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## Time Correlations between the Two Sidebands of the Resonance Fluorescence Triplet

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A new type of time correlation analysis of resonance fluorescence is presented. A strontium atomic beam is excited by a 28-Å-off-resonance laser. The photons of the two sidebands of the fluorescence triplet are shown to be emitted in a well-defined time order. A simple interpretation of this effect is given which implies a quantum jump of the atom from the lower to the upper state through a multiphoton process.

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Resonance fluorescence (i.e., scattering of radiation by free atoms irradiated by a resonant or quasisonant laser beam) has been extensively studied during the last few years. First, it has been predicted<sup>1</sup> and observed<sup>2</sup> that, for two-level atoms and at high laser intensities, the fluorescence spectrum consists of three components (fluorescence triplet). More recently, the distribution of time intervals between photoelectric counts recorded on the scattered light has been measured, giving evidence for an antibunching of the fluorescence photons originating from a single atom.<sup>3</sup>

These two types of experiments emphasize, respectively, the frequency or time features of resonance fluorescence. One can also consider the possibility of a mixed analysis dealing with the time correlations between fluorescence photons previously selected through frequency filters<sup>4</sup> (the frequency resolution  $\Delta\nu$  introduces, of course, an uncertainty  $\Delta t = (\Delta\nu)^{-1}$  in the determination of the emission time). If, for example, the three components of the fluorescence triplet are well separated (their splitting  $\Omega$  being much larger than their widths  $\gamma$ ), one can use filters centered on any one of these components and having a width  $\Delta\nu$  such that  $\gamma \ll \Delta\nu \ll \Omega$ . With such filters, it is possible to determine which components of the triplet the detected photons are coming from and, simultaneously, to study the

statistics of the emission times with a resolution  $\Delta t$  better than the atomic relaxation time  $\gamma^{-1}$ .

In this Letter, we report the first experimental investigation of time correlations between frequency-filtered fluorescence photons. In this experiment, the detuning  $\delta = \omega_L - \omega_0$  between the laser and atomic frequencies  $\omega_L$  and  $\omega_0$  is much larger than the Rabi nutation frequency  $\omega_1$  (off-resonance excitation) so that the splitting  $\Omega = (\omega_1^2 + \delta^2)^{1/2}$  is simply equal to the detuning  $\delta$ . The three components of the triplet are therefore located at  $\omega_L$  for the central component (Rayleigh scattering) and  $\omega_A = \omega_L + \Omega \approx 2\omega_L - \omega_0$  and  $\omega_B = \omega_L - \Omega \approx \omega_0$  for the two sidebands. The experiment hereafter described shows that the photons of these two sidebands, selected by two filters centered at  $\omega_A$  and  $\omega_B$ , are correlated and emitted in a well-defined order ( $\omega_A$  before  $\omega_B$ ).

We use a strontium atomic beam ( $^1S_0 - ^1P_1$  resonance line;  $\lambda_0 = 460.7$  nm) irradiated by the 28-Å-off-resonance blue line of an argon-ion laser ( $\lambda_L = 457.9$  nm). The multimode-laser light (1 W power) is focused onto the atomic beam (laser-beam waist less than 10  $\mu\text{m}$ ) and focused back by a spherical mirror in order to double the laser intensity in the interaction region. In these conditions, the Rabi nutation frequency  $\omega_1$  is much smaller than the detuning  $\delta$  ( $\omega_1/2\pi = 80$  GHz and  $\delta/2\pi = 4000$  GHz) and the central line of the fluorescence triplet is about  $10^4$  (i.e.,  $4\delta^2/\omega_1^2$ ) times

more intense than the two sidebands (the wavelengths of which are  $\lambda_A = 455.1$  nm and  $\lambda_B = \lambda_0$ ).

