review articles

Evidence for the quantum nature of light

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A unique property predicted by the quantum theory of light is the phenomenon of photon antibunching. Recent theoretical predictions and experimental observations of photon antibunching in resonance fluorescence from a two-level atom are reviewed.

THE advent of photon correlation experiments, as pioneered by Hanbury-Brown and Twist[1], began a new era in the field of optics. The experiments of classical optics involved a measurement of the first-order correlation function of the electromagnetic field and as such involved only the interference between the probability amplitudes of a single photon. Phenomena such as diffraction, Young's interference experiment and spectral measurements may be categorised as being in the domain of one photon or linear optics. Photon correlation experiments represent a fundamental deviation from these experiments as they involve the interference between different photons and as such are in the realm of nonlinear optics. The quantum-mechanical interpretation of the Hanbury-Brown Twiss effect was given by Glauber[2]. However, adequate explanations of the Hanbury-Brown Twiss effect and related photon correlation experiments have been given which invoke a classical description of a fluctuating electromagnetic field. However, Glauber[2] pointed out that photon correlation experiments offer the possibility of observing a uniquely quantum-mechanical effect—namely photon antibunching. We describe here the recent prediction by Carmichael and Walls[3] and observation by Kimble, Dagenais and Mandel[4–6] and G. Leuchs, M. Rateike and H. Walther (personal communication) of this unique quantum-mechanical effect in photon correlation experiments of fluorescent light from a two-level atom. This marks a dramatic change from all previous photon correlation experiments since the photon antibunching observed cannot be explained on the basis of a classical description of the radiation field. We begin with a brief outline of events leading up to the present investigations.

The quantum theory of light

The quantum theory of light beginning with Planck[7] and Einstein[8] played a central part in the development of quantum theory during this century. As the quantum theory was developed a sophisticated theory for the interaction of photons and electrons namely quantum electrodynamics evolved. Amongst the predictions of quantum electrodynamics was that the emission of light from an atom would experience a small shift away from the resonance line of the atom. The experimental observation of this shift, known as the Lamb shift, came as a major triumph for the quantum theory of light[9]. However, for many experiments in optics especially in the field of physical optics the classical picture of light as propagating waves of electromagnetic radiation was adequate to describe the observations. The idea developed that it was not necessary to quantise the light field but only necessary to quantise the atoms and thus describe their interaction by a semiclassical theory. Modifications of this concept such as neoclassical radiation theory also developed. These theories can explain phenomena such as the photoelectric effect, spontaneous emission and even give estimates of the Lamb shift. (A discussion on the present status of such theories as an alternative to quantum electrodynamics is given in ref. 10.) An early attempt to find quantum effects in a physical optics experiment was made by Taylor[11] who performed Young's interference experiment at very low intensities such that on the average only one photon was incident on the screen at a time. Integrating over a long detection time Taylor obtained an interference pattern as predicted by classical wave theory with no evidence of any unique quantum effects. Taylor's experiment may also be explained on the basis of the quantum theory which interprets it as the interference of the quantum-mechanical probability amplitudes for the photon to go through one slit or the other[12,13]. Essentially the experiments of physical optics were in the regime of one photon or linear optics. We need to go outside the experiments of one photon optics in order to detect any uniquely quantum-mechanical effects.

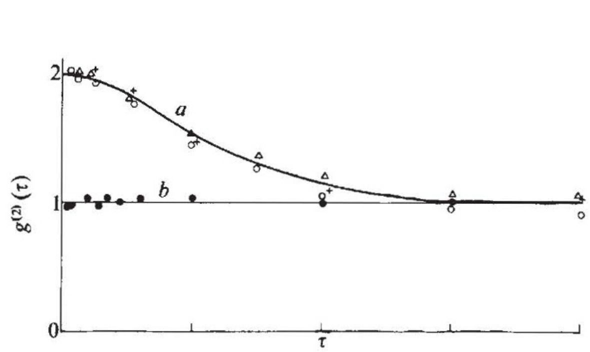

Fig. 1 Second order correlation function $g^{(2)}(\tau)$ for: *a*, thermal light; *b*, coherent light. Experimental points from Arrechi *et al.*[17].

