Statistical properties of nonlinear resonance-diffraction radiation

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It is shown that photon antibunching may manifest itself in the diffraction cone of a directly propagating laser beam under intense resonance excitation of the atomic system, i.e., the photocount variance may fall below the Poisson value and dips may appear in the intensity-fluctuation spectrum against the shot-noise background. The antibunching is associated with the anomalous correlators of the scattered-radiation field, which reflect the evolution of the induced atomic dipole moment and the interference in the scattering of light by groups of atoms. Variants of the proposed experiment are discussed: the observation of heterodyning and self-beating of the scattered radiation in the diffraction cone and the possibility of distinguishing between the contributions from the normal and anomalous field correlators.

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1. In this paper it is shown that in the case of nonlinear resonance scattering of an intense monochromatic light wave by a system of N two-level atoms, photon antibunching is possible in the diffraction cone of the traveling wave. The photocurrent correlator and its spectrum, as well as the variance of the number of photocounts, are investigated. Antibunching in the scattering by an individual atom was predicted in Refs. 1–3. After the experimental confirmation of the theory for $N=1$,[4] intensive searches were undertaken for a macroscopic ($N\gg 1$) source of light with photon antibunching. A review of such searches among the parametric processes of nonlinear optics will be found in Ref. 5. An idealized model—the Dicke system in a resonantor without damping (a conservative system of "N atoms and one field mode") was treated in Ref. 6. In this model, the nonstationary antibunching with periodic vanishing of the variance of the number of photons is associated with complete periodic "transfer back and forth" of energy between the atomic system and the distinguished field mode. Antibunching in the radiation of a macroscopic system has not yet been observed experimentally.

An experimental study of the angular distribution of the intensity in resonance diffraction of laser light by an atomic beam (in connection with the problem of observing atoms and ions in traps) has recently been published.[7] As estimates show, it is also possible to investigate the statistical characteristics of the diffraction component of the radiation in such experiments.

The characteristics of the statistics of the light in resonance diffraction are determined by the normal correlators of the secondary radiation field of the type $\langle E_\alpha^{(-)}(1)E_\beta^{(+)}(2)\rangle$ and by the anomalous correlators $\langle E_\alpha^{(-)}(1)E_\beta^{(-)}(2)\rangle$ and $\langle E_\alpha^{(+)}(1)E_\beta^{(+)}(2)\rangle$ (1 and 2 refer to times and points on the photodetector, and α and β specify the Cartesian components of the field vector). In this case the tendency toward antibunching is associated with the temporal behavior of the anomalous correlators, or more accurately, with the evolution of the atomic dipole moment and the interference due to scattering by groups of atoms that are manifested in these correlators. We recall that "monatomic" antibunching ($N=1$) is associated with the evolution of the population of the excited level. The nontrivial part played by the anomalous correlators in the statistics of radiation has been discussed in a number of papers.[3,8–14]

2. We shall consider the transformation of a monochromatic light wave (of amplitude $\mathbf{E}_0$ and frequency ω_0 with the wave vector $\mathbf{k}_0=(\omega_0/c)\mathbf{n}_0$) by a system of N two-level atoms (with the transition frequency ω_{21} and transition dipole moment $\mathbf{d}$) under the assumption that there are many atoms ($N\gg 1$), that the atoms are optically oriented, and that $|\omega_0-\omega_{21}|\ll\omega_0$. The characteristic dimension L of the region in which the atoms interact with the incident light wave is assumed to be large as compared with the transition wavelength λ ($L\gg\lambda$).[1)] The photocurrent, i.e., the number of photopulses in the observation time T, is recorded at a distance $R\gg L$ in a solid angle Ω near the direction $\mathbf{n}_0$ of the directly transmitted light beam. In this section we assume that the transmitted laser beam itself is excluded.

The variance of the number of photocounts in the observation time T can be expressed in the form

$$\langle\Delta m^2\rangle_T=\int_0^T dt_1\int_0^T dt_2[K(\tau)-\langle I\rangle^2],\qquad \tau=t_2-t_1. \tag{1}$$

Here $K(\tau)$ is the photocurrent correlator under steady-state conditions,

$$K(\tau)=\langle I(0)I(\tau)\rangle, \tag{2}$$

and $\langle I\rangle$ is the average value of the photocurrent in reciprocal seconds. The photocurrent $\langle I\rangle$ includes a term $\langle I_1\rangle\propto N$ due to the scattering of photons by single atoms and a term $\langle I_2\rangle\propto N(N-1)$ due to interference effects in scattering by two atoms, and is essentially determined by the resonance diffraction:

$$\langle I\rangle=\langle I_1\rangle+\langle I_2\rangle=Nq\alpha\gamma\bar\rho_{22}+N(N-1)\,q\beta\gamma\,|\bar\rho_{21}|^2. \tag{3}$$

