Broadening the bandwidth of entangled photons: A step towards the generation of extremely short biphotons

M. Hendrych,[1,*] Xiaojuan Shi,[1,2] A. Valencia,[1] and Juan P. Torres[1,2]
[1]*ICFO-Institut de Ciencies Fotoniques, Mediterranean Technology Park, Castelldefels, 08860 Barcelona, Spain*
[2]*Department of Signal Theory and Communications, Universitat Politecnica, Campus Nord D3, 080340 Barcelona, Spain*
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We demonstrate a technique that allows full control of the bandwidth of entangled photons independently of the frequency band of interest and of the nonlinear crystal. We theoretically show that this technique allows generating nearly transform-limited biphotons with almost one octave of bandwidth (hundreds of terahertz) which corresponds to correlation times of just a few femtoseconds. The presented method becomes an enabling tool for attosecond entangled-photons quantum optics. The technique can also be used to generate paired photons with a very high degree of entanglement.

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I. INTRODUCTION

The development of methods for the generation of entangled photon pairs (biphotons) with a specific bandwidth has been of great interest in recent years. Narrow bandwidth of frequency correlations is important for the design of efficient atom-photon interfaces [1] in quantum networks [2], for long-distance quantum communications [3], or to enable direct measurements of temporal correlations with current photodetectors [4,5].

On the other hand, some applications such as quantum optical coherence tomography [6] and nonlinear microscopy [7] require wide bandwidths. Wide bandwidths are a requisite for the generation of biphotons with very short correlation times [8] and when high fluxes of biphotons are desired [9]. A bandwidth of hundreds of terahertz can generate biphotons with a few femtoseconds of correlation time. These short temporal biphotons are of particular interest in the fields of quantum metrology [10] and for some protocols for timing and positioning measurements [11]. Thanks to the strong correlations of the entangled photons that allow remote compensation of chromatic dispersion [12], the narrow temporal correlation embedded in the biphoton can be transmitted over large distances.

Spontaneous parametric down-conversion (SPDC) is the most convenient source for the generation of entangled photon pairs. When an intense laser beam illuminates a nonlinear material, with a certain small probability a pump photon might split into two lower-energy photons. In analogy with a classical pulse, the temporal correlation width of the biphoton is determined by its spectral bandwidth and spectral phase [13]. The bandwidth of the SPDC pairs is determined by the type of phase matching (type I or type II), by the length and the dispersive properties of the nonlinear material, by the geometry of the SPDC configuration (collinear or noncollinear), and by the spectral and spatial characteristics of the pump beam. By changing any of these parameters, it is possible to modify the bandwidth and therefore the temporal correlations of the SPDC photons. For example, by choosing a particular material [periodically poled lithium niobate (PPLN)] and pumping it at a specific wavelength ($\lambda = 1885$ nm), an ultrabroad bandwidth of ~ 1080 nm has been recently reported [14]. Similarly, by using a very thin crystal of thickness 0.1 mm, a spectrum of 174 nm was produced [15]. However, from these experiments, no conclusion can be drawn about the biphoton's temporal length because the spectral correlations were not measured. In contrast, Dayan *et al.* [7] produced a bandwidth of 100 nm that yielded a biphoton with 23 fs of correlation time.

An alternative approach to modify the SPDC bandwidth is to use chirped quasi-phase-matched crystals [16]. A frequency correlation bandwidth of ~ 300 nm at a central wavelength of 812 nm has been achieved. Nevertheless, when this technique is used to obtain narrow temporal biphotons, compensation of the spectral phase is required as the biphotons are not transform limited.

In this paper, we demonstrate experimentally a technique to increase the SPDC bandwidth in order to generate ultrashort near transform-limited biphotons. It employs angular dispersion (pulse-front tilt) to modify the effective group velocity and group-velocity dispersion of the interacting waves. Differently from other approaches, the method is not based on the choice or on the engineering of the nonlinear material and it works in any frequency band and in any nonlinear crystal of interest. That offers two advantages. First, the wavelength can be chosen in the region where single-photon detectors exhibit high detection efficiency. Second, advantage can be taken of materials with high nonlinear coefficient that naturally do not provide the desired bandwidth.

