Second order coherence of broadband down-converted light on ultrashort time scale determined by two photon absorption in semiconductor

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Abstract: We study the photon correlation properties of broadband parametric down-converted light. The measurement of the photon correlation is carried out thanks to a modified Hanbury Brown-Twiss interferometer based on two photon absorption in GaAs detector. Since this method is not affected by the phase matching conditions of the detecting apparatus (so called "final state post-selection"), the detection bandwidth can be extremely large. This is illustrated by studying, with the same apparatus, the degree of second order coherence of parametric light in both degenerate and non-degenerate cases. We show that our experiment is able to determine the coherent as well as the incoherent contributions to the degree of second order coherence of parametric light with a time resolution in the fs range scale.

OCIS codes: (270.4180) Multiphoton processes; (270.5290) Photon statistics.

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Photon correlation properties are now harnessed in numerous experiments and applications, at the classical level (photon correlation spectroscopy [1], high resolution profiling [2]) as well as at the quantum level (quantum cryptography, quantum teleportation,…) [3]. The determination of the correlation properties of photon fields is thus of paramount importance, especially these of two-photon pair states generated by parametric fluorescence. Several schemes have been proposed to analyze the quantum correlation of this kind of light. In Hanbury Brown–Twiss (HBT) interferometry [4], the incident beam is split into two sub-beams by a beam splitter, sent on two separate photon counters the outputs of which are submitted to a correlation analysis. This technique has been used to characterize parametric fluorescence light sources for more than twenty years [5] and has a time resolution limited by the detector response time, i.e. in the nanosecond range [6,7]. To circumvent this bandwidth issue, Abram *et al.* measured these time correlations by sum frequency generation of the two delayed sub-beams in a nonlinear crystal [8]. More recently, following a pioneering experiment of Dayan *et al.* [9], several teams increased the performances of this latter experiment by compensating the dispersion and taking advantage of high non-linearity of current nonlinear crystals and detector improvement [10,11]. These experiments allow an excellent temporal resolution – down to the fs range – but are confined to narrow bandwidth final states imposed by phase-matching conditions which "postselect" only the contribution of photon pairs that are complementary in energy [12]. The response time of the detector can also be circumvented by the use of time-gated detection by up-conversion scheme as investigated in Ref [13]. However, the resolution time is then limited by the duration of the sampling pulse while the bandwidth is ultimately limited by the phase-matching acceptance of the up-conversion nonlinear crystal.

Recently, F. Boitier *et al.* [14] have developed a new technique based on two photon conductivity in semiconductors that enables the characterization of optical sources with output power down to 0.1 µW, bandwidth in the 1.3 to 1.6 µm range and time resolution in the femtosecond range. Experimentally, the system is rather similar to a Hanbury Brown–Twiss (HBT) interferometer but, in our case, the two delayed sub-beams are recombined in a two-photon counting device [15]. We will refer our technique to Two-Photon Counting (TPC) interferometry. Since, as sketched on Fig. 1a, two-photon absorption (TPA) in semiconductors occurs for photon energies larger than the semiconductor midgap and smaller than its gap, the detection bandwidth is very large, giving access to fs timescale correlation measurement and ultralarge bandwidth. Indeed, the virtual state lifetime can be estimated by the second Heisenberg uncertainty relation $\Delta E.\Delta\tau \sim \hbar/2$, leading to $\Delta\tau \sim 0.5$ fs for $\Delta E = E_g/2$ *i.e.* 0.7 eV [16]. It has been shown that the two photon counting rate measures the degree of second order coherence (DSOC) $g^{(2)}(\tau)$ [17] given by:

$$g^{(2)}(\tau) = \frac{\left\langle \hat{E}^{(-)}(t+\tau)\hat{E}^{(-)}(t)\hat{E}^{(+)}(t)\hat{E}^{(+)}(t+\tau)\right\rangle}{\left\langle \hat{E}^{(-)}(t)\hat{E}^{(+)}(t)\right\rangle^2} \qquad (1)$$

where τ is the delay between the two beams, $\hat{E}^{(+)}(t)$ and $\hat{E}^{(-)}(t)$ are the complex electric field operator and its hermitian conjugate respectively while $\langle \; \rangle$ stands for quantum expectation.

