Measuring photon bunching at ultrashort timescale by two-photon absorption in semiconductors

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Photon beams exhibit temporal correlations that are characteristics of their emission mechanism. For instance, photons issued from incoherent sources tend to be detected in bunches. This striking 'bunching' behaviour has been observed in the seminal experiment by Hanbury-Brown and Twiss (HBT) in the fifties, who measured the time of arrival of partially coherent photons on two separate photon-counting modules[1]. Since then, HBT interferometry has become a widespread technique to study photon correlations down to only the nanosecond range, because of the detector-limited bandwidth, preventing the observation of bunching for real thermal sources. It has been suggested later that two-photon absorption (TPA) could measure the photon temporal correlations at a much shorter timescale[2,3], as it involves an almost simultaneous absorption of two photons, within a maximum delay given by the Heisenberg principle. Here, for the first time, this prediction is experimentally demonstrated using TPA in a GaAs photon-counting module. We have observed photon bunching in the femtosecond range for real blackbody sources (an enhancement of six orders of magnitude in the time resolution of present techniques), opening the way to monitor optical quantum statistics at the ultrashort timescale.

The Hanbury-Brown and Twiss (HBT) experiment actually measures the degree of second-order coherence (DSOC) $g^{(2)}(\tau) = \langle I(t) I(t+\tau) \rangle / \langle I(t) \rangle^2$ of optical fields, where $I(t)$ is the intensity of the fields, τ is the optical path time difference between the two detectors and $\langle \rangle$ designates statistical averaging. A straightforward calculation, in which the fields are treated in terms of classical fluctuating electromagnetic waves, shows that $g^{(2)}(0)$ is always larger than 1 and is equal to 2 for chaotic sources[3]. The experiment was first carried out in the radiofrequency domain, using radiotelescopes, where the undulatory nature of light is unquestionable. It was later extended by the two authors to the optical domain, where they measured a value of $g^{(2)}(0) = 1.93$ using photocounters and a narrowband partially coherent light source (a 546.1 nm Hg discharge lamp followed by an interference filter)[4]. Although this result can still be interpreted classically in wave-like terms, its interpretation in terms of light corpuscles—more suited to photon counters—is more subtle, as it implies that photons emitted by incoherent sources are correlated: this is the so-called 'photon bunching effect'. Actually, the HBT experiment was a great surprise at that time and fostered a lot of controversy in the literature[5–8]. Later, Glauber[7] developed a unifying quantum theory of optical coherence. Using a quantum approach of the photodetection process, he demonstrated the following quantum expression for $g^{(2)}(\tau)$:

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

where $\hat{E}^{(+)}(t)$ and $\hat{E}^{(-)}(t)$ are the complex electric field operator and its Hermitian conjugate respectively[9] and $\langle \rangle$ now stands for quantum expectation. Using expression (1), the value of 2 for $g^{(2)}(\tau = 0)$ for chaotic fields is retrieved. It is also shown that values below 1 can be obtained using non-classical sources of light such as single-photon sources ('photon antibunching' effect)[10]. As first shown by Fano, the HBT effect can be considered as the result of quantum path interferences between two photons emitted by two different atoms and reaching the two detectors[5,11,12]. All of these studies concerning the respective merits of the classical versus quantum description led to the modern theory of quantum optics, which proved so successful later. A more recent example of such a duality of interpretation concerns the intriguing 'ghost' imaging effect, which was predicted and observed first using non-classical quantum correlated photon sources, and which was later observed using the second-order coherence properties of classical thermal light[13–16].

HBT experiments are usually carried out by splitting the optical beam on a beam splitter, retarding one of the two subsequent optical beams by a delay τ and measuring the intensity correlation between the two beams on two distinct detectors using a coincidence counting technique[17,18]. Surprisingly, the value 2 of $g^{(2)}(\tau = 0)$, interpreted as a bunching of photons, has never been experimentally observed with real blackbody sources. The reason for this is the time response τ_r of photon detectors (that is, the minimum resolution time), which is at best in the nanosecond range. As was shown by Scarl, the experimental value of $g^{(2)}(\tau = 0) - 1$ scales as τ_c/τ_r, where τ_c is the inverse of the optical source bandwidth, that is, the coherence time[6,18], and is therefore significant only when the correlation time of the source is of the order of the detection time response. Experiments have so far been carried out by means of pseudo-blackbody sources, such as rotating and/or diffusing discs[19] or narrowband-filtered sources[1,6].

