Observation of Photon Antibunching in Phase-Matched Multiatom Resonance Fluorescence

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We report on the observation of photon antibunching in the resonance fluorescence field radiated by a many-atom source. The crucial feature of our experiment is the achievement of a phase-matching configuration similar to that of four-wave mixing. The antibunching term results from a constructive interference of contributions from the N atoms and thus scales as N^2.

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Photons spontaneously emitted by a single two-level atom are "antibunched": The intensity correlation of the light field is minimum at a null delay.[1,2] This photon antibunching effect has a simple quantum explanation: Just after the emission of a first photon, the atom is in its ground state and then it cannot immediately emit a second photon.[3–5] This effect cannot be explained in a classical description of the fluorescence light and its observation has therefore been considered as experimental evidence of the quantum nature of the electromagnetic field.[6–8] The antibunching effect usually vanishes when the field is radiated by many atoms: The reason is that it is a single-atom effect scaling as the number N of atoms, while the background of coincidences between uncorrelated photons due to multiatom scattering scales as N^2.[9]

In this Letter, we report on the observation of an antibunching behavior in the resonance fluorescence light emitted by a many-atom source. The crucial feature of our experiment is the achievement of a phase matching which allows some constructive interference between the contributions of the N atoms.[10] As a result, the antibunching signal scales as N^2, as does the background. Our phase-matched configuration is sketched in Fig. 1. The atoms are irradiated by two counterpropagating pump lasers and we look for correlations between photons emitted in two opposite directions.[11] We will comment later on the relationship between our configuration and four-wave mixing.

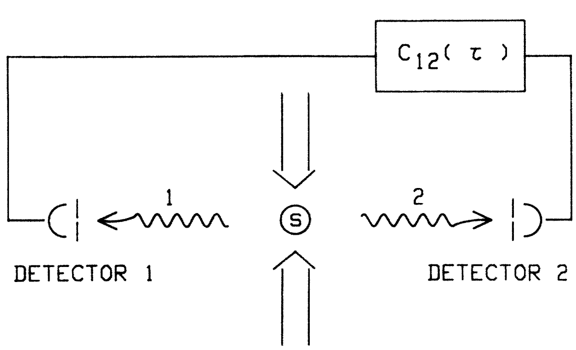

FIG. 1. Experimental scheme. Large arrows indicate counterpropagating laser beams irradiating the atomic source S, and wavy arrows indicate fluorescence photons. $C_{12}(\tau)$ is the intensity correlation between the detection channels 1 and 2, corresponding to exactly opposite directions.

In the experiment, a barium atomic beam (resonance line at 553.5 nm) is irradiated at a right angle by two counterpropagating laser beams from a single-line rhodamine-110 dye laser (standing-wave excitation). The laser and atomic beams have diameters roughly equal to 1 mm. The typical atomic density is $3\times10^9/\text{cm}^3$ which gives a number of atoms in the interaction region $N\cong3\times10^6$ and an optical thickness of the atomic beam about 0.3. The transverse Doppler effect (estimated from the collimation ratio 1:100) is almost equal to the atomic linewidth ($\Gamma/2\pi\cong20$ MHz). The laser frequency is actively stabilized near the atomic resonance (detuning δ).

The fluorescence light is collected in well-defined optical channels with directions perfectly controlled. The stray light from the laser beams is carefully eliminated. The intensity correlation measurements involve coincidence counting electronics including a time-to-digital converter and a multichannel analyzer which yields the time-delay spectrum for two photon detections.

The experimental result for the correlation $C_{12}(\tau)$ between photons emitted in the two opposite directions 1 and 2 (see Fig. 1) is plotted in Fig. 2(a). It reveals a clear antibunching effect: There is a minimum value of $C_{12}(\tau)$ around $\tau=0$. This is very different from the bunching behavior usually associated with multiatomic sources (Hanbury Brown–Twiss effect[12,13]), which was also observed in our experiment by measuring the correlation $C_{11}(\tau)$, between photons emitted in the same direction 1. The experimental result for $C_{11}(\tau)$, given in Fig. 2(b), is obtained by monitoring of the intensity correlation between the two output ports of a beam splitter inserted in beam 1 of Fig. 1.

We have studied the variation of the contrast of the antibunching signal when the holes which limit the detection area are slightly displaced (Fig. 3). The anti-

