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Experimental Evidence for a Photon Anticorrelation Effect on a Beam Splitter: A New Light on Single-Photon Interferences.

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Abstract. – We report on two experiments using an atomic cascade as a light source, and a triggered detection scheme for the second photon of the cascade. The first experiment shows a strong anticorrelation between the triggered detections on both sides of a beam splitter. This result is in contradiction with any classical wave model of light, but in agreement with a quantum description involving single-photon states. The same source and detection scheme were used in a second experiment, where we have observed interferences with a visibility over 98%.

During the past fifteen years, nonclassical effects in the statistical properties of light have been extensively studied from a theoretical point of view [1], and some have been experimentally demonstrated [2-7]. All are related to second-order coherence properties, via measurements of intensity correlation functions or of statistical moments. However, there has still been no test of the conceptually very simple situation dealing with single-photon states of the light impinging on a beam splitter. In this case, quantum mechanics predicts a perfect anticorrelation for photodetections on both sides of the beam splitter (a single-photon can only be detected once!), while any description involving classical fields would predict some amount of coincidences. In the first part of this letter, we report on an experiment close to this ideal situation, since we have found a coincidence rate, on both sides of a beam splitter, five times smaller than the classical lower limit.

When it comes to single-photon states of light, it is tempting to revisit the famous historical «single-photon interference experiments» [8]. One then finds that, in spite of their

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denomination ([1]), none has been performed with single-photon states of light. As a matter of fact, all have been carried out with chaotic light, for which it is well known that quantum second-order coherence properties cannot be distinguished from classical ones, even with a strongly attenuated beam [9]. This is why we have carried out an interference experiment with the same apparatus as used in the first experiment, *i.e.* with light for which we have demonstrated a property characteristic of single-photon states. This single-photon interference experiment is described in the second part of this letter.

Our experimental scheme uses a two-photon radiative cascade dcescribed elsewhere [10], that emits pairs of photons with different ferequencies ν_1 and ν_2. The time intervals between the detections of ν_1 and ν_2 are distributed according to an exponential law, corresponding to the decay of the intermediate state of the cascade with a lifetime $\tau_s = 4.7\,\text{ns}$.

In the present experiment (fig. 1), the detection of ν_1 acts as a trigger for a gate generator, enabling two photomultipliers in view of ν_2 for a duration $w \simeq 2\tau_s$. These two photomultipliers, on both sides of the beam splitter BS, feed singles' and coincidences' counters. We denote N_1 the rate of gates, N_t and N_r the singles' rates for PM_t and PM_r, and N_c the coincidences' rate. Our measurements yield the probabilities for singles' counts during w:

$$p_t = \frac{N_t}{N_1}, \qquad p_r = \frac{N_r}{N_1}, \tag{1a}$$

and the probability for a coincidence

$$p_c = \frac{N_c}{N_1}. \tag{1b}$$

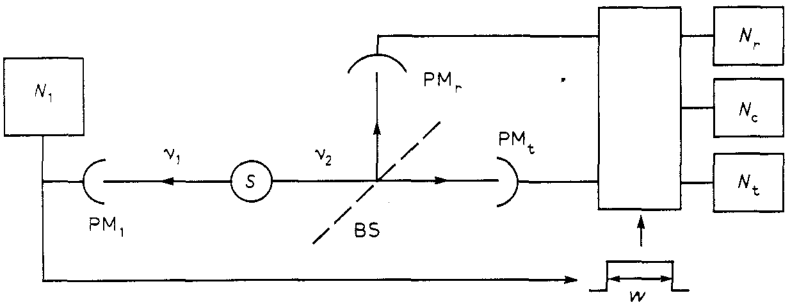

Fig. 1. – Triggered experiment. The detection of the first photon of the cascade produces a gate w, during which the photomultipliers PM_t and PM_r are active. The probabilities of detection during the gate are $p_t = N_t/N_1$, $p_r = N_r/N_1$ for singles, and $p_c = N_c/N_1$ for coincidences.