The experimental setup (described in more detail elsewhere<sup>4</sup>) is sketched in Fig. 1. The density of the atomic beam can reach  $10^{12}$  atoms per cubic centimeter. The fluorescence light is collected by wide-aperture aspherical lenses ( $f/0.8$ ). Bicone-shaped baffles have been found appropriate to reduce the detected stray light to a level smaller than the Rayleigh fluorescence light. The two sidebands are selected in the two detection channels, respectively, by a grating monochromator tuned on  $\lambda_A$  and an interference filter designed for  $\lambda_B$ . The stray light and the Rayleigh fluorescence light (both at the laser wavelength) are well rejected by the two filters (the transmission at the laser wavelength compared to the maximum transmission is  $3 \times 10^{-4}$  for the monochromator and  $10^{-3}$  for the interference filter). The photomultiplier dark rate is small enough to be ignored. The overall detection efficiency (including collection solid angle, filter transmission, and photomultiplier quantum efficiency) is of one count per 500 emitted photons. The pulses from the two detection channels are amplified and shaped. They then drive a time-to-amplitude converter (TAC) connected to a pulse-height analyzer (PHA) which accumulates the number of detected pairs as a function of the detection separation time. The cable delays are adjusted so that a null separation time yields a point in the middle range of the spectrum which therefore exhibits negative as well as positive values for the delay  $\tau$  between the emission of the two  $\omega_A$  and  $\omega_B$  photons.

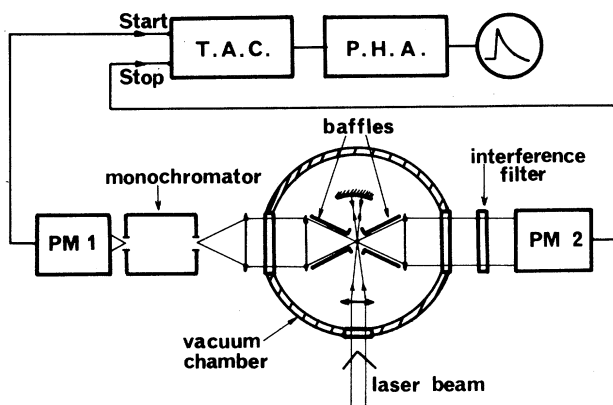


FIG. 1. Experimental setup. The strontium atomic beam is along the axis of the vacuum chamber (PM, photomultiplier; TAC, time-to-amplitude converter; PHA, pulse-height analyzer).

A typical result is shown in Fig. 2(a). The large signal which stands out above an accidental coincidence background indicates that the emissions in the two sidebands are strongly correlated. In addition, the abrupt rising of the signal across the null delay  $\tau = 0$  means that the two correlated photons are emitted in a given time order: the photon in the sideband  $\omega_A$  before the photon in the sideband  $\omega_B$ . Then, the correlation signal decreases exponentially with the delay  $\tau$ . The measured time constant ( $4.7 \pm 0.2$  ns) coincides with the radiative lifetime of the upper level  $^1P_1$  of strontium.<sup>6</sup> The background is mainly due to accidental coincidences between photons at the laser frequency not rejected by the frequency filters.

In order to check the importance of the frequency selection at the sideband wavelength, we have varied the wavelength of the monochromator [Fig. 2(b)]. Shifting it towards the laser wavelength increases the background because of a poorer rejection of the Rayleigh and stray light. The signal exhibits a maximum at the expected wavelength  $\lambda_A$  proving that the signal is due to the sidebands of the triplet. We thus get experimental evidence for the sidebands of the fluorescence triplet in conditions where they are  $10^4$  times weaker than the central component. The excellent signal to background ratio emphasizes the interest of correlation methods since the sidebands could not be easily detected in the frequen-

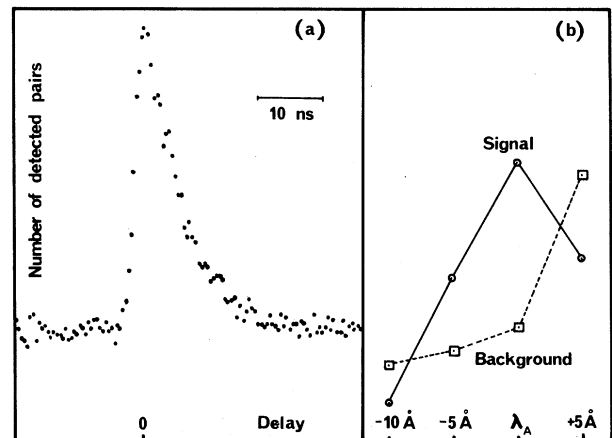


FIG. 2. (a) Typical experimental curve giving the number of detected pairs of photons ( $\omega_A, \omega_B$ ) as a function of the emission delay. The maximum channel height is 1000 counts for an accumulation time of 6 h. The channel width is 0.4 ns. (b) Variation of the signal and background heights when the monochromator wavelength is varied around  $\lambda_A = 4551$  Å.

cy spectrum even with our high-rejection monochromator.