Angular dispersion is a powerful enabling tool in many different areas of optics. It can be used to introduce negative group-velocity dispersion in a beam propagating in free space [17], it is a key element in many techniques for pulse compression, it enables the generation of femtosecond second-harmonic waves [18,19], and it has made possible the observation of temporal solitons [20], where the natural dispersion of the material would have made such observation impossible. In quantum optics, angular dispersion enables us to tailor the frequency correlations of entangled photons and allows us to generate frequency-uncorrelated and frequency-correlated pairs of photons [21]—quantum states that are not easily produced in most experimental configurations currently used [22,23].

*martin.hendrych@icfo.es

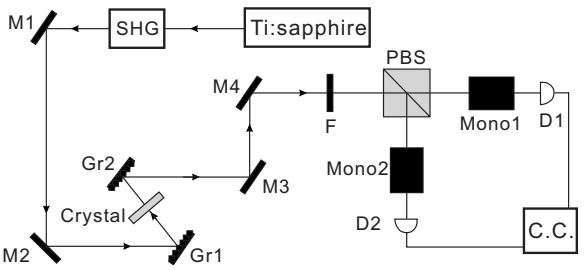

FIG. 1. Experimental setup. SHG: second-harmonic generation; M: mirrors; Gr: gratings; F: long-pass filter; PBS: polarizing beam splitter; Mono: monochromators; D: detectors; and C.C.: coincidence electronics.

II. PULSE-FRONT TILT

Let us consider collinear SPDC, where the down-converted photons copropagate along the direction of the pump beam. The state of the down-converted photons at the output of the medium may be written as

$$|\psi\rangle = \int d\mathbf{q}_s d\mathbf{q}_i d\Omega_s d\Omega_i \Phi(\Omega_s,\Omega_i,\mathbf{q}_s,\mathbf{q}_i)|\Omega_s\rangle|\Omega_i\rangle, \qquad (1)$$

where Ω_j $(j=s,i)$ denote the signal and idler photons' angular frequency detunings from the central angular frequency ω_j^0, i.e., $\omega_j=\omega_j^0+\Omega_j$, and $\mathbf{q}_j$ are the corresponding transverse wave-number vectors. The spectral and spatial properties of the down-converted photons are described by the joint spectrum [13],

$$\Phi(\Omega_s,\Omega_i,\mathbf{q}_s,\mathbf{q}_i) \propto E_\omega(\Omega_s+\Omega_i)E_q(\mathbf{q}_s+\mathbf{q}_i) \times \mathrm{sinc}\left(\frac{\Delta kL}{2}\right)\exp\left\{i\frac{s_kL}{2}\right\}, \qquad (2)$$

where E_ω and E_q are the spectral and transverse wave-number distributions of the pump beam at the input face of the nonlinear crystal of length L, respectively. $\Delta k=k_p-k_s-k_i$ is the phase mismatch along the longitudinal direction with $k_j=[(\omega_j n_j)^2/c^2-|\mathbf{q}_j|^2]^{1/2}$, and $s_k=k_p+k_s+k_i$. n_j is the index of refraction and c is the speed of light.

To illustrate the principle behind the proposed method, let us consider the scheme shown in Fig. 1. In contrast to typical collinear SPDC configurations, the nonlinear crystal is placed between two optical elements that introduce angular dispersion, e.g., a pair of prisms or diffraction gratings. Angular dispersion ϵ causes the front of the pulse to be tilted by an angle ξ given by $\tan\xi=-\lambda_0\epsilon$, where λ_0 is the central wavelength of the pulse and angular dispersion $\epsilon=m/(d\cos\beta_0)$, where m is the grating's diffraction order, d is the groove spacing, and β_0 is the output diffraction angle.

The introduction of angular dispersion modifies the spatial distribution of the pump E_q and the phase mismatch Δk of the joint spectrum of Eq. (2). If the gratings introduce opposite angular dispersion in such a way that $\tan\xi_s=-\tan\xi_p/\alpha_p$ and $\alpha_p\alpha_s=1$, it is possible to modify Δk only and to effectively modify the group velocity and the group-velocity dispersion of the pump, signal, and idler photons. Here, $\alpha_j\equiv\cos\theta_j/\cos\beta_j$ with θ_j and β_j being the incidence and diffraction angles of the pump beam at grating Gr1, respectively.