Here q is the quantum yield of the photodetector, γ is the radiation constant, $\bar\rho_{22}$ is the steady-state population per atom of the upper level 2, and $\bar\rho_{21}$ is the steady-state value of the off-diagonal element of the density matrix of the atom [without the factor $\exp(-i\omega_0 t)$]. The factors α and β in (3)

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are determined by the angular dependence of the intensity of monatomic and diatomic scattering:

$$\alpha=\int_{\Omega} d\mathbf{n}\,F(\mathbf{n}),\qquad |\mathbf{n}|=1, \tag{4a}$$

$$\beta=\int_{\Omega} d\mathbf{n}\,F(\mathbf{n})\,V^{-2}\left|\int_{V} d\mathbf{r}\exp\{ik_0(\mathbf{n}-\mathbf{n}_0)\mathbf{r}\}\right|^2, \tag{4b}$$

$$F(\mathbf{n})=(3/8\pi d^2)\,|\mathbf{d}-\mathbf{n}(\mathbf{nd})|^2, \tag{5}$$

and $V\approx L^3$ is the volume of the interaction region. The factor β reflects the sharp directivity of the diffraction component along $\mathbf{n}_0$. The following estimates are justified by Eqs. (4) and (5):

$$\begin{gathered}\langle I_1\rangle\propto N\Omega,\ \langle I_2\rangle\propto N\varkappa,\\ \varkappa\approx N\Omega\ \text{ for }\ \Omega\leqslant(\lambda^2/L^2),\\ \varkappa\approx N(\lambda^2/L^2)=n\lambda^2L\ \text{ for }\ \Omega>(\lambda^2/L^2).\end{gathered} \tag{6}$$

Here $n=N/V$ is the concentration of atoms. The system is further assumed to be optically thin: $n\lambda^2L\ll 1$.

In the solid angle $\Omega_c=\lambda^2/L^2$ of spatial coherence (the diffraction cone), the random distribution of the atoms does not destroy the time correlations in the scattered radiation due to the evolution of the atoms. As was shown in Ref. 3, for observations in the diffraction cone, the statistical properties of the radiation are determined by the interference between the monatomic scattering and the diffraction component. The corresponding contribution to the photocurrent correlator can be expressed in the form

$$K(\tau)=\langle I\rangle\delta(\tau)+K_1(\tau)+\ldots+K_4(\tau). \tag{7}$$

Here the first term represents the shot noise,[15] and the rest are informative terms corresponding to the various ways of partially factorizing the fourth-order correlator of the scattered-radiation field. The quantity

$$K_1(\tau)\propto\langle E_\alpha^{(-)}(1)E_\alpha^{(+)}(1)\rangle\langle E_\beta^{(-)}(2)\rangle\langle E_\beta^{(+)}(2)\rangle$$

is independent of τ and is proportional to the average intensity of monatomic scattering and to the average intensity of the diffraction component; the term

$$K_2(\tau)\propto\langle E_\alpha^{(-)}(1)E_\beta^{(+)}(2)\rangle\langle E_\beta^{(-)}(2)\rangle\langle E_\alpha^{(+)}(1)\rangle+\{1\leftrightarrow 2\}$$

describes the beating of the monatomic fluorescence with the sharply directional diffraction component; and the term

$$K_3(\tau)\propto\langle E_\alpha^{(+)}(1)E_\beta^{(+)}(2)\rangle\langle E_\alpha^{(-)}(1)\rangle\langle E_\beta^{(-)}(2)\rangle+\text{c.c.}$$

represents similar beating but expressed in terms of the anomalous correlator. The quantities $K_{1,2,3}$ arise from scattering by groups of three atoms and contain the factor $N(N-1)(N-2)$. The term

$$K_4\propto|\langle E_\alpha^{(+)}(1)\rangle\langle E_\beta^{(+)}(2)\rangle|^2,$$

corresponds to complete factorization (scattering by groups of four atoms) and contains the factor $N(N-1)(N-2)(N-3)$.