We interpret now these results with a perturbative approach which is valid since the laser is far enough from resonance (the expansion parameter  $\epsilon = \omega_L/2\delta$  is equal to  $10^{-2}$  in our experiment).<sup>7</sup> At the lowest order, we have the elastic-Rayleigh-scattering process [Fig. 3(a)] giving rise to the central component of the triplet at  $\omega_L$  (energy conservation). The rate of this process is  $\Gamma\epsilon^2$  per atom where  $\Gamma$  is the natural width of the upper level  $e$ . It is proportional to the laser intensity  $I_L$ . The two sidebands are explained by a nonlinear scattering process [Fig. 3(b)] involving the absorption of two laser photons and the emission of two fluorescence photons having frequencies  $\omega_\alpha$  and  $\omega_\beta$  linked by the energy conservation relation  $2\omega_L = \omega_\alpha + \omega_\beta$ . An intermediate resonance occurs when the atom reaches the upper state  $e$  after the absorption of the second laser photon. This explains why  $\omega_\alpha$  and  $\omega_\beta$  are distributed in two sharp lines (width  $\Gamma$ ), respectively, centered on  $\omega_A = 2\omega_L - \omega_0$  and  $\omega_B = \omega_0$ . The rate of the whole process is  $\Gamma\epsilon^4$  per atom which means that the weight of each sideband is proportional to  $I_L^2$ . The diagram of Fig. 3(b) suggests to divide the scattering process in two steps. First, the atom jumps from  $g$  to  $e$  by a three-photon process (absorption of  $\omega_L$ , emission of  $\omega_\alpha$ , absorption of  $\omega_L$ ) which takes place during a very short time, smaller than  $10^{-12}$  s in our experiment (inverse of the energy defect  $\delta$  in the nonresonant intermediate states). Then, the atom spontaneously emits  $\omega_\beta$  with a mean lifetime  $\Gamma^{-1}$ .<sup>8</sup>

This picture provides a simple interpretation of

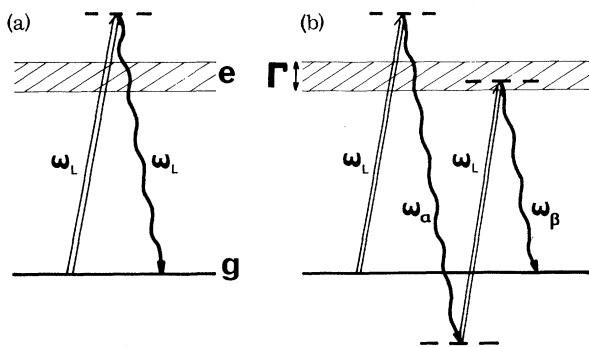


FIG. 3. (a) First-order elastic-Rayleigh-scattering process. (b) Second-order nonlinear scattering process giving rise to the two sidebands of the fluorescence triplet.

all experimental features such as the strong correlation between the photons of the two sidebands (they are emitted by pairs), the time ordering between them, and the exponential decay with a time constant  $\Gamma^{-1}$ . The “percussional” character of the excitation of  $e$  suggests that, if a structure exists in the upper level, the exponential decay of the correlation signal may be modulated at a frequency equal to the spacing between the sub-levels, as in perturbed correlations in atomic or nuclear cascades.<sup>9</sup> Finally, two important features of the experiment can be made clear. First, it is not necessary to use a single-mode laser. The intensity fluctuations do not destroy the correlation signal since their correlation time (inverse of the laser bandwidth) is long compared to the characteristic time of the excitation process  $\delta^{-1}$ .<sup>10</sup> Second, as in the antibunching experiment, the observed correlation signal is due to pairs of photons emitted by the same atom (single-atom effect). Nevertheless, it has been observed with a great number  $N$  of atoms in the field of view ( $N = 2 \times 10^4$ ). As a matter of fact, multiatom effects (mainly Rayleigh-Rayleigh coincidences giving rise to the background and proportional to  $N^2\epsilon^4$ ) could overcome the signal (proportional to  $N\epsilon^4$ ) but they are reduced to a sufficiently low level by the filters which reject the Rayleigh frequency  $\omega_L$ .

As a conclusion for this perturbative discussion we can emphasize the unusual fact that the detection of the first filtered photon is a signature of a quantum jump of the atom from the lower to the upper state through a multiphoton process. Note the difference with the situation where no filters are used (antibunching experiment<sup>3</sup>) and where the detection of one photon is associated with a quantum jump from the upper to the lower state.