The effects of angular dispersion on the SPDC spectrum can be better understood by expanding Δk in a Taylor series about the central frequencies ω_j^0. For a quasi-continuous-wave pump, the frequency detunings of the down-converted photons must be anticorrelated in order to satisfy energy conservation, i.e., $\Omega_s=-\Omega_i$, and Δk becomes

$$\Delta k \approx (N_s'-N_i')\Omega_s - \frac{1}{2}(g_s'+g_i')\Omega_s^2+\cdots, \qquad (3)$$

where $N_j'=N_j+\tan\xi_p\tan\rho_j/c$ and $g_j'=g_j-[\tan\xi_p/c]^2/k_j^0$ play the role of effective inverse group velocity and effective group-velocity dispersion, respectively. The inverse group velocity $N_j=(dk_j/d\omega_j)_{\omega_j^0}$ and group-velocity dispersion $g_j=(d^2k_j/d\omega_j^2)_{\omega_j^0}$ are modified in the presence of the pulse-front tilt ξ_p and Poynting-vector walkoff ρ_j. The new effective values of group velocity and group-velocity dispersion depend on the angular dispersion experienced by the pump beam and allow us to control the SPDC bandwidth.

For example, let us consider type-II SPDC where the polarizations of the down-converted photons are mutually orthogonal. If tilt ξ_p is chosen such that the effective group velocities are equal $(N_s'=N_i')$, the lowest nonzero term in phase mismatch Δk is of the second order which results in an increase in the bandwidth. We obtain a type-II process where the dependence of the bandwidth on the length of the nonlinear crystal is $1/\sqrt{L}$ instead of the typical $1/L$ [13]. The value of the pulse tilt that maximizes the bandwidth is

$$\xi_{\mathrm{II}}^{\max}=\tan^{-1}\left\{\frac{c(N_i-N_s)}{\tan\rho_s-\tan\rho_i}\right\}. \qquad (4)$$

On the other hand, in a type-I process, the polarizations and the group velocities of the signal and idler photons are equal. The bandwidth is increased if the tilt is chosen such that $g_s'=g_i'=0$. In such case, the first nonzero term in Δk is of the fourth order and the dependence of the bandwidth on the length of the crystal is $1/L^{1/4}$. The tilt that maximizes the bandwidth is

$$\xi_{\mathrm{I}}^{\max}=\tan^{-1}(c^2g_sk_s^0)^{1/2}, \qquad (5)$$

where $k_j^0=k_j$ $(\omega_j^0,\mathbf{q}_s=\mathbf{q}_i=\mathbf{0})$.

III. BANDWIDTH CONTROL

To demonstrate the feasibility of the proposed technique, an experiment was performed (see Fig. 1). A 2-mm-thick type-II BBO crystal flanked with two gratings Gr1 and Gr2 was pumped by the second harmonic (Radiantis Blue Stream, 40% efficiency in the picosecond regime) of a picosecond Ti:sapphire laser (Coherent Mira 900-P) tuned at 810 nm. The down-converted photons were separated by a polarizing beam splitter and fed into monochromators Mono1 and Mono2 (Jobin Yvon MicroHR). Single-photon detectors (Perkin-Elmer SPCM-AQR-14-FC) measured single counts and coincidence electronics counted coincidence counts within a 3 ns window. The joint spectrum was obtained by

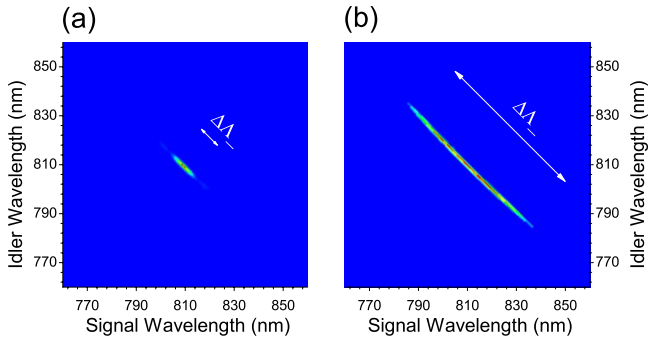

FIG. 2. (Color online) Measured joint spectral density: (a) tilt $\xi_p=0°$ and (b) tilt $\xi_p=38°$. The joint spectrum broadens sevenfold as expected.

measuring the number of coincidences while the two monochromators scanned the plane from 760 to 860 nm.

