expansion (23) as functions of the degenerate wavelength $\lambda_0 = 2\pi c/\omega_0$ calculated for the n_o in BBO by differentiating the Sellmeier equation. Lower order terms have larger positive values. **Right:** The wave detuning $\Delta(\nu)$ vs. relative frequency detuning ξ for a 5 mm - long BBO crystal is shown for the degenerate wavelengths of 1000 nm (the lower curve), 1200 nm, 1431.4 nm, 1600 nm and 1800 nm (the upper curve).

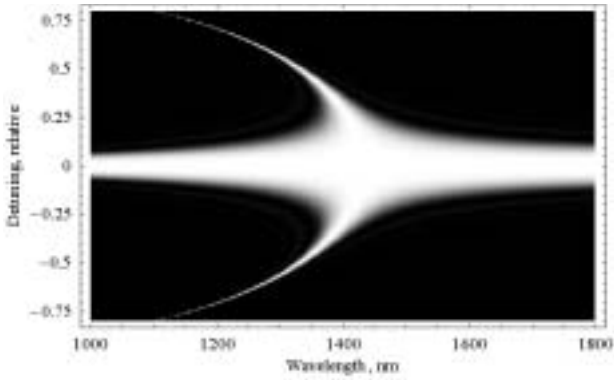

Figure 2. Spectral distribution of SPDC light in units of $\xi = \nu/\omega_0$ vs. wavelength. Changing the wavelength implies changing of the phase matching angle to provide for the collinear degenerate solution at this wavelength.

equal to the inverse spectral width of light applies. Taking this into account, we see that the FWHM of both correlation functions for a 5 mm - long BBO crystal is determined by the condition $\Delta(\nu) \approx 3\,\mathrm{cm}^{-1}$. From Fig.2 we find that his condition is attained at $\xi \approx \pm 0.24$, which yields, given the wavelength, the correlation time of approximately 20 fs. This time gets shorter for a shorter crystal, e.g. for $L = 0.5$ mm it becomes 12 fs, and so on. However, we see that the correlation time saturates for small L, and is limited by the material transparency window.

It is instructive to plot the spectral distributions for different crystal orientations, corresponding to collinear degenerate phase matching at different wavelengths around 1431.4 nm, see Fig.2. From the plot we notice that the spectral distribution branches at shorter wavelengths. This is consistent with the right part of Fig.1, where we see that the equation $\Delta(\nu, \lambda_0) = 0$ has three solutions in the short wavelength range, that all become a degenerate $\nu = 0$ solution at the "critical" $\lambda_0 = 1431.4$ nm due to disappearing of the leading term μ_1 at this wavelength. At longer wavelengths, there is only one solution $\nu = 0$.

Let us point out that, with changing the sign of μ_1 from positive to negative, the non-collinear solution to the phase matching equations disappears, and only the collinear solution remains, see Fig.3. At that point SPDC

becomes a broadband source of collimated two-photon light. Moreover, at the degenerate wavelength slightly shorter than the critical wavelength, "double" phase matching becomes possible, which results in emission of the entangled states of the form $|1\rangle_{\lambda_0}|1\rangle_{\lambda_0} + |1\rangle_{\lambda^+}|1\rangle_{\lambda^-}$ 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 optical and other applications where frequency encoding of information is more robust than polarization encoding.

