# Destructive Interference between Opposite Time Orders of Photon Emission

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(Received 23 April 1991)

We predict that the intensity correlation between the Rayleigh line and a sideband of the fluorescence triplet displays destructive interference between opposite time orders of photon emission. We have measured this effect in natural barium. To our knowledge such an interference effect has not been observed before in atomic physics.

PACS numbers: 42.50.Dv, 32.80.−t

The study of time correlations between successively detected photons from a weak light source requires measurement of the detection times. The obtained information is in some sense complementary to the information obtained from the observation of the light spectrum. An intermediate situation arises when one measures the correlations between photons originating from various spectral lines. When the delay time between two successively detected photons is at most of the order of the inverse of the frequency width $\gamma$ of the spectrometer, the indeterminacy $\gamma^{-1}$ of the time between emission and detection of a photon allows for the possibility that the first detected photon was actually emitted after the second one. This implies that interference may arise between two opposite time orderings of the emission instants, provided that both orderings contribute to the transition from a single initial to a single final state. For large delay times, the order of emission must be the same as the observed order of detection, and the interference must disappear. Although not uncommon in elementary particle physics, interference between different time orderings has, to our knowledge, not been observed in atomic physics.

In this Letter we predict destructive interference between time orderings appearing in the photon correlation function between the central component and a sideband of the fluorescence triplet of a two-state atom. This interference leads to a vanishing correlation function at zero detection-time difference, and it must disappear when the delay time between the two detection instants is larger than $\gamma^{-1}$. We have observed the predicted interference structure in the spectrally resolved photon correlations of resonance fluorescence of barium.

We recall that the lines in the fluorescence triplet can be understood as spontaneous decay between the dressed states of the atom-field system. Denoting the ground state and excited state of a two-state atom by $|g\rangle$ and $|e\rangle$, and the number of photons in the driving field by $n$, then the dressed states that diagonalize the atom-field Hamiltonian are [1]

$$|1\rangle = c_+|g;n\rangle - c_-|e;n-1\rangle, \qquad |2\rangle = c_-|g;n\rangle + c_+|e;n-1\rangle, \tag{1}$$

where $c_\pm = [(\Omega' \pm \Delta)/2\Omega']^{1/2}$, $\Omega' = (\Omega^2+\Delta^2)^{1/2}$ with $\Omega$ the Rabi frequency and $\Delta$ the detuning of the driving light frequency $\omega_l$ from resonance. These states (1) differ in energy by $\hbar\Omega'$. The fluorescence triplet arises from spontaneous decay down the ladder of pairs $|1\rangle$ and $|2\rangle$ for decreasing values of the photon number $n$, provided that the frequency separation is larger than a typical linewidth [2], which is of the order of the spontaneous decay rate $A$. (The variation of the Rabi frequency with the photon number $n$ may be neglected for an intense driving field.)

The central Rayleigh line situated at the frequency $\omega_l$ results from the transitions $|1\rangle \rightarrow |1\rangle$ and $|2\rangle \rightarrow |2\rangle$, whereas the transitions $|1\rangle \rightarrow |2\rangle$ and $|2\rangle \rightarrow |1\rangle$ produce the sidebands at frequencies $\omega_l \pm \Omega'$. In Fig. 1 the transitions are depicted which produce the lines in the fluorescence triplet. The strengths of the three lines are proportional to squares of transition dipole amplitudes multiplied by the initial-state populations. These amplitudes are simply the matrix elements of the atomic lowering operator $S_- = |g\rangle\langle e|$. The two Rayleigh transitions have amplitudes

$$a_{R1} = \langle 1|S_-|1\rangle = -c_+c_-, \quad a_{R2} = \langle 2|S_1|2\rangle = c_+c_-,$$

