Detection of the Ultranarrow Temporal Correlation of Twin Beams via Sum-Frequency Generation

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We demonstrate the ultranarrow temporal correlation (6 fs full width half maximum) of twin beams generated by parametric down-conversion by using its reverse process, i.e., sum-frequency generation. The result relies on an achromatic imaging of a huge bandwidth of twin beams and on a careful control of their spatial degrees of freedom. The detrimental effects of spatial filtering and imperfect imaging are shown, along with the theoretical model used to describe the results.

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The entangled photon pairs produced by parametric down-conversion (PDC) are the key elements for several quantum communication and metrology schemes. Crucial to these applications is the ability to tailor their temporal correlation properties. In particular, various methods of generating ultrabroadband biphotons (based, e.g., on the engineering of the nonlinear medium [1] or of the pump [2]) have been recently demonstrated. Our approach relies rather on the peculiar X-shaped geometry of the spatiotemporal correlation of twin photons, theoretically outlined in Ref. [3,4], where some of us predicted the possibility of manipulating the temporal bandwidth of PDC entanglement by acting on the spatial degrees of freedom and, in particular, of achieving an unusual relative temporal localization of twin photons (for a few femtoseconds) when their near-field positions are resolved.

In this Letter, we report the experimental observation of such ultranarrow temporal correlation of twin beams (6 fs full width half maximum), detected by means of sumfrequency generation (SFG). The temporal correlation profile is measured by introducing a controlled temporal delay between twin beams and then imaging the output of the PDC crystal onto the input face of a second crystal, where up-conversion takes place.

SFG is used in classical optics as an ultrafast correlator. In quantum optics, recent experiments [5–7] used SFG to probe the temporal correlation of twin photons. Contrary to the Hong-Ou-Mandel scheme, which is insensitive to dispersion (see, e.g., the results reported in Ref. [1]), in the SFG scheme the presence of dispersive optical elements drastically broadens the measured temporal correlation of biphotons. Indeed, the experiments [5,6] used prisms to correct the dispersion introduced by optical lenses, which were effective over a large—but still limited—bandwidth of PDC. As a result, the measured temporal correlation of biphotons was in the range ≈ 30 to 100 fs.

Our setup (Fig. 1) presents a number of distinguishing features: (i) rather than correcting the temporal dispersion introduced by lenses, we implement an achromatic

imaging of the PDC light onto the SFG crystal by using parabolic mirrors; (ii) a broad spatial and temporal bandwidth of the PDC light is imaged (in phase and amplitude) onto the SFG crystal. The nonfactorability of the correlation in space and time [3,4] implies that both collecting a huge spatial bandwidth and performing a perfect spatial imaging are key elements to preserve the ultranarrow temporal localization of twin photons [8]. Indeed, our data will show that the effects of diffraction in free space are detrimental to the temporal localization



FIG. 1 (color online). (a) Experimental layout for observing the twin-beam temporal correlation. (b) Spatiotemporal spectrum of the PDC light measured by an imaging spectrometer just before the SFG crystal. Note the huge spatiotemporal bandwidths reflected by the OAP mirrors. (The color map does not reflect the real intensity; different spectral portions were detected with different attenuations.)

in a way similar to dispersion. These features allow us to preserve the phase conjugation of twin photons over a huge bandwidth (≈ 600 nm), thus enabling the demonstration of their ultranarrow temporal localization—the narrowest measured in experiments (to the best of our knowledge) using the SFG process as a probe for PDC correlation.

The experimental layout is shown in Fig. 1(a). The pump pulse at $\lambda_0 = 527.5$ nm is obtained from the second harmonic of a 1 ps, 1055 nm, 10 Hz repetition rate, Nd:glass laser (Twinkle, Light Conversion Ltd.) and is collimated down to about 0.8 mm (FWHM) at the entrance of a type I 4 mm beta barium borate (BBO) crystal for PDC in the collinear configuration. Just after the crystal, two custommade, high-reflectivity dielectric mirrors (Layertec), with reflectivity R > 99.9% in the 750–1330 nm range and $R \approx$ 0.3% at $\lambda_0 = 527.5$ nm, are used to reflect the PDC radiation. The achromatic imaging is performed by means of two identical 90° off-axis parabolic (OAP) gold mirrors, which reflect a huge portion of the spatiotemporal spectrum of the PDC radiation, as shown in Fig. 1(b), and image the output plane of the PDC crystal onto the entrance face of a second BBO crystal, identical to the first one, placed at the plane $4f_{OAP}$. Two adjacent gold mirrors M1 and M2 are placed in the far-field plane of the PDC source at $2f_{OAP}$. They act separately on the two twin-beam components of the light, because each photon has its twin on the opposite side of the far-field plane due to momentum conservation in the elementary PDC process. Mirror M_2 can be translated to produce a relative delay between twin beams. A 1.5 mm gap between the two mirrors allows us to eliminate the residual input pump. The SFG crystal is placed in the focal plane of the second OAP mirror and is mounted on a micrometer translation stage, permitting us to finely adjust its position relative to the imaging plane $4f_{OAP}$. Both crystals are also mounted on a rotation stage in order to adjust their orientation with the aim of working at exactly the same phase-matching conditions. The farfield radiation emitted by the SFG crystal is observed in the focal plane of a 20 cm focal length lens and the light intensity is monitored by means of a 16 bit scientific CCD camera (Roper Scientific) with 80% detection efficiency at 527.5 nm. The far-field SFG distribution shows a narrow central peak, representing the coherently reconstructed far-field profile of the original pump, lying over a widely spread speckled background (see Ref. [9]). The latter originates from incoherent SFG processes as well as from the residual PDC not up-converted. Note that PDC light is absent at the location of the coherent peak, because the central portion of the PDC far-field is eliminated by the gap between M_1 and M_2 .