The purpose of our paper is the following. We investigate broadband down-converted light (by optical parametric generation or OPG) with our TPC interferometry technique and demonstrate that correlation properties between twin beams can be measured down to the femtosecond range. We show moreover, that TPC interferometry is not confined to the detection of photons complementary in energy, contrary to previous techniques [8,9]. This is due to the fact that (i) the final states correspond to the whole valence-to-conduction band transitions in the semiconductor and (ii) no phase matching limitation are involved in TPA. This is illustrated by studying with the same apparatus the correlation properties in both the degenerate and non degenerate OPG configurations.

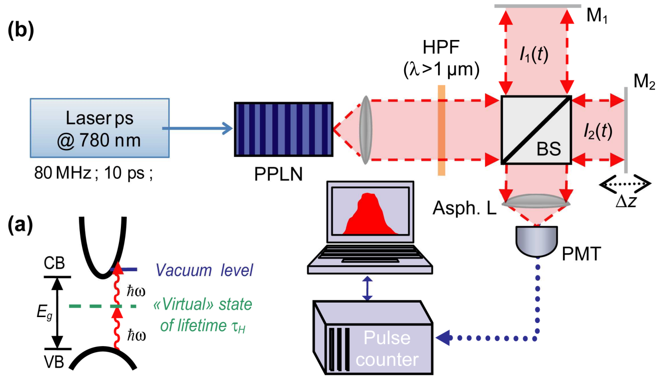

Fig. 1. (a) Two-photon absorption from valence band states to conduction band states in a direct gap semiconductor (e.g. GaAs). In a phototube, the electrons in the conduction band are emitted when reaching the extraction (or "vacuum") level. Only photons arriving within time intervals shorter than the "virtual" state lifetime at midgap τ_H can induce TPA transitions. (b) The HBT apparatus is a Michelson interferometer with two arms: (Asph. L) is a 26 mm aspheric lens, (BS) beam splitter, (HPF) high pass filter, (M1) and (M2) mirrors and (PMT) is the GaAs photomultiplier tube. The source is based on a periodically poled lithium niobate (PPLN) crystal pumped at 780 nm by a mode-locked Ti:Sapphire laser delivering 10-ps pulses at a 80-MHz repetition rate. Estimated focal spot on the detector is 5 µm, far smaller than the detector size.

In the present TPC interferometry experiment, we used a H7421-50 Hamamatsu GaAs phototube as suggested in Ref [15]. so that optical fields with wavelengths between 900 nm and 1,800 nm can be studied. In such a device, the photocurrent is highly amplified by multiplication of photoelectrons emitted from the space charge region of the semiconductor into vacuum, allowing to detect TPC signal in the Geiger mode with experimentally determined 40 dark counts/s (substracted in the following experiments). Figure 1b shows a schematic diagram of the experiment. It is a standard Michelson interferometric apparatus where special attention is given to the complete filtering out of radiation wavelengths shorter that 900 nm in the incoming light, in order to eliminate any direct absorption in the GaAs detector. The investigated parametric downconverter is based on an undoped type 0 35-mm-long periodically poled lithium niobate (PPLN) crystal pumped at 780 nm by a mode-locked Ti:Sapphire laser delivering 10-ps pulses at a 80-MHz repetition rate. By temperature tuning of the quasi-phase matching condition in the PPLN crystal, the second-order coherence can be studied at the degeneracy point of the parametric downconversion as well as far from degeneracy, in our case at respectively 125°C and 128°C.