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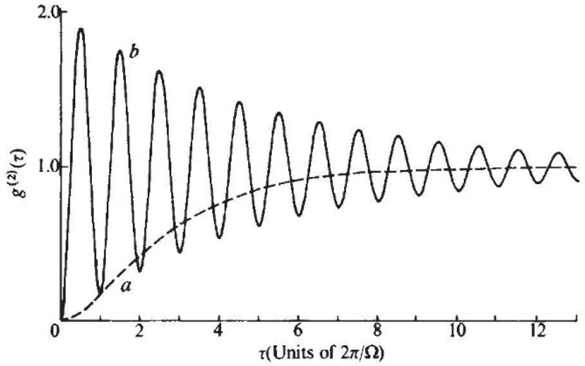

Fig. 2 Second order correlation function $g^{(2)}(\tau)$ for fluorescent light from a two level atom: *a*, low driving field intensity $\Omega \ll \gamma$; *b*, high driving field intensity $\Omega \gg \gamma$. Theoretical predictions from Carmichael and Walls[3].

Photon correlation experiments

The first experiment outside the domain of one photon optics was performed by Hanbury-Brown and Twiss in 1956. They performed an intensity correlation experiment, that is a measurement of $\langle I(t)I(t+\tau)\rangle$. Although the original experiment involved the analogue correlation of photocurrents, later experiments used photon counters and digital correlators[14–18]. In essence these experiments measure the joint photocount probability of detecting the arrival of a photon at time t and another photon at time $t+\tau$. The measurements made in this experiment may be described in terms of the second-order correlation function of the radiation field introduced by Glauber[2] in his formulation of optical coherence theory. This is defined as

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

where $E^{(+)}(t)$ and $E^{(-)}(t)$ are the positive and negative frequency components of the electromagnetic field respectively.

The result of a photon correlation measurement of $g^{(2)}(\tau)$ for a thermal light source using photomultipliers and digital correlators is shown in Fig. 1 curve *a*. We see that the joint counting rate for zero time delay is twice the coincidence rate for large time delays τ. That is, there is a tendency for the photons to arrive in pairs, or a photon-bunching effect. The decay time of the correlations is given by the inverse bandwidth of the light source.

If instead of a thermal light source the experiment is performed with a highly stabilised laser source one obtains a constant correlation function as shown in Fig. 1 curve *b*. This result holds even if the laser and thermal light source have the same bandwidth. Hence there is some fundamental difference between a laser and a thermal light source which may not be apparent in the first-order correlation function but which is manifest in the second-order correlation function. This effect may be understood by considering the photon statistics of different light fields.

Quantum theory of photon correlations

We shall attempt in this section to give a quantum theoretic interpretation of photon correlation experiments. For simplicity we shall concentrate on an interpretation of $g^{(2)}(0)$ which enables us to restrict our attention to a single mode field. For a single mode field we may write $g^{(2)}(0)$ in the form

$$g^{(2)}(0)=1+\frac{\sigma^2-\bar{n}}{\bar{n}^2} \qquad (2)$$

where $\bar{n}$ and σ^2 are the mean and variance respectively of the photon number distribution. Thermal light has a power law photon number distribution with variance $\sigma^2=\bar{n}^2+\bar{n}$ which gives $g^{(2)}(0)=2$. Coherent light on the other hand as produced by a highly stabilised laser has a poissonian photon number distribution[19–21] with $\sigma^2=\bar{n}$ leading to $g^{(2)}(0)=1$. This clearly yields the results shown in Fig. 1 for $g^{(2)}(0)$. To include the decay time of the correlation function one must consider many modes. For a field which has a photon number distribution narrower than a Poisson ($\sigma^2<\bar{n}$) it is possible in principle to obtain a $g^{(2)}(0)<1$. That is the photon coincidence rate for zero time delay is less than that for large time delays τ. This is the opposite effect to the photon bunching observed for thermal light and has been called photon antibunching. This represents a reduction in the photon number fluctuations below that of a poissonian distribution. The extreme case of photon antibunching would be a light beam where the photons arrive evenly spaced. For a single mode field this is equivalent to having a fixed photon number N which gives $g^{(2)}(0)=1-1/N$.