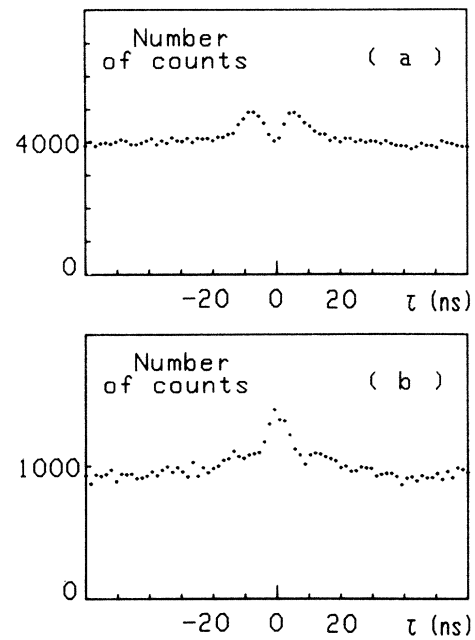

FIG. 2. (a) Intensity correlation C_{12} as a function of the delay τ (1.6 ns per channel). The "hole in the bump" around $\tau=0$ corresponds to the many-atom antibunching effect. (b) Intensity correlation $C_{11}(\tau)$ (Hanbury Brown–Twiss effect). Both curves were obtained for a just-saturating laser power, with a detuning from the resonance $\delta \cong 2\Gamma$. The counting times were 2000 s for (a) and 500 s for (b).

bunching effect disappears when the misalignment angle is more than the coherence angle $\Delta\theta = \lambda/d \cong 0.5$ mrad (where λ is the wavelength of the light and d the transverse dimension of the source). We have thus proved that the effect appears only for a precise phase-matching condition. In order to have nonvanishing counting rates, we have in fact worked with a detection area equal to the coherence area. This smooths the curves of Figs. 2 and 3 and explains the fact that the Hanbury Brown–Twiss effect [Fig. 2(b)] does not reach the ideal peak value equal to 2 times the background.[13]

We want now to explain the major features of the observed correlation signals. Let us first emphasize that the spatial dependence of these signals can be understood in terms of a speckle pattern created by the scattering of a standing wave on randomly distributed scatterers. Indeed, a close examination of the phase factors appearing in the propagation of the fields shows that correlations appear only for the two configurations studied in this paper: photons emitted in the same direction (C_{11}) or photons emitted in opposite directions (C_{12}). In order to explain the time dependence of the signals, we have to use the quantum theory of photodetection.[14] We obtain the following results for

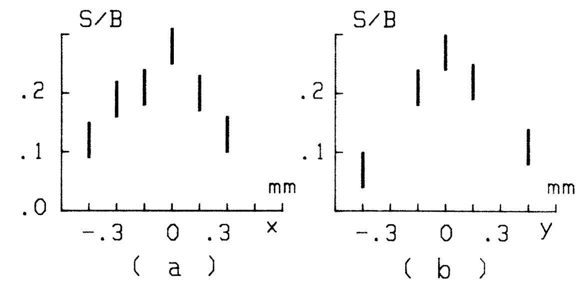

FIG. 3. Ratio between the amplitude S of the antibunching signal (depth of the hole in the bump around $\tau=0$) and the background B as a function of the detector position (detector diameter of 0.6 mm). (a) Corresponds to a horizontal displacement (parallel to the laser beams) and (b) to a vertical displacement (parallel to the atomic beam). The diameter of the coherence area is about 0.55 mm.

$C_{11}(\tau)$ and $C_{12}(\tau)$ [15]:

$$C_{11}(\tau) = I_1^2 + |G_{11}^{-+}(\tau)|^2, \quad (1)$$

$$C_{12}(\tau) = I_1 I_2 + |G_{12}^{++}(\tau)|^2, \quad (2)$$

with

$$I_1 = \mathcal{C}\langle E_1^-(t) E_1^+(t)\rangle, \quad (3)$$

$$G_{11}^{-+}(\tau) = \mathcal{C}\langle E_1^-(t+\tau) E_1^+(t)\rangle, \quad (4)$$

$$G_{12}^{++}(\tau) = \mathcal{C}\langle E_1^+(t+\tau) E_2^+(t)\rangle. \quad (5)$$

(1 and 2 refer to the observation directions discussed above; E^+ and E^- are the positive and negative frequency components of the field operator E; $\mathcal{C}$ is a constant.) The relation (1) is the usual expression for the Hanbury Brown–Twiss effect[13] which gives the intensity correlation $C_{11}(\tau)$ in terms of the intensity I_1 and of the first-order field coherence function $G_{11}^{-+}(\tau)$.[16] The relation (2) for $C_{12}(\tau)$ is very similar but the tested coherence function is now $G_{12}^{++}(\tau)$. This coherence function is less familiar than $G_{11}^{-+}(\tau)$. It has nevertheless been studied since it plays an important role for the problem of squeezing.[17,18] In squeezing experiments,[19] information about G^{++} and G^{-+} is obtained through noise

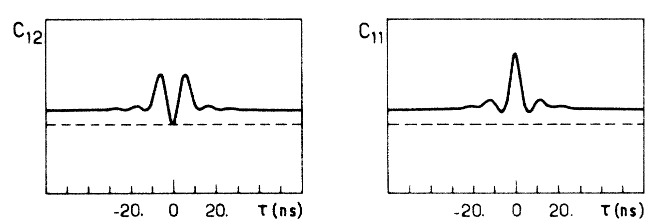

FIG. 4. Theoretical intensity-correlation functions $C_{12}(\tau)$ and $C_{11}(\tau)$, in the case where the Doppler effect can be neglected. Other parameters (detuning, saturation) are set to experimental values. The dashed line corresponds to the background of uncorrelated events.