Figure 2 reports our main result and shows the SFG peak intensity, monitored in the central pixel of the far-field distribution, as a function of the temporal delay, when the second BBO crystal is placed at the optimum imaging plane. Each data point corresponds to the peak intensity averaged over 15 images, each recorded over 2 s (20 laser



FIG. 2 (color online). Temporal correlation profile of twin beams generated by a type I BBO, reconstructed by monitoring the SFG peak intensity as a function of a temporal delay between twin beams. The solid red line shows our theoretical calculation. The dashed blue line is $\operatorname{sinc}^2(\Delta \Omega_f \Delta t/2)$.

shots). The solid line superimposed on the experimental data shows our theoretical predictions, according to the model described in the following.

The scheme is described semianalytically, exploiting the monochromatic and plane-wave pump (PWP) approximation. The model, which is valid in any gain regime of PDC (including the high-gain of the present experiment) is elaborated in detail in Ref. [8], while in the following we report only the key results.

The main quantity of interest, characterizing the entanglement of twin beams generated by PDC is the biphoton correlation (or biphoton amplitude),

$$\psi_{\rm PDC}(\Delta\vec{\xi}) = \langle A_1(\vec{\xi})A_1(\vec{\xi} + \Delta\vec{\xi}) \rangle, \tag{1}$$

where A_1 is the field operator of the down-converted signal field and $\vec{\xi} = (\vec{x}, t)$ is the three-dimensional spatiotemporal coordinate, \vec{x} denoting the transverse position at the output plane of the crystal.

In the PWP limit, the equation describing the generation of PDC light in the first crystal can be exactly solved (see Ref. [4]). In the same limit, the biphoton correlation depends only on the spatiotemporal separation $\Delta \vec{\xi}$. It can be written as the spatiotemporal Fourier transform of the probability amplitude $F_{\text{PDC}}(\vec{w})$ of generating a photon pair in the symmetric Fourier modes \vec{w} and $-\vec{w}$, where $\vec{w} = (\vec{q}, \Omega), \vec{q}$ being the transverse wave vector and Ω the offset from the central frequency $\omega_1 = \omega_0/2$,

$$\psi_{\rm PDC}(\Delta \vec{\xi}) = \int \frac{d\vec{w}}{(2\pi)^3} e^{i\vec{w}\cdot\Delta \vec{\xi}} F_{\rm PDC}(\vec{w}).$$
(2)

The probability amplitude $F_{PDC}(\vec{w})$ is strongly peaked around the curve where phase matching occurs, i.e., where

$$\Delta(\vec{w}) = l_c[k_{1z}(\vec{w}) + k_{1z}(-\vec{w}) - k_0] = 0, \qquad (3)$$

where $k_{1z}(\vec{w}) = \sqrt{k_1(\Omega)^2 - q^2}$ is the *z* component of the signal wave vector, k_0 is the pump wave number, and l_c is the crystal length. Its explicit form (derived, e.g., in

Ref. [4]) reads
$$F_{\text{PDC}}(\vec{w}) = g \frac{\sinh \Gamma(\vec{w})}{\Gamma(\vec{w})} \bigg[\cosh \Gamma(\vec{w}) + i \frac{\Delta(\vec{w})}{2\Gamma(\vec{w})} \times \sinh \Gamma(\vec{w}) \bigg]$$
, with $\Gamma(\vec{w}) = \sqrt{g^2 - \frac{[\Delta(\vec{w})]^2}{4}}$, where g is a gain parameter proportional to the pump amplitude, the non-

linear susceptibility, and l_c . For collinear phase matching, i.e., for $2k_1(0) = k_0$, the region where F_{PDC} takes its maximal value follows a characteristic hyperbolic geometry in the plane (|q|, Ω), as the spatiotemporal spectrum shown in Fig. 1(b). Indeed, a quadratic expansion of Eq. (3), valid close to degeneracy, gives

$$\Delta(\vec{q},\Omega) \approx -\frac{q^2 l_c}{k_1} + k_1'' l_c \Omega^2.$$
⁽⁴⁾