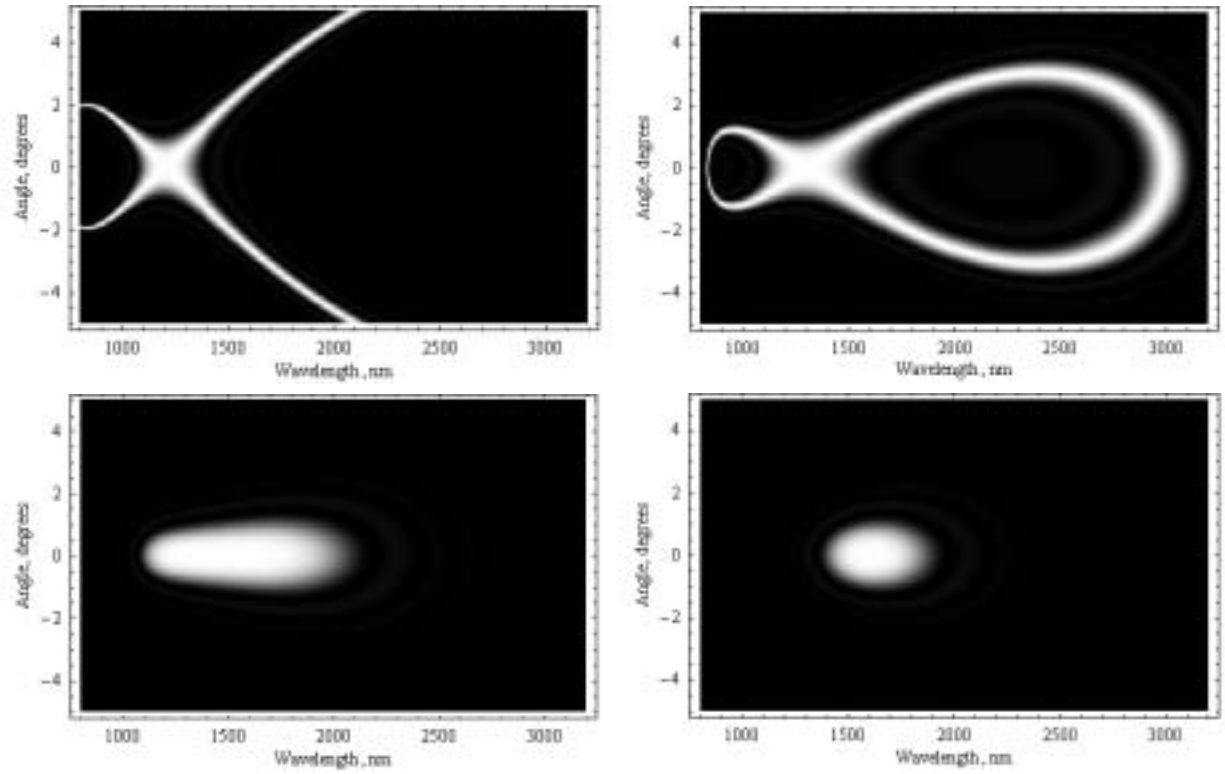

Figure 3. Tuning curves for degenerate collinear wavelengths (equal to twice the pump wavelength) of 1200 nm (upper left); 1300 nm (upper right); 1431.4 nm (lower left); and 1600 nm (lower right). As expected, the "critical" case $\lambda_0 = 1431.4$ nm corresponds to the broadest spectrum of SPDC light irradiated in the collinear with the pump direction.

To address the question of the maximum possible distinction of the $G^{(1)}(t)$ and $G^{(2)}(t)$ widths, we consider a hypothetical case of $\Delta(\xi)$ depending on a single term of the series (23), proportional to an arbitrary even power of ξ. This approach allows us to carry out simple numerical calculations, whose result is shown in Fig.4 for $j = 1, 2, 3$ and 4 (i.e., the second, forth, sixth and eight powers of ξ). The first case describes collinear degenerate SPDC of the Type-I far blue from the critical wavelength. The second case we have just considered and shown that it is already a reasonably good approximation of the rectangular spectrum. Higher order cases approximate such spectrum even better. We see, that in all these examples the relation between the $G^{(1)}(t)$ 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 slightly narrower than $G^{(1)}(t)$.