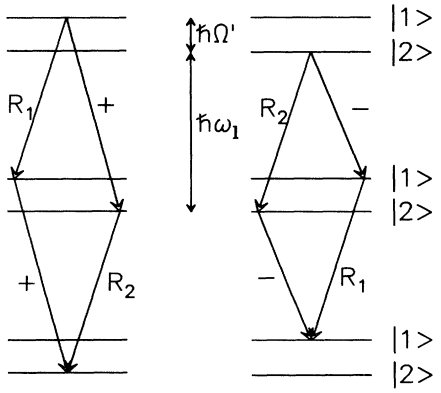


FIG. 1. The two pairs of emission cascades which lead to destructive interference near zero detection-time difference. The transitions which result in the Rayleigh peak are denoted as $R_1$ and $R_2$ for $|1\rangle \rightarrow |1\rangle$ and $|2\rangle \rightarrow |2\rangle$, respectively.

and the amplitudes for the sidebands are

$$a_- = \langle 1|S_-|2\rangle = c_+^2, \quad a_+ = \langle 2|S_-|1\rangle = -c_-^2 .$$

The steady-state populations of the dressed states are

$$p_1 = c_+^4/(c_+^4 + c_-^4), \quad p_2 = c_-^4/(c_+^4 + c_-^4),$$

and the intensities of the three lines, expressed as photon emission rates, are

$$I_- = Ap_2 a_-^2 = I_+ = Ap_1 a_+^2 = Ac_+^4 c_-^4/(c_+^4 + c_-^4),$$

$$I_R = Ap_1 a_{R1}^2 + Ap_2 a_{R2}^2 = Ac_+^2 c_-^2 .$$

The same picture of spontaneous transitions between dressed states can be used to derive expressions for the spectrally resolved correlation functions $F(\alpha,\beta;t)$ [3,4], defined as the joint probability density for observing a photon from the component $\alpha$ at time zero, and a photon from $\beta$ at time $t$ ($\alpha,\beta = +,R,-$). For $t=0$ this picture shows that $F=0$ for $\alpha=\beta=+$, or $\alpha=\beta=-$, demonstrating that photons within a sideband display antibunching. Conversely, for large positive values of $\Delta$ photons from opposite sidebands tend to arrive in pairs, with the photon from sideband + arriving first [3–5]. The corresponding transition cascade is $|1\rangle \rightarrow |2\rangle \rightarrow |1\rangle$. Furthermore, since the emission of a Rayleigh photon does not change the state of the system, it has been concluded that Rayleigh emissions are not correlated with emissions in the sidebands [4].

This picture modifies if we account for interference between successive emissions. Consider the cascades $|1\rangle \rightarrow |1\rangle \rightarrow |2\rangle$ and $|1\rangle \rightarrow |2\rangle \rightarrow |2\rangle$. In Fig. 1 these two cascades are depicted. Both correspond to emission of a photon in the sideband + and a Rayleigh photon, but in opposite time orderings. They correspond to transitions with the same initial and final states of the atom and fluorescence field combined. Moreover, the amplitudes $a_+ a_{R2}$ and $a_{R1} a_+$ add up to zero. This gives the possibility of complete destructive interference. The use of spectrometers with a resolution $\gamma$=FWHM (full width at half maximum) obeying $A \ll \gamma \ll \Omega'$, needed to separate the lines, gives rise to an indeterminacy of the emission times of the order of $\gamma^{-1}$, so that we expect interference to arise for a time difference $t$ between the two detections smaller than $\gamma^{-1}$. A detailed treatment of the filtered fields leaving the spectrometers will be published elsewhere. It gives the result

$$F(+,R;t) = I_+ I_R (1 - e^{-\gamma|t|/2})^2 . \qquad (2)$$

The same result holds for $F(-,R;t)$. This result represents antibunching between a Rayleigh photon and a photon from a sideband. In contrast to the known phenomenon of antibunching [6], which results from the nonemitting nature of the final state of the first emission, in the present case vanishing of (2) for $t=0$ results from the indeterminacy of the intermediate state, and of the time ordering of emission.