([1]) Usually, the single-photon character is stated by showing that the amount of energy flowing during a certain characteristic time (coherence time, or time of flight between source and detector) is small compared to $h\nu$. The necessity of the concept of photon is thus postulated, probably on the basis that the detection process appears discrete. But it is well known that this argument is not fully conclusive, since all the characteristics of the photoelectric effect can be assigned to the fact that the «atomic detector is controlled by the laws of quantum mechanics» (see ref. [1], or: W. E. LAMB and M. O. SCULLY, in *Polarisation, Matière et Rayonnement*, ed. Société Française de Physique, Presses Universitaires de France, 1969).

During a gate, the probability for the detection of a photon ν_2, coming from the same atom that emitted ν_1, is much bigger than the probability of detecting a photon ν_2 emitted by any other atom in the source. We are then in a situation close to an ideal single-photon state emission, and we can expect the characteristic behaviour of such a state, *i.e.* an anticorrelation between detections occuring on both sides of the beam splitter.

We calculate now the minimum coincidence rate predicted by a classical wave-description of the experiment of fig. 1, involving the intensity $I(t)$ impinging on the beam splitter. Let us define the time-averaged intensity for the n-th gate, open at time t_n:

$$i_n = \frac{1}{w} \int_{t_n}^{t_n+w} I(t)\,\mathrm{d}t\,. \tag{2}$$

The semi-classical model of photodetection (see footnote ([1])) yields

$$p_\mathrm{t} = \alpha_\mathrm{t} w \langle i_n \rangle\,, \qquad p_\mathrm{r} = \alpha_\mathrm{r} w \langle i_n \rangle\,, \tag{3a}$$

$$p_\mathrm{c} = \alpha_\mathrm{t} \alpha_\mathrm{r} w^2 \langle i_n^2 \rangle\,, \tag{3b}$$

where α_t, α_r are global detection efficiencies, and brackets indicate averages defined over the ensemble of gates:

$$\langle i_n \rangle = \frac{1}{N_1 T} \sum_{n=1}^{N_1 T} i_n\,, \tag{4a}$$

$$\langle i_n^2 \rangle = \frac{1}{N_1 T} \sum_{n=1}^{N_1 T} i_n^2 \tag{4b}$$

(T is the total duration of the experiment).

The standard Cauchy-Schwarz inequality:

$$\langle i_n^2 \rangle \geqslant \langle i_n \rangle^2 \tag{5}$$

holds for our average. Therefore, a classical description of this «triggered experiment» would yield counting rates obeying the inequality

$$p_\mathrm{c} \geqslant p_\mathrm{r} p_\mathrm{t}\,, \tag{6}$$

or equivalently

$$\alpha \geqslant 1 \qquad \text{with} \qquad \alpha = \frac{p_\mathrm{c}}{p_\mathrm{r} p_\mathrm{t}} = \frac{N_\mathrm{c} N_1}{N_\mathrm{r} N_\mathrm{t}}\,. \tag{7}$$

These inequalities mean clearly that the classical coincidence probability p_c is always greater than the «accidental coincidence» probability, which is equal to $p_\mathrm{r} p_\mathrm{t}$. The violation of inequality (7) thus gives an «anticorrelation» criterion, for characterizing a nonclassical behaviour.

The actual values of the counting rates for our experiment are obtained by a straightforward quantum-mechanical calculation. Denoting N the rate of excitation of the cascade, and ε_1, ε_t and ε_r the detection efficiencies (including collection solide angle, optics *transmission, and detector efficiency),* we obtain

$$N_1 = \varepsilon_1 N\,, \tag{7a}$$

$$N_t = N_1 \varepsilon_t (f(w) + Nw)\,, \tag{7b}$$

$$N_r = N_1 \varepsilon_r (f(w) + Nw)\,, \tag{7b'}$$

$$N_c = N_1 \varepsilon_t \varepsilon_r (2f(w)Nw + (Nw)^2)\,. \tag{7c}$$

The quantity $f(w)$, very close to 1 in our experiment, is the product of the factor $1 - \exp[-w/\tau_s]$ (overlap between the gate and the exponential decay in the cascade) by a factor slightly greater than one related to the angular correlation between ν_1 and ν_2 [11].