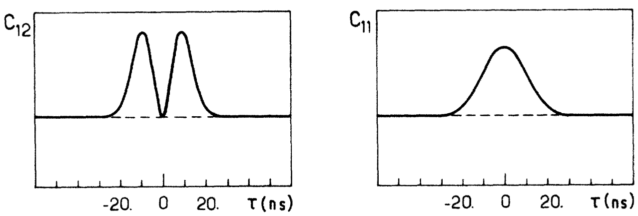

FIG. 5. Theoretical intensity correlation functions $C_{12}(\tau)$ and $C_{11}(\tau)$, in the weak-excitation limit. Other parameters (detuning, Doppler width) are set to experimental values. The dashed line corresponds to the background of uncorrelated events.

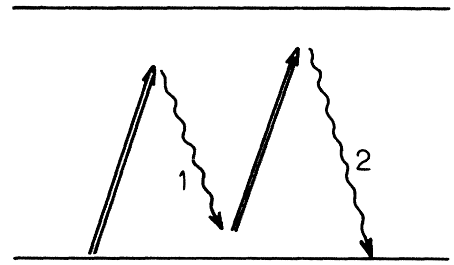

FIG. 6. Diagrammatic representation of the four-wave mixing process responsible for the correlation signal: absorption of two pump photons (large arrows) and spontaneous emission of two fluorescence photons (wavy arrows).

analysis in homodyne detection.[20] The situation is different here since we obtain this information through intensity-correlation measurements in a way similar to the Hanbury Brown–Twiss interferometry.[21]

We can now understand the striking difference between the temporal behaviors of the two signals $C_{12}(\tau)$ and $C_{11}(\tau)$. The field coherence functions can indeed be related to correlation functions of the emitting atomic dipoles. One finds that G_{11}^{-+} is related to $\langle S^+(t+\tau)S^-(t)\rangle$ while $G_{12}^{++}(\tau)$ is related to $\langle S^-(t+\tau)S^-(t)\rangle$ ($S^- = |g\rangle\langle e|$ and $S^+ = |e\rangle\langle g|$, where $|g\rangle$ and $|e\rangle$ denote the ground and excited states of a two-level atom). The quantity $|\langle S^-(t+\tau)S^-(t)\rangle|$ is minimum for $\tau=0$ as a consequence of the operator identity

$$S^-S^- = |g\rangle\langle e|g\rangle\langle e| = 0. \quad (6)$$

We have thus explained the antibunching behavior of $C_{12}(\tau)$ contrasted with the bunching one of $C_{11}(\tau)$ [$|\langle S^+(t+\tau)S^-(t)\rangle|$ is maximum for $\tau=0$].

The calculated temporal behavior of $C_{12}(\tau)$ is shown in Figs. 4 and 5 which correspond to the two marginal situations where either the Doppler effect or the saturation can be neglected. In the first case (Fig. 4), which corresponds to the idealized situation of a perfectly collimated atomic beam, we have used the methods of resonance fluorescence theory[22] in a form adapted to the treatment of multiatom fluorescence.[23] The second marginal situation (Fig. 5) is reached at the limit of a nonsaturating laser excitation. The correlation signals can here be calculated in a perturbative expansion of the field scattered by the atoms.[3,24] The transverse Doppler effect associated with the imperfect collimation of the atomic beam has been included in this approach.[25] It clearly appears on these figures that $C_{12}(\tau)$ is antibunched and $C_{11}(\tau)$ bunched. Moreover, these computed curves are in reasonable agreement with the observed signals of Fig. 2, although they do not include simultaneously the saturation and Doppler effects.

Various sets of curves have been computed for different values of the parameters, in order to select the experimental conditions. We have been led to choose a just-saturating excitation since the contrast of the antibunching signal is strongly affected by saturation. The antibunching signal also disappears when the Doppler width is much greater than the atomic linewidth and than the detuning. Both saturation and Doppler effects have a more dramatic effect when the laser is tuned at exact resonance. This explains why we have obtained our best experimental results for a slightly detuned excitation.

In conclusion, we have observed photon antibunching in the fluorescence field of a many-atom source in a phase-matching configuration identical to the one of four-wave mixing.[26] We can in fact consider that we have studied correlations between the two photons produced by the "spontaneous" four-wave mixing process of Fig. 6. It could be tempting, when looking at this diagram, to predict that the two spontaneous photons are bunched. But there is in fact an infinity of such diagrams corresponding to various frequencies for the fluorescence photons. A proper summation of the contributions of all these diagrams leads to the observed antibunching behavior.[24,25,27] Such an effect can thus be contrasted with the bunching evidenced in other phase-matched nonlinear processes, such as parametric splitting.[28]

Institut d'Optique and Laboratoire de Spectroscopie Hertzienne de l'Ecole Normale Supérieure are Laboratoires associés au Centre National de la Recherche Scientifique.

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