Figure 2 shows the experimental results. Figure 2(a) depicts the joint spectral density for tilt $\xi_p=0°$ when the gratings were removed. The typical frequency anticorrelation of the SPDC photons can be observed. Figure 2(b) corresponds to the situation with tilt $\xi_{II}^{max}=38°$ that maximizes the bandwidth. This was accomplished by taking grating Gr1 with a groove spacing $d=1/1200$ mm and diffraction angle $\beta_p=52°$. In order to satisfy $\tan\xi_s=-\tan\xi_p/\alpha_p$ and $\alpha_p\alpha_s=1$, grating Gr2 had $d=1/600$ mm and $\beta_s=18°$.

A more quantitative comparison between the cases with and without pulse-front tilt is plotted in Fig. 3. The graphs in the upper row correspond to the spectra of signal single counts measured after one of the monochromators: (a) without tilt and (b) with tilt $\xi_p=38°$. The spectra of idler photons are not shown for being almost identical. The squares are experimental data and the solid lines are the theoretical prediction.

Figures 3(c) and 3(d) depict the profile of the number of coincidence counts along the antidiagonal (straight line at −45°) of Fig. 2 without tilt and with tilt, respectively. The variable $\Lambda_-=(\Lambda_s-\Lambda_i)/\sqrt{2}$, where Λ_j is the detuning from the

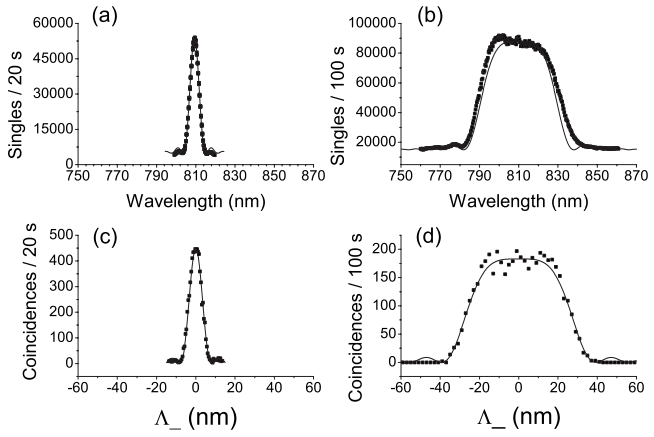

FIG. 3. (a) Signal single counts for $\xi_p=0°$; $\Delta\lambda_s=5.2$ nm. (b) Signal single counts for $\xi_p=38°$; $\Delta\lambda_s=41$ nm. (c) Coincidences along the antidiagonal for $\xi_p=0°$; $\Delta\Lambda_-=7.5$ nm. (d) Coincidences along the antidiagonal for $\xi_p=38°$; $\Delta\Lambda_-=52$ nm. Solid lines represent the theoretical prediction; squares are the experimental data.

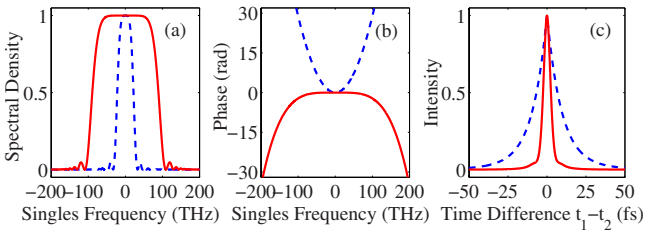

FIG. 4. (Color online) (a) Spectral density $|\Phi|^2$, (b) phase $s_kL/2$, and (c) correlation time $|\Psi(t_1-t_2)|^2$ of the biphoton for type-I BBO crystal without tilt (dashed line) and with tilt (solid line). The shrinking of the temporal correlation is clearly observed.

central wavelength, is associated with this antidiagonal. Without tilt, the full width at half maximum (FWHM) bandwidth was measured to be $\Delta\Lambda_-\sim7.5$ nm [Fig. 3(c)]. Applying a tilt $\xi_p=38°$, the bandwidth broadened to $\Delta\Lambda_-\sim52$ nm [Fig. 3(d)]. The sevenfold increase in the bandwidth is achieved without any modification of the nonlinear crystal or the working wavelength, only by introducing angular dispersion into the pump beam and the down-converted photons. It is this independence of the material properties and of the working wavelength which makes the method proposed here so promising for controlling the bandwidth of the SPDC photons.