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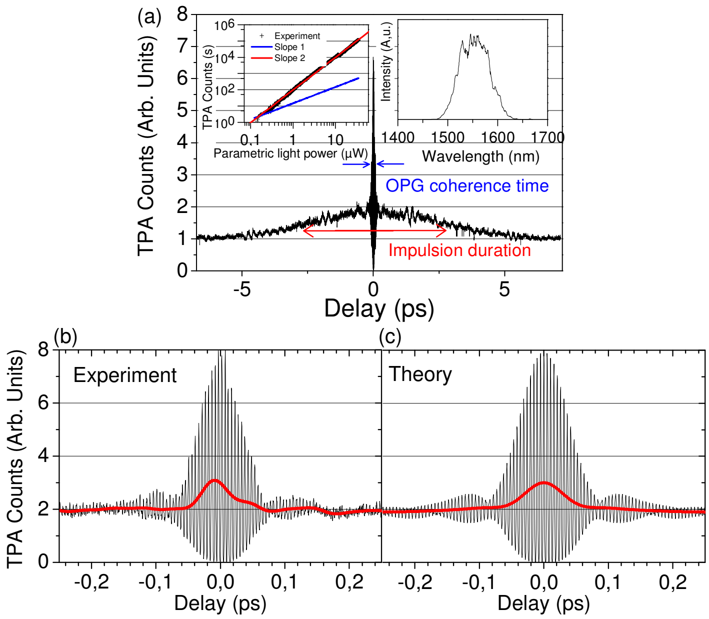

Fig. 2. (a) DSOC spectrum, i.e., variation of the TPA photocounts as a function of the delay τ, of the TPA Michelson set-up of Fig. 1. The left inset shows the number of photoelectron counts as a function of the incident power. The quadratic behaviour clearly indicates a TPA process with an OPG in high gain regime. The right inset shows the spectrum of the degenerate OPG. (b) Zoom on small delay times τ which exhibits the DSOC $g^{(2)}(\tau)$ features of the degenerate down-converted light. The red curve is $TPA_{LPF}(\tau)$ described in Eq. (2). (c) Theoretical modeling using the model described in the text.

Figure 2a shows a TPC interferogram carried out on the OPG light tuned at degeneracy, i.e. centred at 1.56 µm. The spectrum of the OPG emitted light is indicated in the inset of Fig. 2a. The broad peak in Fig. 2a reflects the pulse duration of the laser pump whereas the sharp peak gives access to the OPG correlation properties. This shows that, as far as the correlation properties of OPG photons are concerned, the experiment can be considered as cw. Figure 2b is a zoom of the Fig. 2a on the central peak where interference patterns are clearly observable. The red curve is the result of a low pass filter on the interferogram ($TPA_{LPF}(\tau)$): it is shown in Ref [18]. and [14] that it is equal to:

$$TPA_{LPF}(\tau) = 1 + 2g^{(2)}(\tau) / g^{(2)}(0) \; ; \qquad (2)$$

it thus measures the DSOC factor $g^{(2)}(\tau)$ and points out the photon coincidences. The measured correlation time of about 70 fs is consistent with the 100 nm source bandwidth (see right inset of Fig. 2a) while, using $g^{(2)}(\tau) \to 1$ for $\tau \approx 0.2$ ps, one has $g^{(2)}(0) \approx 2$, meaning that, in our experimental configuration, the properties of the DSOC are very close to what is obtained with chaotic light

We now show that the same TPC apparatus can be used to determine the correlation properties of the OPG beam away from degeneracy. The PPLN crystal temperature is changed in order to obtain two different radiations, the spectra of which is displayed in the inset Fig. 3a. The TPC interferogram of the two simultaneous radiations is shown in Fig. 3a while Fig. 3b shows the result with the signal only (the idler was attenuated by a dichroic mirror). Figure 3a shows that the influence of the two wavelengths is clearly observed. Using $TPA_{LPF}(\tau)$ (see Eq. (2), we can estimate a coherence time of about 140 fs which is again in

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compliance with the OPG bandwidth of 50 nm. Once again, we measure $g^{(2)}(0) \approx 2$ in both cases.