One can obtain an expression for the quantity $[K(\tau)-\langle I\rangle^2]$ in Eq. (1) in terms of the characteristics of the atomic system by using the methods of Ref. 3, which are based on the diagram technique for nonstationary perturbation theory. The result is

$$\begin{aligned}K(\tau)-\langle I\rangle^2=\langle I_2\rangle\delta(\tau)+\langle I_2\rangle q\varkappa F_0\gamma\{2[\bar\rho_{12}\rho_{21}^{(11)}(\tau)-|\bar\rho_{21}|^2]\\ +\bar\rho_{22}[\rho_{12}^{(12)}(\tau)+\rho_{12}^{(21)}(\tau)\exp\{2i\theta\}]+\text{c.c.}\}.\end{aligned} \tag{8}$$

Here $F_0=F(\mathbf{n}_0)$ and the fact the $\langle I_1\rangle\ll\langle I_2\rangle$ when $\Omega\lesssim(\lambda^2/L^2)$ [see the estimates (6)] is taken into account. The quantities $\rho_{ik}^{(lm)}(\tau)$ are the matrix elements of the solution of the well-known quantum kinetic equations for a two-level system in an external field for the initial condition $\rho_{ik}^{(lm)}(0)=\delta_{li}\delta_{mk}$. The shift of the phase θ of the stationary dipole moment of the atom with respect to the field of the exciting light wave is separated out in (8):

$$\bar\rho_{21}=|\bar\rho_{21}|\exp\{i\theta\},\quad \theta=\operatorname{arctg}[\gamma/2(\omega_{21}-\omega_0)]. \tag{9}$$

Let us write the expression for the variance of the number of photocounts in the form

$$\langle\Delta m^2\rangle_T=\langle m\rangle_T\{1+q\varkappa F_0\xi_T\}, \tag{10}$$

where $\langle m_T\rangle\approx\langle I_2\rangle T$ is the average number of photocounts in the time T (provided $\langle I_1\rangle\ll\langle I_2\rangle$). The quantity ξ_T is determined by the evolution of the density matrix of the atoms. In particular, the contribution from the anomalous correlators vanishes at $\tau=0$. This property reflects the recovery of the dipole moment of the atom at the time $t_2=t_1+\tau$ after the emission of the "first" photon at time t_1. The vanishing of the photocurrent correlator at $\tau=0$ is an unavoidable indication of antibunching; this condition may ensure a stronger indication: $\xi_T<0$ for long observation times.

Let us give the results of calculating ξ_T under the conditions

$$T\gg\max\{\gamma^{-1},\ |\nu_0|^{-1},\ V_0^{-1}\},\ \nu_0=\omega_0-\omega_{21},\ V_0=(|\mathbf{d}\mathbf{E}_0|/2\hbar)$$

for the limiting cases of weak and strong fields:

1) $V_0^2\ll\gamma^2$,

$$\xi_\infty\approx-(32V_0^2/\gamma^2)\sin^4\theta=-\left\{2\gamma^2V_0^2\Big/\left(\frac{\gamma^4}{4}+\nu_0^2\right)\right\}, \tag{11a}$$

2) $V_0^2\gg\gamma^2,\nu_0^2$,

$$\xi_\infty\approx 4\cos^2\theta+\frac{\gamma^2}{V_0^2}=\left\{4\nu_0^2\Big/\left(\frac{\gamma^2}{4}+\nu_0^2\right)\right\}+\frac{\gamma^2}{V_0^2}. \tag{11b}$$

The dependence of ξ_∞ on the parameter $x=V_0^2/\gamma^2$ at $\nu_0=0$ has the form

$$\begin{gathered}\xi_\infty(x;\nu_0=0)=x\left(x-\frac{1}{16}\right)\Big/\left(x+\frac{1}{8}\right)^3,\\ \min_{(x)}\xi_\infty(x;\nu_0=0)\approx-0.25\end{gathered} \tag{12}$$

(for $x\approx 0.02$).

Equation (8) remains valid even in the presence of rapid (transverse) phase relaxation (where the constant $\Gamma\gg\gamma/2$ occurs in place of $\gamma/2$ in the equations for the off-diagonal elements of the atomic density matrix). Then the parameter $\varkappa$ in (6) and (8) is replaced by

$$\varkappa'=(\gamma/\Gamma)\varkappa;\quad \cos^2\theta=\nu_0^2/(\Gamma^2+\nu_0^2).$$

In that case antibunching does not appear for long observation times: $\xi_\infty\geqslant 0$, since

$$(|\bar\rho_{21}|/\bar\rho_{22})\leqslant(\gamma/2\Gamma)$$

[see Eq. (8)]; in particular, the limiting equation (11a) is not applicable. However, the compensation of the Gaussian fluctuations at $\nu_0 = 0$ remains and begins even in a field strong enough to saturate the transition, i.e., when $V_0^2 \gg \gamma\Gamma$ (the condition $V_0^2 > \Gamma^2$ is not necessary).