3.2. Type-I SPDC: non-collinear case

Let us consider degenerate but non-collinear Type-I SPDC. As before, we will be concerned only with the longitudinal phase matching, assuming the transverse phase matching perfectly satisfied for the selected pair of transverse modes. A transverse mode in free space is conventionally defined as a single Fresnel zone. In this

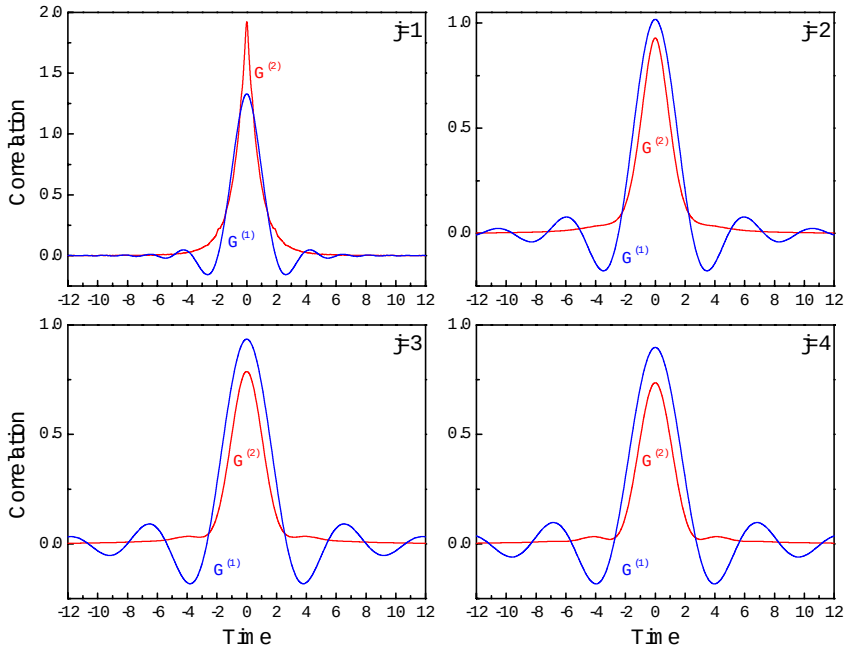

Figure 4. $G^{(1)}(t)$ and $G^{(2)}(t)$ calculated for individual terms of the series (23) $j = 1$ through 4. While the time scales for the plots are different, we see that on each plot the ratio between the correlation functions widths remains approximately the same.

case, the wave detuning $\Delta(\nu)$ (21) takes on the following form:

$$\Delta(\nu) = k_p - \frac{\omega_0 + \nu}{c} n(\omega_0 + \nu) \cos[\theta_0 + \delta\theta(\nu)] - \frac{\omega_0 - \nu}{c} n(\omega_0 - \nu) \cos[\theta_0 + \delta\theta(-\nu)], \tag{24}$$

where θ_0 is the perfect phase matching angle and the variation $\delta\theta$ is assumed to be smaller than a single mode angular size for the entire spectrum ν, which allows us to approximate the cosines in Eq.(24) as

$$\cos[\theta_0 + \delta\theta(\nu)] \approx \cos\theta_0 - \delta\theta(\nu) \sin\theta_0 \tag{25}$$

and to notice that the angular variation term $\delta\theta$ disappears in the earlier considered collinear case ($\theta_0 = 0$).[24] In the non-collinear case, however, this term cannot be disregarded. Similarly to Eq.(22), we can expand it into series

$$\delta\theta(\nu) = \sum_{m=1}^{\infty} B_m \xi^m. \tag{26}$$

Substituting (26) and (22) into the wave detuning (24) we find that the latter consists of two terms:

$$\Delta(\nu) = \Delta_z(\nu) \cos\theta_0 + \Delta_\theta(\nu) \sin\theta_0, \tag{27}$$

where the first term $\Delta_z(\nu)$ is already familiar to us from Eq.(23). The second term is equal to

$$\Delta_\theta(\nu) = \frac{2\omega_0}{c} \sum_{m=1}^{\infty} B_{2m} \xi^{2m} \left(n(\omega_0) + \sum_{j=1}^{\infty} \mu_j \xi^{2j} \right) + \frac{2\omega_0}{c} \sum_{m=1}^{\infty} B_{2m-1} \xi^{2m} \sum_{j=1}^{\infty} (d_{2j} + d_{2j+1}) \xi^{2j}. \tag{28}$$