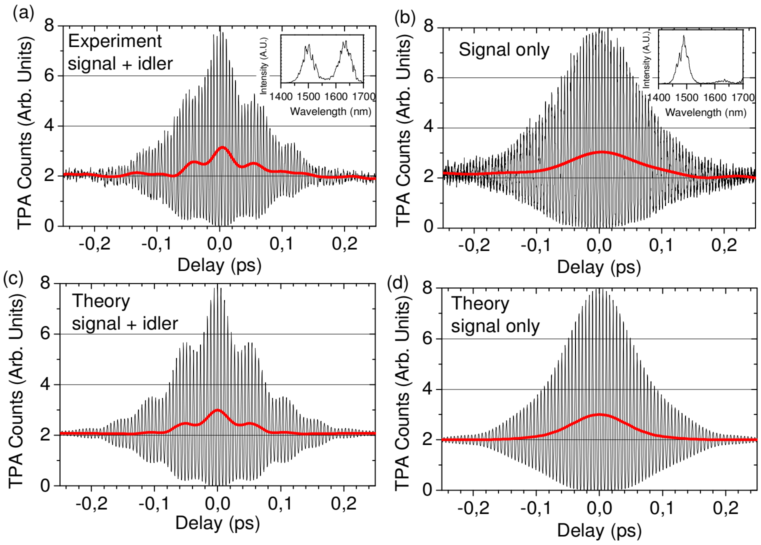

Fig. 3. (a) Zoom on the non-degenerate OPG part of the DSOC spectrum. The inset shows the spectrum of the non-degenerate OPG. (b) Zoom on the non-degenerated OPG part of the DSOC spectrum in the case where the idler wavelengths were attenuated by a dichroic mirror. The inset shows the spectrum after attenuation. (c) Theoretical modeling of the mutual DSOC spectrum of Fig. 3a. (d) Theoretical modeling of the mutual DSOC spectrum of Fig. 3b. The red curve is $TPA_{LPF}(\tau)$ described in Eq. (2).

For a proper interpretation of our experimental results and to get some physical insights, we developed a quantum optics theoretical model of the experiment. We briefly present the main steps of the calculation while a detailed description of the model will be presented elsewhere. To derive the creation and annihilation operators, we use the continuous variables version [19] of the formalism suggested by Huttner *et al.* [20].

The annihilation operator $\hat{a}(z_c,\omega)$ at the output of the PPLN crystal is related to the operators $\hat{a}(0,\omega)$ and $\hat{a}^\dagger(0,\omega_p-\omega)$ at the crystal entrance as follows:

$$\hat{a}(z_c,\omega)=\left[\mu(z_c,\omega)\hat{a}(0,\omega)+i\,\nu(z_c,\omega)\hat{a}^\dagger(0,\omega_p-\omega)\right]\exp\left\{i\left[\Delta k(\omega)/2+k(\omega)\right]z_c\right\}. \quad (3)$$

where $k(\omega)$ is the wavevector in the PPLN crystal at pulsation ω and $\Delta k(\omega)$ is the phase mismatch parameter given by

$$\Delta k(\omega)=k(\omega_p)-k(\omega)-k(\omega_p-\omega)-\frac{2\pi}{\Lambda} \quad (4)$$

with Λ the quasi-phase matching period of the PPLN crystal. In Eq. (3), $\mu(z,\omega)$ and $\nu(z,\omega)$ are the usual parametric propagation factors:

$$\mu(z,\omega)=\cosh\left[\gamma(\omega)z\right]-i\frac{\Delta k(\omega)}{2\gamma(\omega)}\sinh\left[\gamma(\omega)z\right] \quad (5)$$

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$$\nu\left(z,\omega\right)=\frac{g\left(\omega\right)}{\gamma\left(\omega\right)}\sinh\left[\gamma\left(\omega\right)z\right] \tag{6}$$

where $g\left(\omega\right)$ is the parametric gain:

$$g\left(\omega\right)=\frac{d_{\mathrm{eff}}}{c}\sqrt{\frac{2\omega\left[\omega_{\mathrm{p}}-\omega\right]Z_0 I_{\mathrm{p}}}{n\left(\omega\right)n\left(\omega_{\mathrm{p}}-\omega\right)n\left(\omega_{\mathrm{p}}\right)}}, \tag{7}$$

d_{eff} is the effective nonlinear coefficient, c is the speed of light, Z_0 (= 377 Ω) the vacuum impedance, I_{p} the incident pump intensity and $\gamma\left(\omega\right)$ is the effective parametric amplification factor:

$$\gamma\left(\omega\right)=\sqrt{g\left(\omega\right)^2-\Delta k\left(\omega\right)^2/4}. \tag{8}$$