Let us now consider a classical description of the electromagnetic field. We consider a field described by a fluctuating amplitude E. These fluctuations are taken into account by introducing a probability distribution $P(E)$ for the complex field amplitude. A calculation of $g^{(2)}(0)$ yields

$$g^{(2)}(0)-1=\frac{\int P(E)(|E|^2-\langle|E|^2\rangle)^2\,\mathrm{d}^2E}{(\langle|E|^2\rangle)^2} \qquad (3)$$

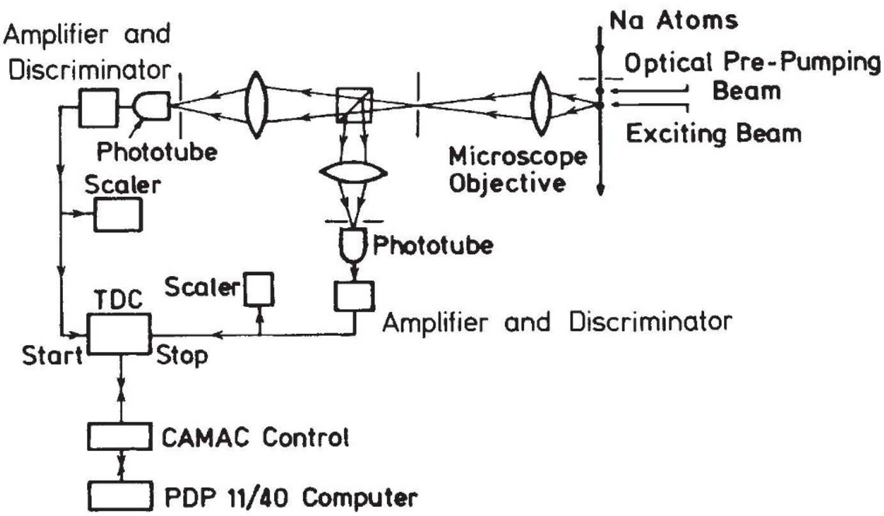

Fig. 3 Experimental arrangement used by Kimble *et al.*[4].

For a thermal field which has a gaussian distribution for $P(E)$ we recover $g^{(2)}(0)=2$ whereas for a coherent field with a stabilised amplitude $P(E)$ is a delta function and hence $g^{(2)}(0)=1$. Hence the Hanbury-Brown and Twiss effect for chaotic fields and indeed for the coherent laser field is adequately described by classical theory. However, since the right hand side of equation (3) is a positive semi-definite quantity, we see that classical theory requires $g^{(2)}(0)\geq 1$ and hence does not allow photon antibunching.

The formulation of the quantum theory closest in formal appearance to the classical theory involves the diagonal expansion of the density operator of the radiation field in terms of coherent states[2,22],

$$\rho=\int P(\mathscr{E})|\mathscr{E}\rangle\langle\mathscr{E}|\,d^2\mathscr{E} \tag{4}$$

where the coherent states $|\mathscr{E}\rangle$ are eigenstates of the positive frequency part of the electromagnetic field

$$E^{(+)}|\mathscr{E}\rangle=\mathscr{E}|\mathscr{E}\rangle \tag{5}$$

with eigenvalue $\mathscr{E}$. The function $P(\mathscr{E})$ does not have the character of a probability distribution since for certain nonclassical states of the radiation field it may be negative or highly singular.