IV. TEMPORAL CORRELATION

The application of this method for the generation of entangled photons with very narrow temporal correlations can be seen in Fig. 4. In general, temporal biphoton $\Psi(t_1,t_2)$ (t_j is the clicking time of jth detector) is given by the amplitude and phase of the joint spectrum $\Phi(\Omega_s,\Omega_i)$. Figure 4 shows the spectral density (a), spectral phase (b), and temporal shape (c) of a biphoton for the case of 2-mm-thick type-I BBO SPDC at 810 nm. Type-I phase matching was chosen because of its naturally broader spectrum. In the case without tilt, the spectral phase follows a quadratic dependence. In the case with tilt in the region where the spectral density varies, the spectral phase is almost constant due to its fourth-order dependence on the frequency. It is this fact that makes it possible to generate nearly transform-limited biphotons. The mentioned increase in bandwidth translates into ultrashort biphotons with a temporal correlation of a few femtoseconds. This contrasts with other methods where the increase in the bandwidth is not directly accompanied by a decrease in the correlation time [16,24]. That can be caused by a particular shape of the spectral phase or when the phase relationship between individual frequencies, and thus coherence, is lost. This is, for example, the case of white-light continuum generated by Kerr self-phase-modulation.

Figure 4(c) was obtained by performing a Fourier transform of Eq. (2), both numerically and analytically. The dashed and solid lines correspond to the case $\xi_p=0°$ and $\xi_p=\xi_I^{max}=16.2°$, respectively. The FWHM bandwidth of the spectral density obtained without tilt is 96 nm (2π 44 THz) that corresponds to a correlation time of 19 fs (rms width) (13.4 fs FWHM). This should be compared with the case when the optimum angular dispersion is applied that gives a

bandwidth of 465 nm (2π 197 THz) and a correlation time of 6.4 fs (rms width) (4.6 fs FWHM). It should be noted that working at a shorter wavelength, it is possible to further shorten the biphoton because the correlation time scales with $\lambda^2/(c\Delta\lambda)$.

V. ENTROPY OF ENTANGLEMENT

Pump beams with pulse-front tilt can also be employed to obtain a very high degree of entanglement. Up to now, low values of the entropy of entanglement ($E\sim 1-2$ ebits) of paired photons in the frequency continuum have been reported [25]. A higher degree of entanglement can be achieved using broadband SPDC photons. For entangled photons of the form $\Phi(\Omega_s,\Omega_i)\sim\exp\{-(\Omega_s+\Omega_i)^2/B_p^2\}\exp\{-(\Omega_s-\Omega_i)^2/B_c^2\}$, the entropy of entanglement [26] depends on the ratio between the bandwidth of the pump beam B_p (typically 2π 5 MHz) and the bandwidth of the biphoton B_c. For a bandwidth of $\Delta\lambda_s=31$ nm ($B_c\sim 2\pi$ 16.4 THz) at 1064 nm [27], one has values of $B_c/B_p\sim 3.3\times 10^6$ and $E\sim 21$ ebits. The method presented here allows to reach values of $\Delta\lambda_s>500$ nm ($B_c>2\pi$ 420 THz), therefore allowing us typical ratios greater than 8.4×10^7 and $E>26$ ebits.

Recently it has been shown [28] that signal-to-noise ratio (SNR) of detection and imaging in noisy backgrounds can be exponentially improved when entangled photons are used. The SNR increases by a factor of the order of 2^E. In our case, the number of photons needed to detect an object would be reduced by $\sim 10^7$.

VI. SUMMARY

In conclusion, a scheme to broaden the bandwidth of the SPDC photons was experimentally demonstrated. The method is based on the introduction of angular dispersion (pulse-front tilt) into the pump and down-converted photons which allowed us to demonstrate a sevenfold increase in the original bandwidth without changing the nonlinear crystal or the frequency band. The potentiality of the proposed method to generate ultrashort nearly transform-limited biphotons, and with a high degree of entanglement, has also been discussed. The described method can allow biphotons to enter the realm of *attosecond quantum optics* since it allows the generation of entangled photons with correlation times below 1 fs.

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