The purpose of this analysis is to see if using the non-collinear case could allow us to cancel the leading term ξ^2 not only at 1431.4 nm, but at any desired wavelength. To verify this, we gather all terms proportional to ξ^2 in Eq.(27) and solve the equation for their sum equal to zero, with respect to the signal wavelength and the crystal axis orientation. The solution will be limited to only the wavelength-degenerate case. This solution is shown on the left side of Fig.5. It includes the collinear case for $\theta_a = 20^o$ and $\lambda_0 = 1431.4$ nm as well as a range of other solutions. Upon close consideration, however, we notice that all of these solutions represent *nearly* collinear SPDC. At nearly-collinear orientation, the tuning curves $\theta(\lambda)$ for the Type-I SPDC are known to depend very strongly on the frequency detuning, experiencing a discontinuity at the exact collinear orientation, where their slope turns to infinity. Therefore it is not surprising that the quadratic terms always cancel for the nearly-collinear geometry; it is unfortunate that they do not cancel anywhere else. An example of the tuning curve for nearly collinear SPDC is shown on the right of Fig.5. From this curve we see that indeed, the spectral width of the degenerate wavelength signal is somewhat larger than for other wavelengths, but is far from the "critical" case in Fig.3. One possible reason for this is that near the collinear orientation, even though the ξ^2 term disappears, the coefficients of the higher-order terms in $\Delta_\theta(\nu)$ become very large.

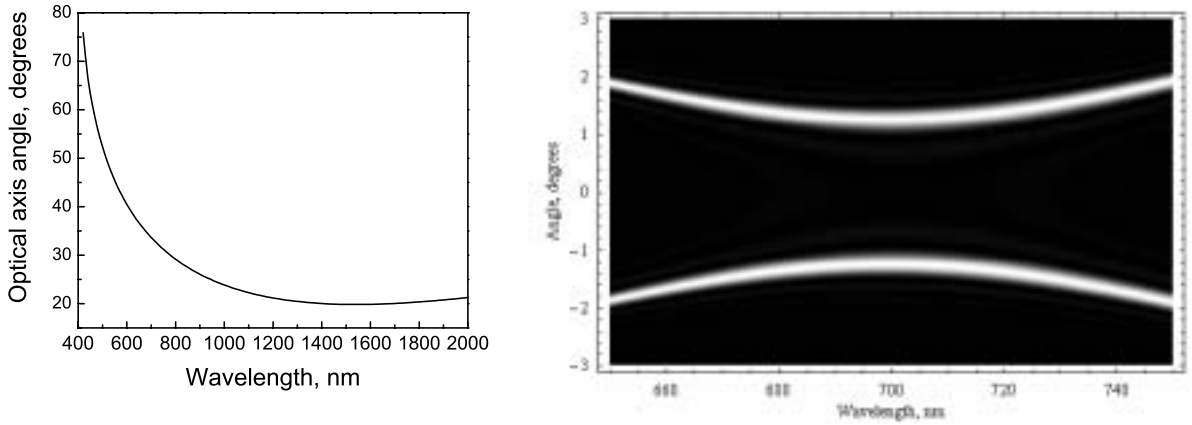

Figure 5. Left: The optical axis orientation θ_a and the degenerate signal wavelength λ_0 zeroing the ξ^2 term in Eq.(27). **Right:** A phase matching curve for the Type-I SPDC degenerate at 702.2 nm. The optical axis angle $\theta_a = 33.6^o$ is very close to the perfectly collinear orientation $\theta_a = 33.53^o$.