Of course the condition $n\lambda^2 L \ll 1$ for optical thinness under which the calculation was made greatly limits the magnitude of the antibunching effect [see formulas (6) and (10)]. This limitation can be weakened, however, if we are interested in comparatively low exciting-light intensities corresponding to the greatest manifestation of antibunching [see Eq. (12) and subsequent ones; we are now discussing the case $\Gamma = \gamma/2$]. The linear absorption does not qualitatively alter the effect provided only that the intensity in a large part of the scattering region remains within the range of values for which $\xi_\infty < 0$. Under these conditions, the nonlinear corrections to the absorption are of the order of $\varkappa V_0^2/\gamma^2$, so only the requirement $\varkappa \ll 1$ is necessary.

3. Let us briefly discuss the manifestation of a tendency to antibunching in another variant of the experiment—in the case of heterodyning. Let us assume that beating of the diffraction component of the scattering is observed, either with the directly propagating laser beam, i.e., in directions including $\mathbf{n}_0$, or with a beam diverted from it. As usual, we shall assume the heterodyne signal to be considerably stronger than the investigated secondary radiation of the medium in the diffraction cone, and that it is coherent (i.e., has Poisson statistics). We denote the field of this signal at the surface of the photodetector by $\mathscr{E}_h$, and its phase with respect to the field of the exciting laser light, by φ_h. We shall assume that the wave fronts are concordant at the photodetector surface. In the case of heterodyning under the above conditions, the variance of the number of photocounts is determined by correlators of the form

$$|\mathscr{E}_h|^2\langle E^{(-)}(1)E^{(+)}(2)\rangle,$$

$$|\mathscr{E}_h|^2\exp\{-2i\varphi_h\}\langle E^{(+)}(1)E^{(+)}(2)\rangle(+\text{c.c.});$$

both of these correlators contain factors of the order of $\varkappa$. The expression that replaces (8) in Eq. (1) is

$$K(\tau)-\langle I_h\rangle^2=\langle I_h\rangle\delta(\tau)+\langle I_h\rangle q\varkappa F_0\gamma\{[\bar\rho_{12}\rho_{21}^{(11)}(\tau)-|\bar\rho_{12}|^2]\times[1+\exp\{2i(\theta-\varphi_h)\}]$$
$$+\bar\rho_{22}[\rho_{12}^{(12)}(\tau)+\rho_{12}^{(21)}(\tau)\exp\{2i\varphi_h\}]+\text{c.c.}\}. \quad (13)$$

Here $\langle I_h\rangle$ is the average photocurrent due to the heterodyne signal, i.e., to the transmitted laser light or to some other reference radiation.

As is evident from Eq. (13), the extent to which the photocount statistics deviates from the Poisson formula is independent of the intensity of the heterodyne signal but depends substantially on the phase shift φ_h. If $\varphi_h = 0$, Eq. (13) is the same as Eq.(8) (except for the substitution $\langle I_h\rangle \leftrightarrow \langle I_2\rangle$), so that we obtain the same result for ξ_T as in the case of self beating [in particular Eqs. (11) and (12)]. In analyzing all the radiation escaping into the diffraction cone when $\mathscr{E}_h \approx E_0$ and $\varphi_h \approx 0$ (in the case of an optically thin system), antibunching is found only when there is sufficient detuning from the resonance, and it turns out to be weaker than in the self beating of the diffraction component.

4. The variance of the number of photocounts found above can be measured in an atomic-beam experiment with enough atoms in the interaction region to eliminate the Doppler broadening.[1,2,4,7] An experiment with a thin activated film is also possible; introducing the transverse relaxation constant $\Gamma \gg \gamma/2$ (see Sec. 2) corresponds approximately to taking the effect of the electron-vibrational interaction into account.[16] In performing an atomic-beam experiment for comparison with the results (11) and (12), one can make the measurements and process the data statistically by a method that makes it possible to exclude the effect of particle-number fluctuations in the interaction region, which tends to suppress the appearance of antibunching.[17]