A calculation of $g^{(2)}(0)$ using the P representation yields

$$g^{(2)}(0)-1=\frac{\int P(\mathscr{E})(|\mathscr{E}|^2-\langle|\mathscr{E}|^2\rangle)^2 d^2\mathscr{E}}{\langle|\mathscr{E}|^2\rangle^2} \tag{6}$$

This appears similar in form to the classical expression of equation (3), however, since $P(\mathscr{E})$ may for certain fields which have no classical description take on negative values then $g^{(2)}(0)$ may be less than unity in quantum theory.

While this particular feature of quantum theory was recognised it remained to find a light field which exhibited the property of photon antibunching. It was suggested that certain processes of nonlinear optics for example sub-second harmonic generation[23–26] and two-photon absorption[27–33] would exhibit photon antibunching. To date there has been no experimental verification of these predictions. It was resonance fluorescence from a two-level atom that led to the first experimental observation of photon antibunching.

Resonance fluorescence from a two-level atom

The topic of resonance fluorescence from a two-level atom has been the subject of considerable theoretical and experimental investigation. For weak incident fields the light is coherently scattered, whereas for strong incident fields when the Rabi frequency Ω ($\Omega=2\kappa\mathscr{E}/h$ where $\mathscr{E}$ is the driving field amplitude and κ is the atomic dipole matrix element) exceeds the Einstein A coefficient γ of the atom the spectrum of the scattered light splits into three peaks with the two sidebands displaced from the central peak by the Rabi frequency. This spectrum first predicted by Mollow[34] was verified in detail by the experiments of Wu *et al.*[35] and Hartig *et al.*[36] following an earlier experiment of Schuda *et al.*[37]. These experiments used an atomic beam of sodium atoms which were optically pumped to prepare a pure two-level system. These atoms were then subjected to irradiation from a highly stabilised dye laser tuned to resonance with the atomic transition. This system was to prove of further interest to physicists.

Carmichael and Walls[3] predicted that the second-order correlation function of the light emitted by a single atom undergoing resonance fluorescence would exhibit the property of photon antibunching. This was confirmed in subsequent calculations by Cohen-Tannoudji[38] and Kimble and Mandel[39]. The results obtained by Carmichael and Walls for the second order correlation function $g^{(2)}(\tau)$ in the steady state are

$$g^{(2)}(\tau)=\left[1-\exp\left(\frac{-\gamma\tau}{2}\right)\right]^2 \quad \Omega\ll\gamma \tag{7}$$

$$g^{(2)}(\tau)=\left[1-\exp\left(\frac{-3\gamma}{4}\tau\right)\cos\Omega\tau\right] \quad \Omega\gg\gamma \tag{8}$$

in the limiting cases of very weak and very strong driving fields. The behaviour of $g^{(2)}(\tau)$ is plotted in Fig. 2. This system clearly displays the property of photon antibunching since in both limits $g^{(2)}(\tau)$ starts at zero. For low-intensity driving fields $g^{(2)}(\tau)$ rises monotonically to a background value of unity, whereas for high driving field intensities $g^{(2)}(\tau)$ rises above unity before reaching a steady state value of unity by damped oscillations.

This behaviour can be understood as follows. A measurement of $g^{(2)}(\tau)$ records the joint probability for the arrival of a photon at time $t=0$ and the arrival of a photon at time $t=\tau$. Consider now the driven two-level atom as our source of photons. The detection of a fluorescent photon prepares the atom in its ground state since it has just emitted this photon. The probability of seeing a second photon at $\tau=0$ is zero since the atom cannot re-radiate from the ground state. One must allow some time to elapse so that these may be a finite probability for the atom to be in the excited state and hence a finite probability for the emission of a second photon. In fact $g^{(2)}(\tau)$ is proportional to the probability that the atom will be in its excited state at time τ given that it was initially in the ground state.

