QUANTUM OPTICS

Photon bunching two by two

Although the bunching of photons emitted from an incoherent source is well known, the nanosecond response times of conventional photon-counting detectors have prevented it from being observed directly. Using the ultrafast two-photon absorption characteristics of a semiconductor detector, such effects can now be studied at femtosecond timescales.

Giuliano Scarcelli

Imagine a star or an incandescent lamp that emits photons spontaneously. Now, imagine a photodetector that collects photons from this light source. You might expect that the arrival and detection of photons from such a chaotic light source would occur at random. Yet this is not the case. Surprisingly, the probability of detecting many photons in close succession is higher than that for those farther apart. Such 'photon bunching' behaviour has intrigued physicists since it was first identified by Robert Hanbury Brown and Richard Twiss (HBT) in 1956 (ref. 1). However, despite 50 years of research, photon bunching from an ideal broadband chaotic source has never been observed directly. This is because the characteristic period over which photons from such a source are correlated — determined by the coherence time of the source — is far shorter than the typical nanosecond response time of a conventional photodetector. Now, on page 267 of this issue[2], Boitier and colleagues show that by exploiting the two-photon absorption characteristics of a semiconductor-based photon detector, the temporal resolution with which these correlations are measured can be extended down to the scale of femtoseconds, opening an entirely new regime for the study of the HBT effect.

Hanbury Brown and Twiss first discovered photon bunching by using a novel type of stellar interferometer consisting of a single half-silvered mirror to split and redirect starlight to two photomultiplier tube detectors separated by a distance of several metres (Fig. 1a). The original aim of their experiment was to show how the size of distant stars could be measured without going to extensive lengths to preserve the coherence of an incoming signal, as required by conventional interferometers. This was achieved by measuring the increasing correlation between the signal intensities of the two detectors as they and the silvered mirror were aligned to point exactly in the direction of a particular star. This has come to be known as the spatial HBT effect.

Soon afterwards, the two researchers set out to investigate a similar effect, but in the temporal rather than the spatial domain, using a narrow emission line from a mercury arc lamp. They found that for a chaotic source, the probability of detecting two photons — one at each detector — with zero delay between their arrival was twice as large as that of statistically independent particles (Fig. 1b).

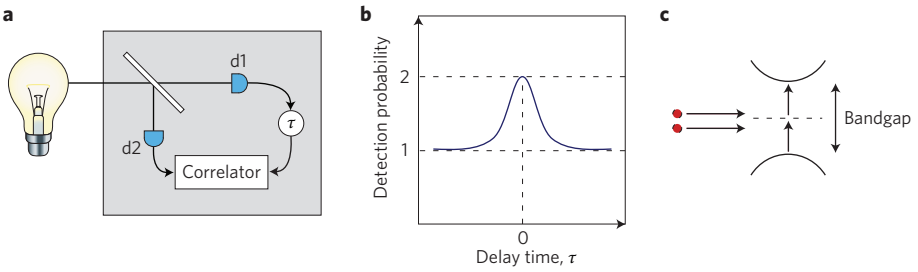

Figure 1 | Measurement of photon bunching in an ultrafast Hanbury Brown and Twiss interferometer. **a**, In a typical photon-bunching temporal measurement, photons from a chaotic source of light are split at a beamsplitter and measured with two photodetectors and a correlator. **b**, The measured joint probability of photodetection shows the 'photon bunching' peak whose maximum occurs at zero delay in the paths of two incident photons. **c**, Boitier and colleagues[2] use the principle of two-photon absorption to generate photoelectrons in a GaAs detector, to achieve detection that is six orders of magnitude faster than is possible with conventional photon-counting devices.

This finding immediately triggered a fierce debate on its physical basis[3] — a debate that was pivotal in the development of the quantum theory of light detection by Glauber[4] and marked the birth of the field of quantum optics. In itself, there was no problem in establishing a classical interpretation of HBT based on the fluctuation of continuous waves of light. But, as Hanbury Brown noted[1], "The trouble of course was due to worrying about photons." The quantum interpretation of HBT, based on the interference between indistinguishable Feynman amplitudes, was harder to envisage by physicists of the day. Yet this picture is now used to explain the photonic HBT effect and similar phenomena shown by other systems of bosons and even fermions[5]. And HBT-type experiments are not restricted to the exploration of quantum optics but are regularly used in particle physics[6] and in fluid dynamics[7].

Given such progress, it is surprising that it has taken so long to get to a point where temporal photon bunching from real thermal sources can be measured directly. So the demonstration by Boitier *et al.* is all the more impressive. Instead of the two photomultiplier tubes of a standard HBT interferometer, they use a Michelson interferometer arrangement consisting of two fully silvered mirrors (in addition to the half-silvered mirror that splits the incoming beam) to direct both signals to a single GaAs semiconductor detector. The detector operates on the principle of two-photon absorption (TPA) such that, ideally, it will only register a detection event when it absorbs two incoming photons within a time interval less than the lifetime of the 'virtual' midgap state of the GaAs (Fig. 1c). The idea of using TPA for coincidence measurements is not new, but because of limitations in the detection sensitivity of typical semiconductor detectors it had never previously been made to work for an HBT experiment. The detector used by Boitier *et al.* strikingly shows no residual one-photon absorption, so the multiplication of photoelectrons emitted from the semiconductor space charge region into the vacuum can be fully exploited. This enables the photocurrent to be highly amplified while keeping the dark count extremely low. The resulting interferometer achieves a temporal resolution that is six orders of magnitude better than a conventional HBT interferometer, enabling the authors to measure a nearly optimal HBT signal (1.8 times the random coincidence level) from a blackbody source with a coherence time of 37 fs.