The comparison of eqs. (7*b*), (7*b*') and (7*c*) clearly shows the anticorrelation: there is a «missing term» $(f(w))^2$ in N_c, related to the fact that a single photon can only be detected once. The quantum-mechanical prediction for α is thus

$$\alpha_{QM} = \frac{2f(w)Nw + (Nw)^2}{(f(w) + Nw)^2}\,, \tag{8}$$

which is smaller than one. The corresponding effect will be strong if Nw can be chosen much smaller than $f(w)$; the experiment is thus designed in order to satisfy this requirement.

The excitation of the atoms is achieved by a two-photon process, using two single-line laser at different frequencies [10]. Several feedback loops control the laser frequencies and intensities, in order to obtain a short- and long-term stability of the excitation rate N within a few percent. The gate w is realized using two time-to-amplitude converters followed by threshold circuits. These «single-channel analysers» are fed by shaped pulses from PM_1 on the START input, and from PM_t or PM_r on the STOP input. The gates corresponding to N_t and N_r can thus be adjusted and superimposed within 0.1 ns. A third time-to-amplitude converter measures the elapsed times between the various detections, and allows a permanent control of the gating system.

The value of w is chosen for a maximum violation of the semi-classical inequality $\alpha \geq 1$, by maximizing the quantity $(1-\alpha)/\sigma_\alpha$, where σ_α is the standard deviation on the measurement of α due to the counting process. This criterion yields $w \simeq 9$ ns.

In fig. 2 the theoretical and experimental values of α are plotted as a function of Nw (see

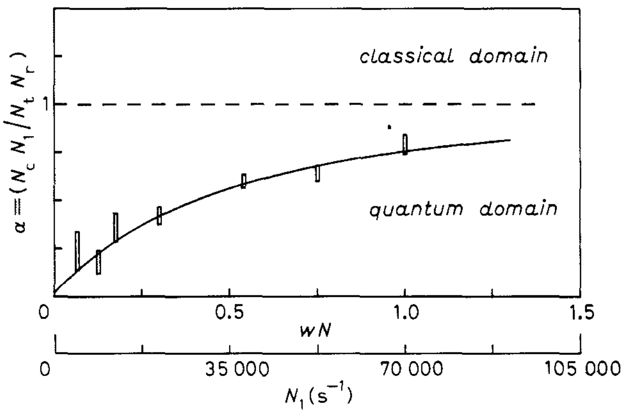

Fig. 2. – Anticorrelation parameter α as a function of wN (number of cascades emitted during the gate) and of N_1 (trigger rate). The indicated error bars are ± one standard deviation. The full-line curve is the theoretical prediction from eq. (8). The inequality $\alpha \geq 1$ characterizes the classical domain.

eq. (8)), or equivalently as a function of the rate of gates $N_1 = \varepsilon_1 N$. A maximum violation of more than 13 standard deviations is obtained for $\alpha = 0.18 \pm 0.06$. For this point, the total counting time was $T \simeq 5$ hours, with $N_1 \simeq 8800\ \mathrm{s}^{-1}$ (including the dark rate $300\ \mathrm{s}^{-1}$), and $N_r \simeq 5\ \mathrm{s}^{-1}$ (dark rate $0.02\ \mathrm{s}^{-1}$). In that case, the number of expected coincidences from the classical theory would be $N_c^{\mathrm{class}} T \geqslant 50$, while we found $N_c^{\mathrm{exp}} T = 9$. Hence the light emitted after each «triggering» pulse has been shown to exhibit a specifically quantum anticorrelation behaviour (2).

By building a Mach-Zehnder interferometer around the beam splitter BS1 (fig. 3), an actual «single-photon» interference experiment can be designed. According to quantum mechanics, the probabilities p_{MZ1} and p_{MZ2} for a detection during the gate in either output of the interferometer are oppositely modulated, as a function of the path difference δ, with a visibility unity.