A number of other techniques have been proposed to alleviate the detector time constraint, but none satisfies the performances necessary for blackbody studies. Sum frequency generation in a nonlinear crystal[20–22] enables quasi-instantaneous measurements. However, low yield and stringent phase-matching conditions

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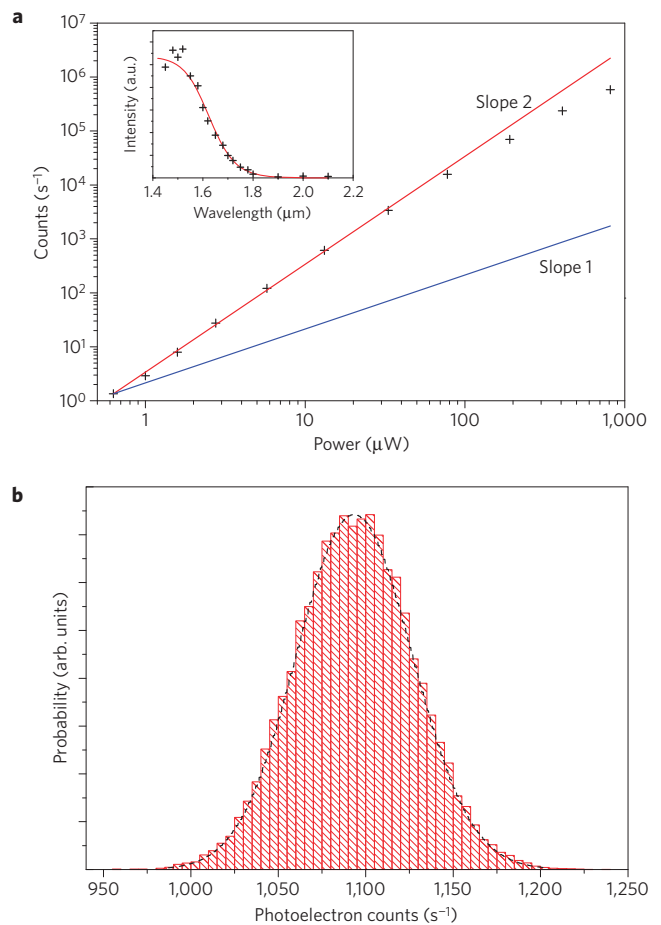*e-mail: emmanuel.rosencher@onera.fr.
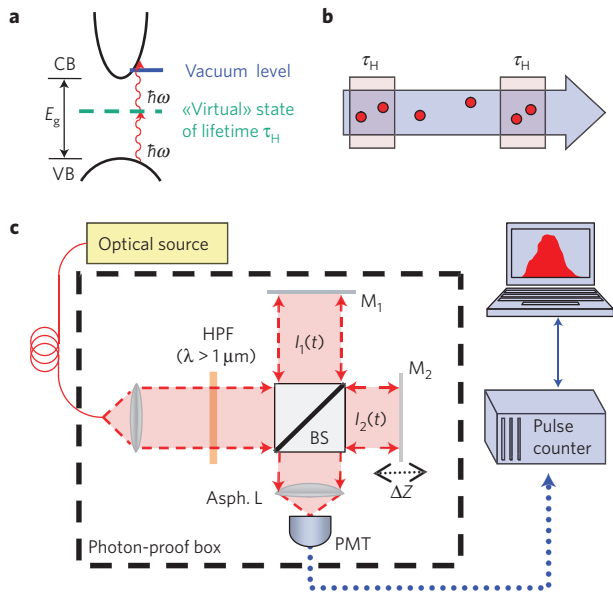
Figure 1 | Principle of TPA and experimental set-up. a, TPA from valence-band (VB) states to conduction-band (CB) states in a direct-gap semiconductor (such as GaAs). In a phototube, the electrons in the conduction band are emitted when reaching the extraction (or 'vacuum') level. **b**, Only photons arriving within time intervals shorter than the 'virtual' state lifetime at midgap τ_H can induce TPA transitions. **c**, The HBT apparatus is a Michelson interferometer with two arms. Asph. L: 26 mm aspheric lens, BS: beam splitter, HPF: high-pass filter, M_1 and M_2: mirrors, PMT: GaAs photomultiplier tube. All of the experiments are carried out in a photon-proof box with optical-fibre light delivery. The estimated focal spot is 5 μm, far smaller than the detector size.