Thus, phase-matched modes are those for which the temporal dispersion occurring along the crystal is compensated by diffraction. Because of this geometry of phase matching, the spatiotemporal correlation (1) assumes an X-shape in any plane containing time and a spatial coordinate. A key result of Refs. [3,4] is that, when twin photons are detected at the same near-field position, the width of their temporal correlation, i.e., the width of $\psi_{PDC}(\Delta x = 0, \Delta t)$, is given by the inverse of the full temporal bandwidth detected (that can extend in principle to the pump optical frequency), and not by the group velocity dispersion bandwidth $1/\sqrt{k_1'' l_c}$ characterizing the far-field correlation. This is a consequence of the cancellation of temporal dispersion by diffraction occurring for all the generated modes [see Eq. (4)]. Therefore, a temporal correlation in the femtosecond range can be achieved in principle, provided that twin photons are localized in the near-field.

The SFG propagation equation in the second crystal can be solved in the limit where the fraction of up-converted light is small. Our analysis in Ref. [8] shows that at the crystal output the SFG intensity distribution is homogeneous in space and time (a mere consequence of the PWP approximation) and has two components: $I_{SFG} = I_{SFG}^{incoh} + I_{SFG}^{coh}$. The incoherent component results from the random up-conversion of photons originally unpaired. All the information on the twin-beam correlation is contained in the coherent component, which originates from the upconversion of phase conjugate photons [10], and has the form [8]

$$I_{\rm SFG}^{\rm coh} = \left| \int \frac{d\vec{w}}{(2\pi)^3} H_+(\vec{w}) H_-(-\vec{w}) F_{\rm PDC}(\vec{w}) F_{\rm SFG}(-\vec{w}) \right|^2$$
(5)

where $F_{\text{SFG}} = \sigma l'_c \operatorname{sinc} \Delta_{\text{SFG}}(\vec{w})/2 \exp i \Delta_{\text{SFG}}(\vec{w})/2$ represents the probability amplitude of up-converting a pair of photons in modes \vec{w} and $-\vec{w}$, which depends on the phase matching $\Delta_{\text{SFG}}(\vec{w})$ in the second crystal. H_+ and H_- are optical transfer functions describing the propagation between the two crystals of the twin components of the PDC light with $q_x > 0$ and $q_x < 0$, respectively, separated in the far-field by mirrors M_1 and M_2 . Equation (5) tells us that the coherent SFG component results from the sum over all

the probability amplitudes of a photon pair being generated in the first crystal in modes \vec{w} , $-\vec{w}$ times the probability amplitude the same photon pair is up-converted in the second crystal.

Assuming that a small temporal delay Δt is introduced by mirror M_2 , in the ideal case of perfect imaging with no dispersive optical elements or losses, we have $H_+(\vec{w}) = 1$, $H_-(\vec{w}) = e^{i\Omega\Delta t}$, and Eq. (5) becomes

$$I_{\rm SFG}^{\rm coh} = \left| \int \frac{d\vec{q}d\Omega}{(2\pi)^3} e^{-i\Omega\Delta t} F_{\rm PDC}(\vec{q},\Omega) F_{\rm SFG}(-\vec{q},-\Omega) \right|^2$$
(6)

$$= |[\psi_{\text{PDC}} \otimes \psi_{\text{SFG}}(\Delta \vec{x} = 0, \Delta t)|^2, \tag{7}$$