Finally, it seems interesting to point out some similarities between the correlation signal described above and those observed in atomic or nuclear cascades  $a-b-c$ . In such a cascade, a pair of photons at different frequencies is also emitted with a given order (the photon associated with transition  $a-b$  before the one associated with  $b-c$ ) and the correlation signal decreases exponentially (with the radiative lifetime of  $b$ ). Actually, the analogy between these two situations is not fortuitous. Resonance fluorescence photons may indeed be considered as photons spontaneously emitted by the combined system of atom plus laser photons interacting together, the so called “dressed atom” which, as a consequence of the quantization of the laser field, has an infinite number of energy levels forming a quasi-

periodic array.<sup>11</sup> A sequence of fluorescence photons therefore appears as a cascade of the dressed atom downwards its energy diagram. Such a picture allows us to interpret the signal here investigated as a radiative cascade signal,<sup>12</sup> where the cascading system is not the bare atom but the dressed one.

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<sup>5</sup>A. Aspect, C. Imbert, and G. Roger, to be published.

<sup>6</sup>D. W. Fahey, W. F. Parks, and L. D. Scheerer, *Phys. Lett.* **74A**, 405 (1979), and references cited therein.

<sup>7</sup>A detailed calculation confirming all the following results will be published elsewhere.

<sup>8</sup>These experimental features might also result from a hyper Raman process bringing the atom from  $g$  to  $e$  through a virtual excitation in a Rydberg state (by the

absorption of two laser photons) and the emission of a  $2\omega_L - \omega_0$  fluorescence photon. But the rate of such a process, evaluated from the corresponding energy defects and oscillator strengths, is shown to be 6000 times smaller than the rate of the process studied in this paper. Moreover, we have experimentally verified that the  $\omega_A$  photons are linearly polarized along the laser polarization, as expected from the diagram in Fig. 3(b). This would not be the case for the hyper Raman process which can bring the atom from  $g$  to any sublevel of  $e$ .

<sup>9</sup>A. M. Dumont, C. Camhy-Val, M. Dreux, and R. Vitry, *C. R. Acad. Sci.* **271B**, 1021 (1970); M. Popp, G. Schäfer, and E. Bodenstedt, *Z. Phys.* **240**, 71 (1970); R. M. Steffen and H. Frauenfelder, in *Perturbed Angular Correlations*, edited by E. Karlsson, E. Matthias, and K. Siegbahn (North-Holland, Amsterdam, 1964).

<sup>10</sup>Furthermore, for the nonlinear process studied here, a multimode laser is more efficient than a single-mode laser with the same intensity by a factor  $\langle I^2 \rangle / \langle I \rangle^2$ , which is equal to 2 for Gaussian fluctuations.

<sup>11</sup>C. Cohen-Tannoudji and S. Reynaud, in *Multiphoton Processes*, edited by J. H. Eberly and P. Lambropoulos (Wiley, New York, 1978), p. 103.

<sup>12</sup>The picture of the dressed-atom cascade remains valid at resonance where a perturbative treatment would no longer be possible at high laser intensities. As shown in Ref. 4, the emissions in the sidebands remain strongly correlated. It must be noted, however, that the correlation signal becomes symmetric.

## Observation of an Optical Stark Effect on Vibrational and Rotational Transitions

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Vibrational and rotational Raman transitions are shown to be shifted to lower frequencies in the presence of a nonresonant, high-intensity optical field. Experimental results using coherent anti-Stokes Raman spectroscopy for hydrogen and nitrogen are presented. The magnitude of the observed shift is in agreement with a calculation which considers the coupling of the optical field to the internuclear separation *via* the molecular electronic polarizability. The magnitude of the shift is predicted to be proportional to the optical field intensity.

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We report the first observation of shifts in molecular vibrational and rotational frequencies induced by an optical field which is not resonant with molecular transition frequencies. The Stark shifts of rotational transitions in polar molecules have been studied extensively using low-frequency applied fields.<sup>1</sup> Changes in transition frequencies due to intermolecular fields have been studied extensively in matrices, liquids, and gas-

es.<sup>2</sup> Apparent spectral shifts that did not involve actual changes in molecular frequencies also have been observed in coherent Raman experiments<sup>3</sup> with use of high-power lasers. The quadratic Stark effect on electronic transitions has been the subject of considerable research.<sup>4</sup> Stark shifts of vibrational frequencies driven near resonance have been investigated,<sup>5</sup> but shifts due to the presence of nonresonant fields