Figure 2 | Characterization of TPA detection response. a, Number of photoelectron counts as a function of incident power. The source is an attenuated 1.55 μm laser diode. The quadratic behaviour clearly indicated a TPA process. The inset shows the detection bandwidth determined with a filtered supercontinuum source. **b**, Histogram of the number of TPA photoelectron counts. The optical source is a continuous-wave ASE incoherent source peaking at 1.55 μm for an incident power of 200 μW. The fit indicates a Poissonian statistics.

still exclude studies of large-bandwidth continuous-wave sources such as thermal blackbodies. Nevertheless, Qu and Singh were able to demonstrate the bunching of photons spontaneously emitted from a laser apparatus below threshold by measuring the enhancement of second harmonic generation yield[22]. However, they did not carry out any intensity autocorrelation measurement in which the estimated time resolution would have been longer than 10 ps because of the phase-matching spectral acceptance. The Hong–Ou–Mandel interferometer also gives access to the femtosecond timescale but addresses very specific entangled photon light states, or single photons in a well-defined temporal, spectral and spatial mode[23].

Since the early days of quantum optics, it has been predicted that two-photon absorption (TPA) can be used to measure photon correlation[2,3]. Indeed, TPA transition rates are directly related to the expectation value of $\hat{E}(t)^{(-)}\hat{E}(t)^{(-)}\hat{E}(t)^{(+)}\hat{E}(t)^{(+)}$. Georgiades et al.[24] have experimentally shown the non-classical behaviour of TPA using an atomic narrowband resonance. More recently, TPA in semiconductors has been used to measure autocorrelation spectra of high-power femtosecond sources[25]. Since then, the technique has been extended to very low-power femtosecond sources[26]. Here, we show for the first time that, indeed, the photocurrent due to TPA in semiconductor devices enables a direct measurement of the second-order coherence of stationary optical sources such as amplified spontaneous emission (ASE) and blackbody sources.

TPA in semiconductors is a well-known effect, which is well understood in the framework of the quantum theory of light[3,27]. In an illustrative way, it can be considered as a transition of an electron from a valence-band state to a 'virtual' state in the gap of energy E_g, followed by a transition to a real state in the conduction band (Fig. 1a). For such a process to occur, the two photons must

be absorbed within a time interval smaller than the Heisenberg lifetime $\hbar/E_g$, that is, a few 10^{-15} s in the optical range (Fig. 1b). It is thus easy to understand that this technique is well fitted to study optical correlation at ultrashort timescales. TPA occurs for all photon energies larger than the semiconductor midgap and smaller than its gap, inducing a photocurrent in the semiconductor. The detection bandwidth is thus very large.

In the present experiment, we used an H7421-50 Hamamatsu GaAs phototube as suggested in ref. 26 so that optical fields of wavelengths between 900 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, enabling photon counting with experimentally determined 40 dark counts per second. These dark counts are subtracted in the following experimental results.