Assuming negligible loss in the beam splitter, the annihilation operator at the detector position z_{d} can be straightforwardly connected to the one at the crystal output according to the following expression:

$$\hat{a}\left(z_d,\omega\right)=\tfrac{1}{2}\left[i\,M\left(\omega\right)\left(1+e^{-i\omega\tau}\right)\hat{a}\left(z_c,\omega\right)-\hat{\nu}\left(\omega\right)\left(1-e^{-i\omega\tau}\right)\right] \tag{9}$$

where $M\left(\omega\right)=e^{i\varphi(\omega)}$ accounts for dispersion experienced by the beam on its path from the crystal output to the TPC detector and $\hat{\nu}\left(\omega\right)$ is the vacuum field operator for the angular frequency ω at the output port of the Michelson interferometer. The TPC signal S_{TPC} is then proportional to [19]:

$$S_{TPC}\propto\left\langle\hat{a}^{\dagger}\left(z_{\mathrm{d}},t\right)\hat{a}^{\dagger}\left(z_{\mathrm{d}},t\right)\hat{a}\left(z_{\mathrm{d}},t\right)\hat{a}\left(z_{\mathrm{d}},t\right)\right\rangle \tag{10}$$

$$\text{with}\quad \hat{a}\left(z,t\right)=\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty}\mathrm{d}\omega\hat{a}\left(z,\omega\right)e^{-i\omega t}. \tag{11}$$

We would like to particularly discuss the DSOC whose value at zero delay – $g^{(2)}(0)\approx 2$ in all our experimental cases – could seem surprising if one considers the expected extra-bunching for a twin-photon beam when compared to chaotic light. Expanding Eq. (10) and extracting only low frequency terms, it can be shown that the calculated DSOC at the detector position is given by:

$$g^{(2)}\left(\tau\right)=g_{\mathrm{I}}^{(2)}\left(\tau\right)+g_{\mathrm{C}}^{(2)}\left(\tau\right), \tag{12}$$

where $g_{\mathrm{C}}^{(2)}\left(\tau\right)$ accounts for the so called "coherent" signal due to TPA induced by correlated twin-photon pairs while $g_{\mathrm{I}}^{(2)}\left(\tau\right)$ accounts for the so called "incoherent" signal due to TPA from signal–signal, idler–idler and uncorrelated signal–idler photons (so called "accidental coincidences").

In Eq. (12), the incoherent term can be written:

$$g_I^{(2)}=1+\tfrac{1}{\Phi^2}\left|\tfrac{1}{\sqrt{2\pi}}\int_0^{\omega_p}d\omega\left|\nu\left(z_c,\omega\right)\right|^2e^{-i\omega\tau}\right|^2=1+\left|g^{(1)}\left(\tau\right)\right|^2 \tag{13}$$

where Φ is the OPG photon flux. To go further in the interpretation, one can notice that

$$g^{(1)}(\tau)=\frac{\Phi_s}{\Phi_s+\Phi_i}g_s^{(1)}(\tau)+\frac{\Phi_i}{\Phi_s+\Phi_i}g_i^{(1)}(\tau) \tag{14}$$

where $g_s^{(1)}(\tau)$ (respectively $g_i^{(1)}(\tau)$) is the degree of first-order coherence of the signal (respectively idler) field alone while Φ_s (respectively Φ_i) is the signal (respectively idler) photon flux such as $\Phi = \Phi_s + \Phi_i$. As a consequence, we have

$$g_I^{(2)}(\tau)=1+\left(\frac{\Phi_s}{\Phi}\right)^2\left|g_s^{(1)}(\tau)\right|^2+\left(\frac{\Phi_i}{\Phi}\right)^2\left|g_i^{(1)}(\tau)\right|^2+2\,\mathrm{Re}\left[\frac{\Phi_s\Phi_i}{\Phi^2}\left|g_s^{(1)}(\tau)\right|\left|g_i^{(1)}(\tau)\right|e^{i(\omega_s-\omega_i)\tau}\right] \tag{15}$$

where, in the right-hand side of Eq. (15), the second term is related to TPA due to signal alone, the third one is related to idler alone, and the last one corresponds to uncorrelated signal–idler TPA. This last term contains the interference term of frequency $\omega_s-\omega_i$ at the origin of the modulation of the red curve on Fig. 3a and 3c. *One should note that a similar expression would be obtained if two chaotic sources with respective central angular frequencies* ω_s and ω_i were simultaneously sent in the TPC interferometer.

