Possibility of observing negative time pair correlation of photons in nonlinear resonance fluorescence

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A type of experiment with an atomic beam for measuring the intensity-fluctuation spectrum of nonlinear resonance fluorescence and the photon-number dispersion is discussed. The conditions for observing photon antibunching, which is characteristic of nonlinear resonance fluorescence, are elucidated.

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A number of experiments on the study of nonlinear resonance fluorescence (NRF) have recently been performed. Grove *et al.*[1] have measured the optical NRF spectrum, and have obtained good agreement with theory. Recently, the observation of photon anti-bunching in NRF in a delayed-coincidence-counting experiment was reported.[2] The experiment was set up in such a way that only a few atoms entered the region of intersection of the atomic beam with the laser beam. Antibunching in NRF is connected with the scattering of light on individual atoms, whereas the quantities contributing to the photocurrent correlator

$$K(\tau)=\frac{1}{2}\langle[J(0),J(\tau)]_+\rangle,$$

which has been measured by Kimble *et al.*,[2] are the constant component $\langle J\rangle^2$, the fluctuations arising as a result of scattering on groups of atoms, and the fluctuations due to the beating of the scattered laser radiation with the NRF of the individual atoms.[3] The indicated components of $K(\tau)$ depend on the various powers of the mean number of atoms in the interaction region, on the dimensions of this region, and on the delay time τ. It is also necessary—for the interpretation of the results of experiment—to take into account the fluctuations in the number of atoms in the interaction region.[4] Thus, the separation of the single-atom effect, which is responsible for the antibunching, in a delayed-coincidence-counting experiment requires the processing of the $K(\tau)$-measurement results on the basis of all the theoretical ideas we have about intensity fluctuations.

Here we assess the possibility of observing the intensity fluctuation spectrum [i.e., the spectrum of the correlatior $K(\tau)$] and the dispersion of the photon-number distribution in an experiment with an atomic beam. We formulate the conditions under which the single-atom effect and the related antibunching effect can be observed independently of the remaining components of the $K(\tau)$ spectrum.

The investigation of negative photon pair correlations is interesting from the theoretical standpoint. Here we have an example of radiation whose fluctuations can be weaker than those of the ideal laser radiation [the dispersion of the photon-number distribution is less than that of the Poisson distribution and the $K(\tau)$ spectrum has dips in a background of shot noise].

For the estimates, we shall use the condition, fulfilled in the experiment by Grove *et al.*,[1] for the mean number, $\bar{N}$, of atoms in the region of intersection of the atomic and laser beams:

$$\bar{N}(\lambda^2/L^2)\approx 1, \tag{1}$$

where λ is the wavelength and L is the linear dimension of the interaction region. If this condition is fulfilled and the observation is carried out a direction, $\mathbf{n}$, significantly different from the direction, $\mathbf{n}_0$, of the laser beam, then we can neglect the effects in the diffraction cone of the incident wave. The photocurrent correlator for the NRF of a system of N noninteracting two-level atoms under conditions of quasimonochromatic excitation can be expressed in terms of the density matrix, $\rho_{ik}(\tau)$, of an atom in a strong field (see Ref. 3) in the form

$$K(\tau)\approx Nei_0\delta(\tau)+Ni_0^2\rho_{22}^{(11)}(\tau)/\bar{\rho}_{22}+N(N-1)i_0^2+N(N-1)e^2q^2\beta(\tau)(\lambda^2/L^2)|K^{(1)}(\tau)|^2. \tag{2}$$

Here $i_0=eq\alpha\gamma\bar{\rho}_{22}$ is the contribution to the photocurrent from the individual atom, e is the electron charge, q is the quantum yield of the photodetector, γ is the radiation constant, and $\bar{\rho}_{22}$ is the steady-state value of the average population of the excited level; the coefficients α and $\beta(\tau)$ are determined by the geometry of the experiment [see also the formula (3)]. The first term in (2) is the single-atom shot noise, the second expresses the effect of successive scattering of two photons by one and the same atom [in $\rho_{22}^{(11)}(\tau)$ the superscripts indicate the initial condition for $\tau=0$]; the next two terms describe the scattering of light by pairs of atoms. In the last term—the two-atom wave noise—the parameter λ^2/L^2 characterizes the solid angle into which the wave vectors of two photons are "drawn" as a result of intereference during scattering on two atoms. If, as in the experiment by Grove *et al.*,[1] the photons are collected in a small solid angle in the direction $\mathbf{n}_\perp(\mathbf{n}_\perp\perp\mathbf{n}_0$, $\mathbf{n}_\perp\perp\mathbf{n}_a$; $\mathbf{n}_a$ is the direction of the atomic beam, $\mathbf{n}_a\perp\mathbf{n}_0)$, then the dominant contribution is made by the wave noise (αN^2). The spectrum of the $K(\tau)$ signal is in this case a convolution of the optical NRF spectrum, which is determined by the first-order correlator $K^{(I)}(\tau)^3$.