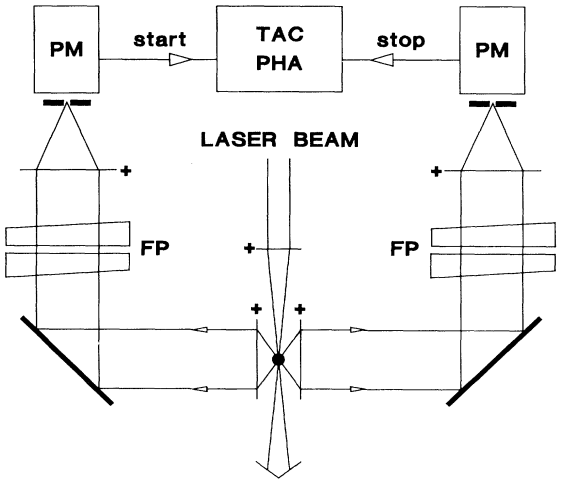


FIG. 2. Experimental setup. FP, Fabry-Pérot spectrometer; PM, photomultiplier; TAC, time-to-amplitude converter; PHA, pulse-height analyzer. The dot between the microscope objectives indicates the position of the atomic beam. The distance between the spectrometers and the atomic beam is 3 m.

The $^1S_0 \rightarrow {}^1P_1$ resonance transition of barium ($\lambda = 553.5$ nm, $A = 1.2\times10^8$ s$^{-1}$) is used for the experiment. In Fig. 2 an overview is given of the experimental setup. An atomic beam is irradiated at approximately right angles by a linearly polarized cw ring dye laser (output power 900 mW). The laser beam is focused on the atomic beam. The focus is placed above the atomic beam in order to obtain a fairly constant Rabi frequency over the interaction volume. Inside the interaction volume Rabi frequencies of up to 5 GHz are achieved. The fluorescence is collected by two microscope objectives (numerical aperture 0.6) placed inside the vacuum chamber at either side of the atomic beam. Two Fabry-Pérot spectrometers, both placed at 3 m from the vacuum chamber, are used to separate the triplet components. The FWHM of a transmission function of one Fabry-Pérot spectrometer is 0.55 GHz and that of the other is 0.82 GHz. Behind the Fabry-Pérot spectrometers an achromatic lens focuses the light beam on a pinhole. In each detection channel a photomultiplier is used to detect the fluorescence light. The photopulses are discriminated by a constant-fraction discriminator. The time differences are measured by a time-to-amplitude converter (TAC). A 0.1-$\mu$s delay cable in the stop channel of the TAC permits the recording of the correlation function at both negative and positive times. Finally, a pulse-height analyzer completes the detection of the time differences of the photopulses. The overall time resolution of the detection electronics is measured to be 3 ns.

In Fig. 3 the number of coincidences between photons from the Rayleigh peak and photons from the lower-frequency sideband are plotted as a function of the time

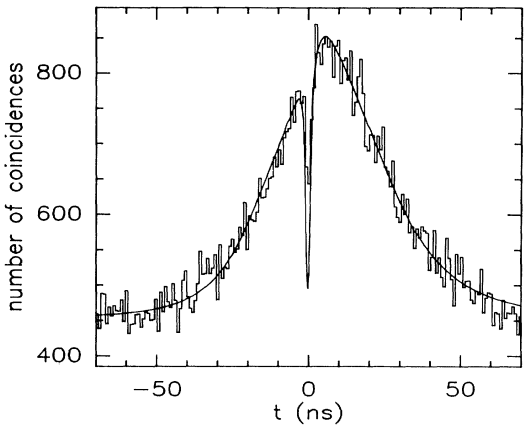


FIG. 3. Intensity correlation spectrum resulting from a measurement in which coincidences are measured between photons from the Rayleigh peak and photons from the lower-frequency sideband; $\Delta = 0.0(3)$ GHz and $\Omega = 4.0(5)$ GHz. The measuring time is $1.6 \times 10^3$ s and the channel width is 0.8 ns. The solid line represents convolution of Eq. (3) with a Gaussian response function.