where $\psi_{\rm SFG}(\Delta \vec{\xi}) = \int d\vec{w} / (2\pi)^3 e^{i\vec{w}\cdot\Delta\vec{\xi}} F_{\rm SFG}(\vec{w})$ is the biphoton amplitude in the SFG crystal. In passing from Eq. (6) to Eq. (7), the convolution theorem and Eq. (2)were used. In the limit of a short SFG crystal, $\psi_{\text{SFG}}(\Delta \vec{\xi})$ behaves as a Dirac delta in the convolution (7), so that the PDC correlation can be exactly reconstructed by monitoring the SFG intensity as a function of Δt . In general, when the SFG crystal has a finite length, the measured signal (6) involves the combined properties of the PDC and SFG processes; however, provided that the two crystals are identical and tuned for the same phase-matching conditions, the two processes are exactly the reverse of each other, and the probability amplitudes F_{PDC} of downconversion and F_{SFG} of up-conversion overlap in the $(\vec{q},$ Ω) space. Thus, the integral in Eq. (6) is able to reconstruct the shape and width of $|\psi_{PDC}(\Delta x = 0, \Delta t)|^2$. The solid red line in Fig. 2 has been plotted from Eq. (6), where the spectral integration was limited to a bandwidth $\Delta \Omega_f =$ 0.9×10^{15} Hz, simulating the finite transmission bandwidth of the setup, and the baseline of the curve was adjusted to account for the experimental background (incoherent SFG + scattering + residual PDC light). It is remarkable that both the theoretical and measured profiles are well fitted by the empirical curve sinc²($\Delta\Omega_f \Delta t/2$), which is just the Fourier transform of a box function of width Ω_f in the frequency domain. This outcome gives clear evidence to our claim that dispersion is canceled by diffraction for the transmitted spectral modes, so that they all contribute to a localized temporal correlation summing up coherently.

The measurement has been repeated in nonideal conditions. Figure 3 shows the effect of a spatial filtering of twin beams, performed by inserting in the far-field of the PDC crystal a circular pinhole that clips an angular portion of the emitted radiation. The temporal correlation profile in Fig. 3 was obtained by placing a 4 mm diameter pinhole at 29 cm from the output face of the crystal. The data show a remarkable broadening of the *temporal* correlation, when filtering is performed over *spatial* degrees of freedom, which represents a first proof of principle demonstration of the nonfactorable character of the PDC correlation [3].



FIG. 3 (color online). Effect of spatial filtering. A pinhole clips an angular sector $\Delta \alpha = 0.79^{\circ}$ of the PDC light (see inset). The reconstructed temporal correlation broadens because the effective spectral bandwidth of phase-matched modes is reduced.

The broadening of the temporal peak occurs because, by cutting spatial modes, phase matching occurs inside a smaller temporal bandwidth $\Delta \Omega_{eff} \approx 0.34 \times 10^{15}$ Hz, as schematically shown by the inset of Fig. 3.

Even more remarkable are the effects of an imperfect imaging of the PDC light onto the SFG crystal. We repeated the measurement by displacing the SFG crystal by an amount $\Delta z_{\rm img}$ away from the optimal imaging plane at $4f_{OAP}$. The results, showing how the temporal correlation progressively undergoes broadening when an imaging error is introduced, are presented in Fig. 4. These data show how the diffraction introduced by displacements of a few hundred microns with respect to the imaging plane (i) reduces the efficiency of up-conversion and (ii) broadens the temporal correlation. The second effect is not trivial and again shows the interdependence of spatial and temporal degrees of freedom, because spatial diffraction in free space has an effect on the temporal correlation similar to temporal dispersion: an error in the imaging plane introduces a propagation phase that prevents the spectral modes in the bandwidth from summing up coherently in a narrow temporal peak. Formally, when the SFG crystal is shifted by $\Delta z_{\rm img}$ from the $4f_{\rm OAP}$ plane, an additional propagation phase is introduced under the integral in Eq. (5), so that $H_+(\vec{w})H_-(-\vec{w}) =$ $e^{-i\Omega\Delta t} \exp\left[-i\frac{q^2c}{\omega_1(1-\Omega^2/\omega_1^2)}\Delta z_{\rm img}\right]$ [8]. If we now make the simple assumption that only phase-matched modes contribute to the coherent SFG peak, we can use Eq. (3) to substitute $q^2 \rightarrow \Omega^2 k_1 k_1''$, with $k_1 = n_1(\omega_1) \omega_1/c$. The transfer function thus becomes $H_+(\vec{w})H_-(-\vec{w}) \rightarrow$ $e^{-i\Omega\Delta t} \exp[-i\frac{\Omega^2 k_1'' n_1(\omega_1)}{1-\Omega^2/\omega_1^2}\Delta z_{\rm img}]$, which describes a quadratic dispersionlike chirp of the twin beams.

In conclusion, we have been able to demonstrate—for the first time, to our knowledge—that the temporal corre-



FIG. 4 (color online). Effect of an imaging error on the reconstructed temporal correlation. When the second crystal is translated by Δz_{img} with respect to the $4f_{\text{OAP}}$ plane, the temporal correlation broadens.

lation of twin beams is as narrow as a few femtoseconds, in a setup that uses the inverse SFG process. This result relies not only on minimizing the temporal dispersion, thanks to an achromatic imaging, but also on the control of the spatial degrees of freedom of twin beams. Our counterexamples of Figs. 3 and 4 indeed show that spatial filtering and free-space diffraction broaden the temporal correlation, thus giving evidence of the interdependence of spatial and temporal degrees of freedom in PDC claimed by theory [3,4,8].

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