It would evidently be of greater experimental interest to measure the intensity-fluctuation spectrum of the diffraction component, i.e., the spectrum of the correlator $K(\tau)$. This spectrum is less sensitive to particle-number fluctuations (the contour is blurred, the extent of the blurring being of the order of the reciprocal transit time); the spectrum may be measured by the standard techniques of optical mixing spectroscopy. The decisive factor here is the signal-to-noise ratio—the ratio of the spectral density of the informative part of the correlator to the spectral density of the shot noise. As is evident from Eq. (8), when $\omega \approx 0$ (but $\omega \neq 0$) the signal-to-noise ratio is $q\varkappa F_0\xi_\infty$. This signal-to-noise ratio is adequate for observing the spectrum with its dips against the shot-noise background when the antibunching is most clearly manifested under conditions of appreciable but no very strong absorption (see Sec. 2). Let us consider the spectrum and the signal-to-noise ratio in more detail under conditions of saturation. The normal correlator [$\sim\rho_{12}^{(12)}(\tau)$ in Eqs. (8) and (13)] corresponds to the well-known nonlinear reso-

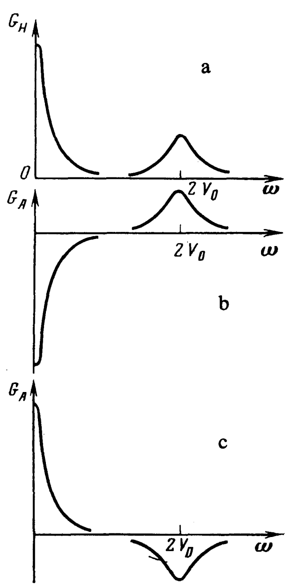

FIG. 1. Components of the photocurrent fluctuation spectrum under saturation conditions, reckoned from the shot-noise level: a—the contribution of the normal correlator, b and c—the contribution from the anomalous correlator (b—for $\theta = \pi/2$ or $\varphi_h = \pi/2$, c—for $\varphi_h = 0$).

nance fluorescence triplet shifted to zero frequency (see case a in Fig. 1). The contribution of the anomalous correlator $[\sim\rho_{12}^{(21)}(\tau)$ in Eqs. (8) and (13)] depends on the shift of the phase θ of the induced dipole moment of the atom (in the case of self beating) or on the phase φ_h of the heterodyne field (in the heterodyne method). When $\theta=\pi/2$ and $\nu_0=0$, or, correspondingly, when $\varphi_h=\pi/2$ and $\nu_0=0$, the anomalous correlations tend to compensate the central component (see case b in Fig. 1). When $\varphi_h=0$ and $\nu_0=0$ only the doubled central component remains (see case c in Fig. 1). As is evident from Eq. (9), the case $\theta=0$ corresponds to $\nu_0=\infty$ and is of no interest. Thus, by altering the detuning of the resonance or the phase of the reference-signal field (in the case of heterodyning) we can alter the character of the self-modulation of the diffraction component of the scattering (or of the modulation of the reference signal). The features of the intensity-fluctuation spectrum mentioned above are retained even in the presence of fast transverse relaxation, and also for a "laser-type" level scheme: resonance at the transition between excited levels with incoherent pumping at one or both of them.[18,19] Let us estimate the signal-to-noise ratio in an atomic-beam experiment under conditions in which the transition is completely saturated. Assume that $\varkappa=0.1$, which is quite adequate for the applicability of Eqs. (8)-(13) and leaves wide possibilities for selecting the intensity of the atomic beam and the dimensions of the interaction region, and let us also set $q=0.2$. Then from (8) we find that the signal-to-noise ratio amounts to 1.4×10^{-2} at the frequency $\omega=2V_0$. Such a value at the entrance to the spectrum analyzer is entirely acceptable for optical mixing methods or for intensity spectroscopy; detailed estimates of the sensitivity of these methods will be found in Ref. 20.

For complete separation of the contributions from the normal and anomalous correlators to the intensity-fluctuation spectrum of the diffraction-scattering component we may suggest heterodyning with the frequency of the reference signal shifted with respect to the frequency of the exciting laser light: $\Delta\omega_h=\omega_h-\omega_0$. In that case the informative part of the photocurrent assumes the character of a periodically nonstationary stochastic process.[21] In the usual method of analyzing the photocurrent spectrum, the spectrum of the normal correlator is shifted by the frequency $\Delta\omega_h$.[22] The contribution from the anomalous correlator reflects the unsteadiness—it contains the factor

$$\exp\{i\Delta\omega_h(t_1+t_2)\}$$

and averages to zero. However, precisely this component is retained provided the circuit of the spectrum analyzer is such that the external signal (i.e., the current of frequency $\Delta\omega_h$) is mixed with the photomultiplier signal after the latter has been transformed by the square-law detector. Let us suppose that the inequalities $\Delta\omega_h\ll\Gamma_f\ll\gamma$ (or Γ) are satisfied, where Γ_f is the damping constant of the high-frequency filter. Then the spectrum of the anomalous correlator is singled out in the subsequent averaging.

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