On the other hand, the coherent term in Eq. (12), can be written

$$g_C^{(2)}(\tau)=\frac{1}{\Phi^2}\left|\frac{1}{\sqrt{2\pi}}\int_0^{\omega_p}d\omega\,\mu(z_c,\omega)\nu(z_c,\omega)e^{i\left[\varphi(\omega)+\varphi(\omega_p-\omega)\right]}\,e^{-i\omega\tau}\right|^2 \tag{16}$$

As reported in previous studies [10–12], the coherent term depends drastically on dispersion since it requires twin-photon pairs to be synchronized.

In our case of high parametric gain, we can assume that

$$\mu(z_c,\omega)\approx\nu(z_c,\omega) \tag{17}$$

Thus, we get:

$$g_C^{(2)}(\tau)\approx\left|\frac{1}{\sqrt{2\pi}}\int_0^{\omega_p}\mathrm{d}\omega\left[\frac{1}{\sqrt{2\pi}}\int\mathrm{d}\tau' g^{(1)}(\tau')e^{i\omega\tau'}\right]e^{i\left[\phi(\omega)+\phi(\omega_p-\omega)\right]}e^{-i\omega\tau}\right|^2 \tag{18}$$

Hence, the coherent term $g_C^{(2)}(\tau)$ is altered by dispersion as would be a coherent short pulse whose complex electric field $E(t)$ is given by $E(t)=g^{(1)}(t)$. Such dependence of the coherent part as a function of the spectral phase manipulations is in good agreement with the previous work of Pe'er *et al.* [21]. One thus expect, a temporal shift of the coincidence delay ($\tau\approx(\omega_s-\omega_i)\phi''(\omega_p/2)$) as well as a broadening and an attenuation of the coherent term that scale as $\sqrt{1+\Delta^4\phi''^2(\omega_p/2)}$ where Δ is the spectral width of the parametric light and $\phi''(\omega_p/2)$ is the second derivative of the spectral phase, known as group delay dispersion, evaluated at the degeneracy frequency $\omega_p/2$. Conversely, one can notice that $g_C^{(2)}(\tau)\approx\left|g^{(1)}(\tau)\right|^2$ (and thus $g^{(2)}(0)\approx 3$) for negligible dispersion.

We have used this theory to account for our experimental results. As shown in Figs. 2 and 3, one can see that a very good agreement is obtained between experimental and calculated TPC interferograms when one accounts for dispersion of the optical elements. In our case, due to the dispersion of the optical components of our setup whose actual value is very close to $\phi''\approx 4000\ \mathrm{fs}^2$, twin-photon wave packets are stretched, leading to a very small coherent term observable in Fig. 3a. Indeed, this corresponds to the conditions when no signal can be measured in the experiment reported by O'Donnell *et al* [11] where only the coherent part of

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the DSOC function can be measured. The observed interference pattern is thus dominated by by the incoherent term (accidental coincidences) whose properties are similar to the case of chaotic light

In summary, two-photon counting interferometry, which harnesses two photon absorption (TPA) in a semiconductor, has been used in order to measure the correlation properties of broadband down-converted light. Intensity fluctuation correlation times can be easily measured in the few femtosecond range. This technique displays an extremely large bandwidth since it is not affected by phase matching conditions. This is illustrated by studying the degree of second order coherence of the down converted light, and away from degeneracy. The TPC interferometer enables us to finely characterize the incoherent part of the down-converted light and to experimentally demonstrate that the behaviour of photon bunching is then rather similar to the one observed in chaotic sources [14]. Work is in progress to enhance the overall quantum efficiency of TPC in order to investigate the low gain regime, as performed in Ref. [9].

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