3.3. Absorption in the crystals and SPDC

So far, discussing two-photon correlation time of the biphotons produced via SPDC, we focused on minimizing the wave detuning $\Delta(\nu)$ which arises due to dispersion of the materials. The dispersion is important for long crystals, because, according to Eq.(7), the phase-mismatch factor is proportional to the crystal length. It seems obvious that decrease of the crystal length leads to the even further reduction of the two-photon correlation time. However, this works only to a certain extent, before the absorption of the crystalline material becomes the limiting factor. Ultimately, the two-photon correlation time cannot be shorter than the spectral width of the transparency window of the nonliner material used for the biphoton generation.

Recalling Eq.(12) we see that the function $h(\nu)$ is defined on the restricted frequency interval only. This is obvious because the power spectrum $S(\nu)$ of any emitted radiation is located between zero frequency and the frequency that corresponds to the excited energy levels of the material. As we have already pointed out, the two-photon correlation time is determined by the width of the Fourier spectrum of function $h(\nu)$ (13). The shortest two-photon correlation time is realized when $h(\nu)$ is distributed over as wide as possible frequency range. This frequency range is the frequency range of the power spectrum of the radiation.

Bulk losses in continuous solid transparent materials can be approximated with the phenomenological dependence

$$\alpha \simeq \alpha_{UV}\ e^{\lambda_{UV}/\lambda} + \alpha_R\ \lambda^{-4} + \alpha_{IR}\ e^{-\lambda_{IR}/\lambda}, \tag{29}$$

where α_{UV}, α_R, and α_{IR} describe the blue wing (primary electronic), Rayleigh, and red wing (multi-phonon) losses of the light, respectively; λ_{UV} and λ_{IR} stand for the edges of the material transparency window. This expression does not take into account resonant absorption due to possible crystal impurities. Unfortunately, even the phenomenological coefficients in Eq.(29) are not always known. For BBO, the transparency range at 0.5 transmittance level for a 0.8 cm long crystal is known to be $0.198 - 2.6$ μm.[25] Therefore, the absolute value of the two-photon correlation time for a bipthoton generated in such a crystal cannot be less than ~ 0.7 fs. This result does not depend on how short the crystal length is.

Two-photon emission by a single two-level atom is an extreme example of an ultra-short nonlinear element producing biphotons. The geometric size of the atom is much less than the emitted photons wavelength so the notion of dispersion is useless for evaluation of the two-photon correlation time in this case. Only the absorption (the matrix element describing the atom parameters) is important. Studies of power spectrum of two-photon transitions in atomic medium have shown[26, 27] that the spectrum width is restricted by the energy (frequency) of the atomic transition. In this case, the two-photon spectrum has nearly rectangular shape and hence closely matches the ultimate case considered in Section 3. The two-photon correlation time cannot be shorter than the inverse width of the spectrum, i.e. the inverse of the atomic transition frequency.

4. CONCLUSION

We have carried out a theoretical study of the limitations imposed on the two-photon correlation time of a biphoton, and demonstrated that in all studied systems, this time is only slightly shorter than the inverse spectral width of an individual photon. Therefore, the strongest possible temporal correlation of a photon pair, desired for a variety of two-photon applications, is ultimately limited to 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. It has been previously shown that for Type-II and non-degenerate Type-I SPDC the leading term defining the dispersion is proportional to the frequency detuning, while for the degenerate Type-I SPDC it is proportional to the square of detuning. We have found a solution eliminating the quadratic term for the degenerate Type-I SPDC, and making the dispersion proportional to the forth power of the frequency detuning. With this simple configuration, very short (20 fs) correlation time can be achieved even with a relatively long (5 mm) crystals. Therefore, although unable to significantly surpass the classical limit, two-photon correlation technique provides an access to femtosecond time domain with much simpler technical means than e.g. ultra short laser pulses.

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