Let the photon-collection angle, Ω, contain only the directions, $\mathbf{n}$, for which $|\mathbf{n}-\mathbf{n}_0|\approx 1$ and $|\mathbf{n}-\mathbf{n}_\perp|\approx 1$. The contribution of the wave noise is significantly reduced as a result of the Doppler effect, which, for $\mathbf{n}_0\perp\mathbf{n}_a$, has no effect on the single-atom signal. When allowance is made for the velocity distribution of the atoms in the beam, the coefficient $\rho(\tau)$ in (2) is equal to

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$$\beta(\tau)=\frac{9\Omega}{64\pi^2 d^4}\int_\Omega d\Omega_n|\mathbf{d}-(\mathbf{n}\mathbf{d})\mathbf{n}|^4\langle\langle\exp\{ik_0(v_1-v_2)\tau\cos\theta\}\rangle\rangle. \quad (3)$$

Here $\mathbf{d}$ is the dipole moment of the transition, $\cos\theta = (\mathbf{n}\cdot\mathbf{n}_a)$ and $\langle\langle\ldots\rangle\rangle$ denotes averaging over the velocities. The violation of the interference conditions as a result of the relative displacement of the atoms during the scattering of the pair of photons leads to a situation in which the ratio of the contributions of the wave noise, $G_2(\omega)$, and the single-atom signal, $G_1(\omega)$, for $\omega \gtrsim \gamma$ turns out to be of the order of

$$\frac{|G_2(\omega)|}{|G_1(\omega)|}\approx N\frac{\lambda^2}{L^2}\frac{1}{\Omega}\frac{\gamma}{\delta\omega_D}, \quad \delta\omega_D=k_0\bar{v}. \quad (4)$$

If the direction $\mathbf{n}_\perp$ lies within the photon-collection angle, then there appears in (4) a factor of the order of $\ln(\delta\omega_D/\gamma)$, which changes the estimate little. Under the condition (1), it is sufficient to take the collection angle $\Omega \gg \gamma/\delta\omega_D$ so as to separate out the intensity-fluctuation spectrum of the single-atom component of the NRF. There appears in this spectrum a negative photon pair correlation at times of the order of

$$\tau_c=\min\{\gamma^{-1}, |\nu_0|^{-1}, (2V_0)^{-1}\},$$

where $\nu_0=\omega_0-\omega_{21}$ is the resonance detuning and $V_0 = \mathbf{d}\cdot\mathbf{E}_0/2\hbar$ is the Rabi frequency; dips appear below the shot-noise level.[3] In a strong field with $V_0^2 \gg \gamma^2, \nu_0^2$ the ratio of the spectrum of the single-atom signal to the spectrum of the shot noise is of the order of $q\alpha$. Antibunching of photons in the case of scattering on a single atom is considered in Refs. 5 and 6.

In photon statistics, the antibunching effect—the "repulsion" of the photons during the period of time for which $\tau \lesssim \tau_c$—leads to a reduction in the dispersion, which may turn out to be smaller than for the Poisson statistics. The dispersion of the number of photoelectrons detected in the time interval T is determined with the correlation function $K(\tau)$:

$$\langle\Delta M^2\rangle_T=\frac{2}{e^2}\int_0^T (T-\tau)[K(\tau)-\langle i\rangle^2]d\tau. \quad (5)$$

The term with the δ function in $K(\tau)$ (the shot noise) gives the Poisson contribution $\langle M\rangle_T = Nq\alpha\gamma\bar{\rho}_{22}T$. If for $K(\tau)$ we use the expression (2), then we should take $\langle i\rangle = Ni_0$. Here allowance for the interference terms in $K(\tau)$ and $\langle i\rangle^2$ does not change the result. A relation similar to (4) arises under the above-indicated observation conditions. Finally, we obtain

$$\langle\Delta M^2\rangle_T=\langle M\rangle_T\{1+\xi(T)\}, \quad (6)$$

where

$$\xi(T)=q\alpha\gamma\frac{2}{T}\int_0^T (T-\tau)[\rho_{22}^{(11)}(\tau)-\bar{\rho}_{22}]d\tau.$$

The quantity $\xi(T)$ can be negative; for example, for $T \gg \gamma^{-1}$

$$\xi(T)\approx\xi(\infty)=q\alpha\frac{2V_0^2[\nu_0^2-3\gamma^2/4]}{[\gamma^2/4+\nu_0^2+2V_0^2]^2}; \quad (7)$$

$\xi<0$ for $\nu_0^2<3\gamma^2/4$. If $\nu_0=0$, $\min\xi(\infty)=-3q\alpha/4$ (for $V_0^2=\gamma^2/8$).

In order to eliminate the effect of the fluctuations in the number of atoms in the interaction region, we can measure during a period of time shorter than the time of flight the quantity $M_1^2-M_1M_2$, where M_1 and M_2 are the numbers of photoelectrons detected in the intervals (t_1, t_1+T) and (t_2, t_2+T), where t_1 and t_2 should be such that $|t_2-t_1| \gg \gamma^{-1}$. In this case M_1 and M_2 are realizations of the statistics of independent quantities (with respect to the behavior of the atoms in the field, but at a fixed number, N, of atoms). Therefore, the averaging over a set of such measurements yields $\overline{\langle M^2\rangle_T - \langle M\rangle_T^2}$, where $\langle\ldots\rangle$ denotes quantum-mechanical averaging and the bar denotes averaging over N. This result allows a comparison with the quantity (6) in which N is replaced by $\bar{N}$.

Notice that it is possible to measure all the components of the fluctuation spectrum of NRF in similar experiments.[3]

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Note added in proof (February 21, 1979). In Refs. 7 and 8 further investigation of photon pair correlations in NRF of individual atoms is carried out with allowance for the interference and transit-time effects. The measurement, proposed in the present paper, of the intensity fluctuation spectrum of NRF is an alternative method of studying antibunching of photons. The fluctuations in the number of atoms has little effect on this spectrum for $\tau_s \equiv L/\bar{v} \gg \tau_c$ (they lead to a broadening of the order of τ_s^{-1}). The contribution of the fluctuations in the number of atoms to the dispersion of the number of photocounts is of the order of, or greater than, the contribution of the single-atom effect. A method of detecting the dispersion corresponding to the condition $N \to \text{const} = \bar{N}$ is described above.

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