Figure 1c 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, to eliminate any direct absorption in the GaAs detector. Two kinds of highly incoherent optical source have been used: an Er^{3+}-doped fibre ASE source emitting at a centre wavelength of 1.55 μm (BT 1200 Photonetics) and a halogen lamp exhibiting blackbody
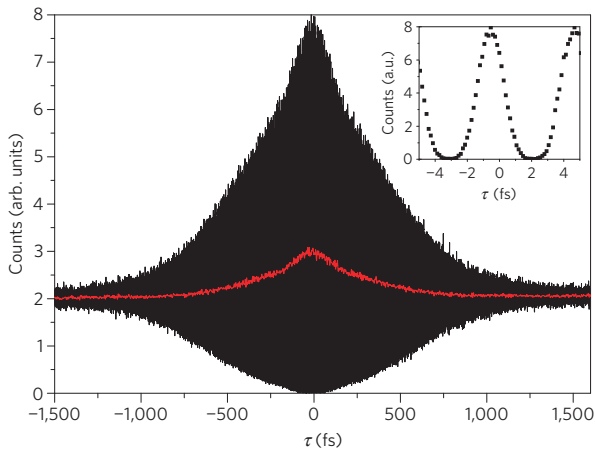
Figure 3 | Variation of the TPA photocounts as a function of the delay τ in the Michelson set-up. The optical source is a continuous-wave ASE incoherent source peaking at 1.55 μm for an incident power of 200 μW. The inset shows an enlarged view of the interferogram. The solid red line, obtained by filtering out the high-frequency components of the interferogram, is proportional to the DSOC.

characteristics at 3,000 K ($\pm$100 K). For each incoming light beam in the arms of the interferometer, the instantaneous photocurrent emitted from GaAs is given by:

$$J(t) = \alpha P(t) + \beta \frac{P(t)^2}{S}$$

where α and β are the one- and two-photon quantum efficiency of the semiconductor device. P is the incident power and S is the spot area. α should be as close to zero as possible and β corresponds to a TPA coefficient of 10 cm GW^{-1} on a 1-μm-thick depletion layer. Alignment and optimization were done by a Z-scan technique[28]. Figure 2a shows the $J(P)$ curve, which exhibits two orders of magnitude of quadratic behaviour from 0.6 to 100 μW using a 1.5 μm attenuated laser diode[26], that is, five orders of magnitude in the photocurrent leading to a good agreement for the value of β. Deviation at high power is due to the deadtime overlap of the detector. No signal related to one-photon absorption α is observable. The detection bandwidth, determined using a filtered supercontinuum source (from 1.4 to 2.2 μm), is shown in the inset of Fig. 2a.

Figure 2b shows a typical histogram of TPA photocounts for the ASE source at 200 μW, that is, the probability to count N TPA photoelectrons as a function of N during a time T ($T = 10$ ms here). N is related to the number of photons n incident on the detector by $N = \eta_2 n^2$, where η_2 is the TPA quantum efficiency linearly connected to β. A Poissonian behaviour ($\langle \Delta N^2 \rangle = N$) is observed on the full optical power range whatever the optical source, that is, laser, ASE or halogen lamp. Such a behaviour, reminiscent of partition noise in a single-photon detector with a low quantum efficiency[3], is surprising at first sight for a two-photon detector: a simple calculation shows indeed that partition noise in a low-efficiency TPA detector leads to a super-Poissonian behaviour with $\langle \Delta N^2 \rangle = 2N$. A likely explanation of the observed Poissonian behaviour is the following. Our TPA detector actually operates in the counting regime, yielding the same single 'click' whether two photons or more are simultaneously incident on the detector, whereas it does not click in the presence of 0 or 1 photon. The quantum observable $\hat{O}_{\text{TPA}}$ associated with such a measurement is therefore $\hat{O}_{\text{TPA}} = \hat{I} - |0\rangle\langle 0| - |1\rangle\langle 1|$. From this expression, it can easily be shown that the two-photon photocount noise is Poissonian at the limit of small quantum efficiency, whatever the statistics of the incident light.
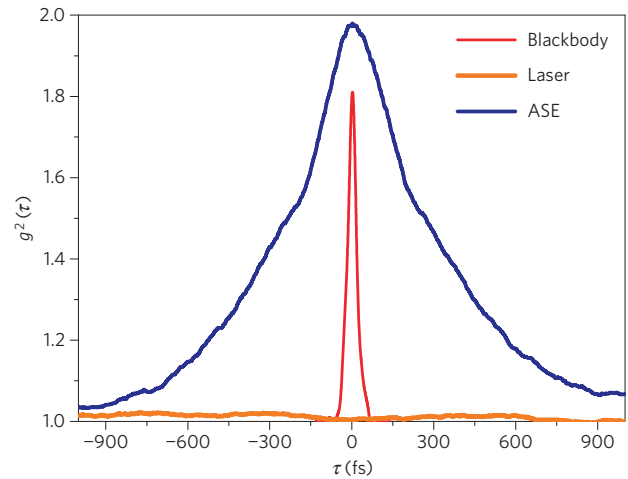
Figure 4 | DSOC of a 1.55 μm diode laser and two highly incoherent sources, an ASE and a 3,000 K blackbody source as determined by the TPA experiments.