difference $t$. Since the experiments are performed in a thermal atomic beam the average number of coincidences in a channel of the pulse-height analyzer $\langle n(t) \rangle$ is given by [7]

$$\langle n(t) \rangle \propto \langle N \rangle + F(\alpha,\beta;t)\xi(t)/I_\alpha I_\beta , \quad (3)$$

where $\langle N \rangle$ is the average number of atoms in the observation volume (stray light and dark current are negligible, as can be checked by repeating the measurement with the atomic beam switched off). The value of $\langle N \rangle$ is about 0.8 in the experiment. The transit function $\xi(t)$ describes the conditional probability that a photon is observed from an atom in the region seen by the photomultiplier that triggers the stop of the TAC following a detection of a photon from the same atom in the region seen by the photomultiplier that triggers the start of the TAC. Since these regions do not coincide perfectly in our experiments, $\xi(t)$ is slightly asymmetric. Note that the asymmetry in $\xi(t)$ would still be present if we used a single observation channel that is directed by a beam splitter into two spectrometers. The pinholes in front of the spectrometers would still impede perfect coincidence of the two separate observation regions. For $t \gg \gamma^{-1}$ the correlation function $F(+,R;t)$ is independent of $t$. Therefore, for $t \gg \gamma^{-1}$ the measured correlation function only reflects the behavior of $\xi(t)$. The transit function $\xi(t)$ would be measured by performing coincidence measurements in a situation where the correlation function $F(t)$ is independent of time. This would be the case for correlations between two Rayleigh photons.

Near $t = 0$ the number of measured correlations shows a minimum. The FWHM of the dip represented by (2) is approximately $5\gamma^{-1}$. In our experiments the filter time, $\gamma^{-1}$, is about 0.3 ns. Therefore the calculated width of the interference dip is about half the time resolution of the detection electronics (3 ns). A measurement of this structure thus yields a dip with an amplitude that is half the amplitude of the actual dip, as can be found from a convolution of (2) with a Gaussian response function that models the detection electronics. This is consistent with the depth of the dip we have measured (Fig. 3).

We have also performed an experiment in which one of the Fabry-Pérot spectrometers is tuned to the higher-frequency sideband of the fluorescence triplet and the other to the Rayleigh line. The same structure was measured near $t = 0$, as expected. Correlation experiments in which the spectrometers are tuned to the other combinations of lines in the triplet have also been performed [8]. In these experiments no interference structures have been observed.

It is noteworthy that this interference effect cannot be understood in terms of atomic processes only. As a result of the uncertainty in the time delay between emission and detection, two opposite emission time orders can contribute to the observed time order of detection. Therefore the quantum indeterminacy in the emission order originates in the spectrometers, which are separated from the emitting atom by a macroscopic distance of 3 m. Nevertheless, the spectrometers can be described in a classical way. The interference effect can only be understood in terms of the combined system of the emitting atom and the spectrometers.

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[1] C. Cohen-Tannoudji and S. Reynaud, J. Phys. B **10**, 345 (1977); **10**, 365 (1977).
[2] B. R. Mollow, Phys. Rev. **188**, 1969 (1969).
[3] C. Cohen-Tannoudji and S. Reynaud, Philos. Trans. R. Soc. London A **293**, 223 (1979).
[4] H. F. Arnoldus and G. Nienhuis, J. Phys. B **17**, 963 (1984).
[5] A. Aspect *et al.*, Phys. Rev. Lett. **45**, 617 (1980).
[6] H. J. Kimble, M. Dagenais, and L. Mandel, Phys. Rev. Lett. **39**, 691 (1977).
[7] H. J. Kimble, M. Dagenais, and L. Mandel, Phys. Rev. A **18**, 201 (1978).
[8] C. A. Schrama, Ph.D. thesis, Rijksuniversiteit Utrecht, 1991 (unpublished).