Besides representing a landmark observation in quantum optics, this achievement opens many new possibilities for the exploration and exploitation of photon-bunching effects. For example, ultrashort coincidence counters can be used to characterize bright single-photon sources for quantum communication[8] and could enable the flux of non-classical photons to be enhanced while still maintaining distinctive quantum features[9]. In addition, it will be interesting to see the implications of this experiment on the spatial domain. Fast two-photon correlation detection may broaden the scope of recent imaging protocols based on the spatial HBT effect[10,11] as well as being useful for high-flux quantum lithography and microscopy[12]. To realize such possibilities, however, further technical improvements are needed, most notably with respect to the overall quantum yield, which at the present level does not allow accurate retrieval of the photon statistics of light sources, and to the spectral bandwidth, which may mask the effects produced by non-classical light. Moreover, the TPA method is a zero-delay zero-displacement coincidence counter; it does not directly probe the complete behaviour of HBT correlation functions, which may limit its range of applicability. ❐

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MOLECULAR PHYSICS

Molecules standing to attention

The combination of quantum-state selection and shaped femtosecond laser pulses provides a tool for creating samples of isolated molecules with precisely defined and controlled spatial orientation.

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Much of our present understanding of molecular properties has come from studies of isolated gas-phase molecules, often prepared in supersonic molecular beams. Such gas samples are randomly oriented; that is, inherently all possible orientations of the molecules are present. Any measurement made on such a sample therefore suffers an averaging over all molecular orientations, and this substantially reduces the information obtained (see Fig. 1a). To avoid this orientational averaging, it is necessary to constrain and define the direction of the molecules. This can be done 'invasively', for example by trapping the molecules in an inert-gas matrix or by absorbing them onto a metal surface, but it is desirable to define the orientation of the molecules in the gas phase, so as to measure their native properties. On page 289 of this issue, Omair Ghafur and co-workers[1] report a method that enables just that, by marrying two established techniques: molecular-quantum-state selection and forced molecular orientation by combining an intense shaped laser field with a static electric field.

In the experiment of Ghafur *et al.*, a linearly polarized laser field is used to produce a high degree of localization of the molecular axes of nitric oxide molecules. This process, referred to as molecular-axis alignment, arises as a result of a torque on the molecules resulting from the interaction between the laser-induced molecular dipole moment and the intense electric field of the laser. This torque causes the molecules to rotate towards the direction of the laser polarization (see Fig. 1b). Laser-induced molecular-axis alignment has become well established in recent years[2], but the degree of alignment produced in the sample is normally limited by the initial incoherent populations of rotational quantum states present in the gas sample — each rotational quantum state corresponds to a different classical rotational velocity, hindering simultaneous alignment of all molecules in the sample. Ghafur and colleagues[1] work around this problem by sending the molecular beam first through a hexapole electric field. This delivers only the molecules in the ground rotational quantum state to the laser interaction region, producing a sample with effectively zero rotational temperature. Starting with this quantum-state-selected gas, Ghafur *et al.* achieve substantially higher degrees of molecular-axis alignment than with the conventional approach, and the degree is further increased by carefully designing the shape of the applied laser pulse.

This laser-alignment process would normally produce equal numbers of molecules with the nitrogen atom pointing up and pointing down (Fig. 1b). But Ghafur *et al.* use in their experiment[1] a static electrical field of moderate strength, to provide a preferred direction for the nitrogen end of the molecule by means of the first-order Stark interaction with the permanent dipole moment of the molecule, thus defining the orientation of the molecules. In this way, the laser and electric fields act in concert to orientate the gas molecules (Fig. 1c).

The duration of the shaped femtosecond laser pulse employed by Ghafur *et al.*[1] is much shorter than the period of molecular rotation, which means that axis orientation happens after the laser pulse (and recurs at regular intervals determined by the rotational period of the nitric oxide molecules). High degrees of molecular orientation are therefore achieved at instants when the laser field is absent, and this facilitates the measurement of molecular properties without disturbance by the strong laser field. This key feature distinguishes the work of Ghafur and colleagues from a recently published approach in which the orientation of quantum-state-selected iodobenzene was reported[3]. In the latter work, molecular orientation was achieved only during the application of the laser field, because a substantially longer unshaped laser pulse was used.

The techniques shown by Ghafur *et al.*[1] should find broad application in molecular science and should enable, for example, studies of the dependence of chemical reactions on the relative orientation of colliding molecules. Controlled molecular orientation has a central role in emerging techniques employing strong-laser-field processes for studying molecular structure,