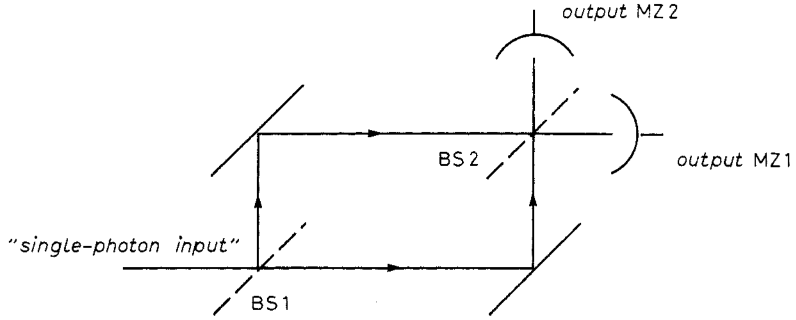

Fig. 3. – Mach-Zehnder interferometer. The detection probabilities in outputs MZ1 and MZ2 are oppositely modulated as a function of the path difference between the arms of the interferometer.

In the actual experiment, the optical system is designed in order to accept the large optical spread of the beam from the source [10] (beam diameter 40 mm for a total divergence 25 mrd), without destroying the visibility of the fringes. This was achieved by observing the fringes in the focal planes of two lenses in view of the outputs MZ1 and MZ2, and working at a path difference around zero.

The two beam splitters BS1 and BS2 are actually two multidielectric coatings on a single $(60 \times 120)\ \mathrm{mm}^2$ silica plate. The planeities of this plate and of the mirrors are close to $\lambda/50$; the orientations are controlled by mechanical stages at about the same precision. The counting rates on both outputs of the interferometer are measured as a function of the path difference δ; δ is varied using a piezo-driven mechanical system, which ensures a parallel translation of the mirror at the required precision.

The interferometer was first checked using light from the actual source, but without any gating system. We found a fringe visibility (3) $V = 98.7\% \pm 0.5\%$, easily reproducible from

(2) A counter experiment has been performed using a pulsed photodiode; the rate N_1 of exciting electrical pulses, and the probabilities $p_t = N_t/N_1$ and $p_r = N_r/N_1$ can be adjusted to the same values than in the actual experiment. But since the light pulse from the diode can be described classically, the expected number of coincidences obeys inequality (7). This point has been verified experimentally in detail.

(3) The fringe visibility is defined by $V = (N_{\mathrm{MZ1}}^{\mathrm{Max}} - N_{\mathrm{MZ1}}^{\mathrm{Min}})/(N_{\mathrm{MZ1}}^{\mathrm{Max}} + N_{\mathrm{MZ1}}^{\mathrm{Min}})$, where $N_{\mathrm{MZ1}}^{\mathrm{Max}}$ and $N_{\mathrm{MZ1}}^{\mathrm{Min}}$ are the maximum and minimum counting rates on output MZ1 when δ is varied (dark rates of the PMTs are subtracted for this calculation).