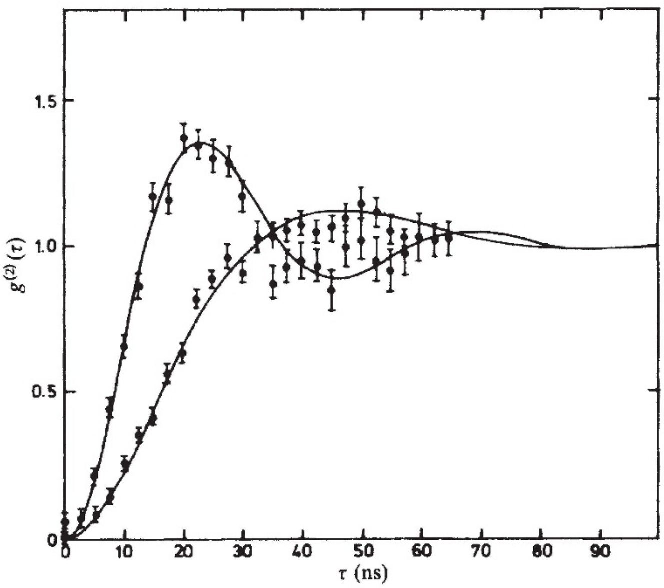

Fig. 4 Photon correlation measurements of fluorescent light from sodium. Experimental results obtained by Dagenais and Mandel compared with theory (solid line). ●, $\Omega/\gamma=2.2$; ○, $\Omega/\gamma=1.1$.

Thus a single atom undergoing resonance fluorescence became a candidate for observing photon antibunching. However, the results (equations (7), (8)) hold only for resonance fluorescence from a single atom. If photons from many atoms contribute to the signal detected then one gets interference effects and the antibunching is diminished and for a large number of atoms lost entirely. In fact for a large number of independently contributing atoms one finds $g^{(2)}(\tau)=2$ in agreement with the central limit theorem[3].

It is clear therefore that an experiment of high sensitivity was necessary if measurements were to be made on the second order correlation function of light from a single atom. Such an experiment was performed by Kimble, Dagenais and Mandel[4]. A sketch of their experimental setup is shown in Fig. 3. In a similar configuration to the experiments which measured the spectrum they used an atomic beam of sodium atoms optically pumped to prepare a pure two-level system. The atomic beam was irradiated at right angles with a highly stabilised dye laser tuned on resonance with the $3P_{3/2}$, $F=3$, $M_F=2$ to $3^2S_{1/2}$, $F=2$, $M_F=2$ transition in sodium. The intensity of the atomic