The second-order coherence spectra are carried out by scanning one arm of the interferometer relatively to the other, taking care of the drastic alignment constraints during the scans. Figure 3 shows the result for the ASE source. The features can be understood using a generalization of ref. 29:

$$\frac{I(\tau)}{I_1 + I_2} = 1 + 2G_2(\tau) + \mathrm{Re}\left[F_2(\tau)\mathrm{e}^{-2i\omega\tau}\right] + 4\mathrm{Re}\left[F_1(\tau)\mathrm{e}^{-i\omega\tau}\right] \quad (2)$$

In equation (2), I is the photoelectron interferometric signal; I_1 and I_2 are the photoelectron signals obtained using each arm of the interferometer separately. The oscillating terms in brackets, written using notation from ref. 29, are shown in greater detail in the inset of Fig. 3. G_2 is the intensity interferometric signal and F_1 and F_2 are reminiscent of the phase interferences. $G_2(\tau)$ is obtained by filtering out the interferogram at low frequency. The result of this procedure is shown in Fig. 3 (red line).

Assuming complete decorrelation for long delay time, g_2 is experimentally determined by plotting $g^{(2)}(\tau) = G_2(\tau)/G_2(\infty)$. The values of $G_2(\infty)$ are determined from the portions of the $G_2(\tau)$ curves that do not show any measurable variation with increasing $|\tau|$, for example, between 2 and 3 ps for the ASE curve (not shown in Fig. 4). Figure 4 compares the DSOC obtained with a stable laser, the ASE and the halogen lamp source. The DSOC at zero time delay $g^{(2)}(0)$ is 1.0 ± 0.1 for the laser, 1.97 ± 0.1 for the ASE and 1.8 ± 0.1 for the halogen lamp. The photon bunching for the real chaotic sources is thus clearly observed and determined. Moreover, the results are in quantitative agreement with the theoretical expectation $g^{(2)}(\tau) = 1 + \mathrm{e}^{-\pi(\tau/\tau_c)^2}$ for a chaotic Gaussian source with coherence times τ_c equal to 534 and 37 fs for the ASE and the halogen lamp respectively, in compliance with the source bandwidth (for example, 300 nm for the latter). Let us stress once again that such a result is not obtainable with a classical Hanbury-Brown Twiss set-up because of the integration time of detectors, which limits the validity of the HBT experiment to τ_c values larger than the integration time, that is, in the nanosecond range.

The experiment still suffers from an overall low quantum yield, which could be enhanced using, for example, resonant three-level quantum-well infrared detectors[30] at the expense of the spectral bandwidth. We plan to extend these studies to the case of broadband non-classical sources and also to the characterization of future intense single-photon sources (with rates as high as 10^{14} s^{-1}), which are of paramount importance in quantum cryptography for instance. The same detection strategy can also lead to interesting

studies concerning spatial photon (anti)bunching, as TPA is highly sensitive to the photon spatial overlap.

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Author contributions

All authors contributed at each step of this work.

Additional information

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