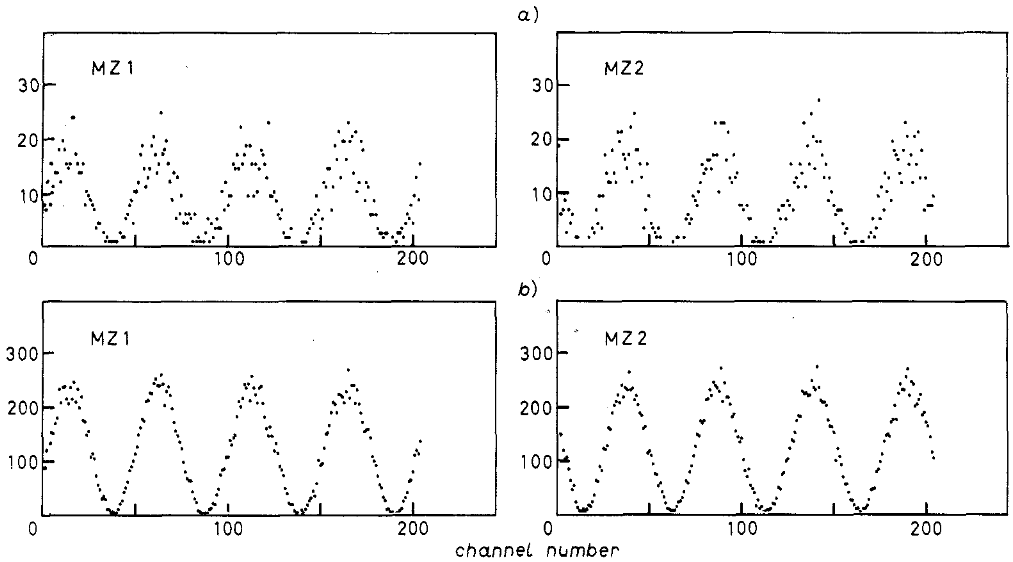

Fig. 4. – Number of counts in outputs MZ1 and MZ2 as a function of the path difference δ (one channel corresponds to a $\lambda/50$ variation of δ). *a*) 1 s counting time per channel *b*) 15 s counting time per channel (compilation of 15 elementary sweeps (like (*a*)). This experiment corresponds to an anticorrelation parameter $\alpha = 0.18$.

day to day within the error limit. In the actual gated experiment, δ was varied around $\delta = 0$ over 256 steps of $\lambda/50$ each, with a counting time of 1 s per step. These sweeps over 5 fringes were stored separately into a computer, then compiled to improve the signal-to-noise ratio. A single sweep and the compiled result for $\alpha = 0.18$ are shown on fig. 4. Several methods of data analysis consistently yielded $V > 98\%$ for any value of α (fig. 5).

Two triggered experiments have thus been performed, using the same source and the same triggering scheme for the detectors. They illustrate the wave-particle duality of light. Indeed, if we want to use classical concepts, or pictures, to interpret these experiments, we must use a particle picture for the first one («the photons are not split on a beam splitter»),since we violate an inequality holding for any classical wave model. On the contrary, we are compelled to use a wave picture («the electromagnetic field is coherently

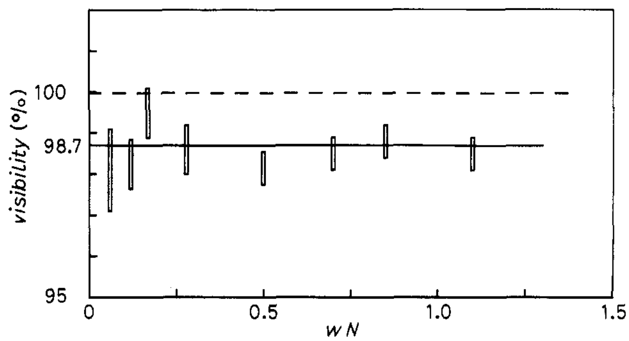

Fig. 5. – Visibility of the fringes in the single-photon regime as a function of wN (compare with fig. 2). A correction (smaller than 0.3%) has been made for dark counts of the PMTs. The estimation of the error bars is conservative.

split on a beam splitter») to interpret the second (interference) experiment. Of course, the two complementary descriptions correspond to mutually exclusive experimental set-ups ([4]).

From the point of view of quantum optics, we will rather emphasize that we have demonstrated a situation with some properties of a «single-photon state». An ideal source of such states would involve the collection of the light at frequency ν_2 in a 4π solid angle, and a shutter triggered by the photons ν_1. One could then carry out many experiments related to nonclassical properties of light, for instance production of sub-Poisson light [12] ([5]).

Although such a scheme can be considered, it would be extremely hard to work out, for practical reasons. Nevertheless, there exists a similar scheme that seems more promising: it consists of pairs of photons emitted in parametric splitting [2, 13, 14]. Due to the phase matching condition, the angular correlation between photons ν_1 and ν_2 is very strong and it becomes possible to produce single-photon states in a single spatial mode.

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([4]) The discussion (and possibly the experiment) can be refined by considering a «quantum nondemolition measurement» of the passage of photons in one arm of the interferometer (N. IMOTO, H. A. HAUS and Y. YAMAMOTO: *Phys. Rev. A*, 32, 2287 (1985) and references therein). Such a device would entail phase fluctuations destroying the interference pattern.

([5]) Instead of the «deletion» scheme proposed in [12], one could also use a feedback loop, activated by ν_1, and reacting on the cascade rate, in order to quiet the Poisson fluctuations in the number of cascades excited in a certain time. See also ref. [14].

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