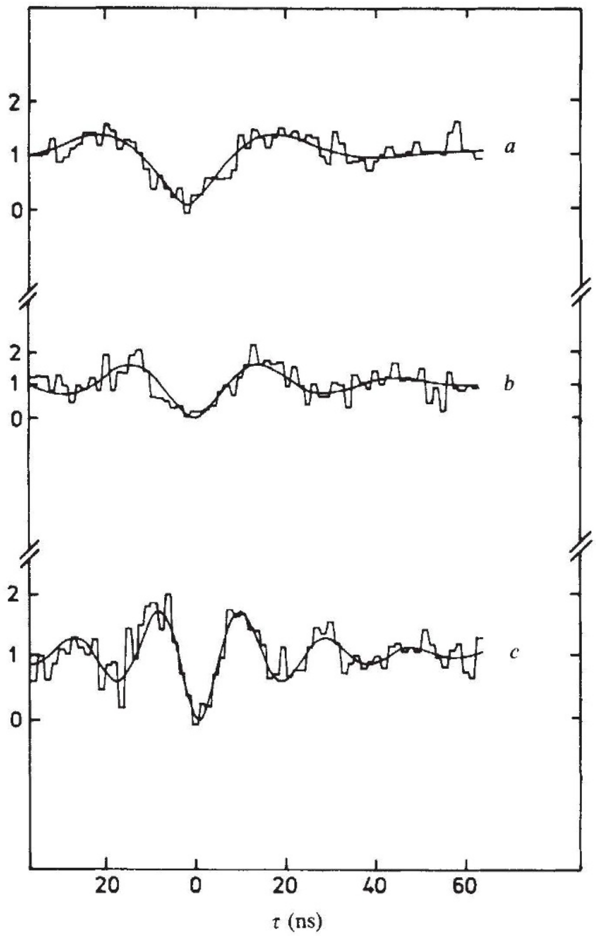

Fig. 5 Photon correlation measurements of fluorescent light from sodium. Experimental points adapted from the results of Leuchs *et al.* (personal communication) compared with theory. *a*, $\Omega/\gamma = 2.2$; *b*, $\Omega/\gamma = 4.3$; *c*, $\Omega/\gamma = 5.7$.

beam was reduced so that on the average no more than one atom is present in the observation region at a time. The fluorescent light from a small observation volume is observed in a direction orthogonal to both the atomic and laser beams.

The fluorescent light in this direction is divided into two equal parts by a beam splitter and the arrival of photons in each beam is detected by two photomultipliers. The pulses from the two detectors are fed to the start and stop inputs of a time to digital converter (TDC) where the time intervals τ between start and stop pulses are digitised in units of 0.5 ns and stored. The number of events $n(\tau)$ stored at address τ is therefore a measure of the joint photoelectric detection probability density which equals $\eta^2 g^{(2)}(\tau)$ where η is the detector efficiency. The initial results obtained by Kimble *et al.*[4] showed the initial positive slope of $g^{(2)}(\tau)$ characteristic of photon antibunching but starting with $g^{(2)}(0) = 1$ rather than zero.

The reason for this disagreement with the theory was pointed out by Jakeman *et al.*[40] who attributed it to number fluctuations in the atomic beam. Though the atomic beam density is such that on the average only one atom is in the observation volume during a correlation time, there are poissonian fluctuations about this mean value so that at times there could be two or more atoms in the observation volume. Thus in the atomic beam experiment one observes the photon antibunching superimposed on the poissonian number fluctuations of the atoms. A calculation including the atomic number fluctuations was carried out by Carmichael *et al.*[41] Similar calculations were made by Kimble *et al.*[5] who obtained very good agreement with experimental results. Some recent experimental results of Dagenais and Mandel[6] are shown in Fig. 4, where when the effects of atomic number fluctuations are taken into account excellent agreement is obtained with the theory. A photon correlation experiment on resonance fluorescence from a beam of sodium atoms has been recently performed by Leuchs *et al.* (personal communication). The results of this experiment are shown in Fig. 5, where with compensation for the atomic number fluctuations excellent agreement is found with the theory. These experiments show clear evidence for the existence of photon antibunching and thus verify the predictions of the quantum theory of light.

Discussion

It could still be argued that one has not observed light with the antibunching property but only inferred from the experimental observation that the fluorescent light from a single atom must possess the antibunching property. This is due to the finite correlation at $\tau = 0$ arising from the number fluctuations of the atoms. It would be useful, therefore, to consider ways of improving experimental methods. Recently advances have been made towards isolating a single atom in an ion trap[42]. This suggests the possibility of performing resonance fluorescence on a single stationary atom which should radiate light with the antibunching character. In addition there are the suggested systems in nonlinear optics for example, sub-second harmonic generation[23–26] and two-photon absorption[27–33] where photon antibunching has been predicted. These experiments involve the nonlinear coupling of electromagnetic waves through a medium which may be in the solid state thereby eliminating problems due to atomic number fluctuations.

Although minor refinements may be possible the experiments performed by Kimble *et al.*[4–6] and Leuchs *et al.* (personal communication) represent a fundamental contribution to our understanding of the nature of light. For the first time photon correlation experiments have detected an effect which is a direct manifestation of the quantum nature of light.

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