Intensity-fluctuation spectrum of spontaneous radiation

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The intensity fluctuation spectrum of spontaneous radiation is calculated quantum-mechanically. The wave properties of the radiation as well as the effect of the excitation fluctuations are taken into account. The result agrees with the published experimental data on the noise spectrum of spontaneous radiation, which cannot be explained adequately by the semiclassical method.

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1. INTRODUCTION

To describe certain spontaneous-emission characteristics, such as the average intensity and the average spectrum of the emission, the semiclassical method is quite adequate. In the semiclassical description of spontaneous emission, the radiative transition in the atom is set in correspondence with a classical charged oscillator with corresponding frequency and damping (see, e.g., [1]). The spontaneous emission is then regarded as emission of a damped electromagnetic wave described within the framework of classical electrodynamics.

Recently reported results on the fluctuation and correlation characteristics of spontaneous emission, however, cannot be interpreted on the basis of semiclassical concepts. [2,3] Aleksandrov *et al.* [2] have measured the fluctuation intensity spectrum of spontaneous emission of gas atoms, and the result of the experiment was not compatible with the semiclassical calculation performed in the same paper. The authors of [2] have succeeded in explaining the experimental data by using a probability approach based on quantum concepts concerning emission and absorption of an electromagnetic field. In this approach, the exponential decay of the atomic excitation was interpreted as a time distribution of the probability of observing a photon.

We present here a consistent quantum-mechanical description of an experiment on the measurement of the spontaneous-emission fluctuation spectrum, wherein the conclusion of Aleksandrov *et al.*, [2] namely that the semiclassical method does not agree in principle with quantum theory in this case, is confirmed. In our analysis we take into account the wave properties of the radiation, something that cannot be done in the probability approach.

We note that a problem that is similar from the fundamental point of view is considered by Fano. [4] He investigated quantum-mechanically the interference of the radiation of two atoms excited at the initial instant, an interference that manifests itself in the form of space-time correlations when this radiation is absorbed by two atoms—"counters." In this paper we use the Konstantinov and Perel' diagram method of nonstationary perturbation theory for the density matrix. [5,6] This makes the analysis easy to visualize and facilitates the comparison with the semiclassical method.

2. REGISTRATION OF RADIATION WITH A PHOTODETECTOR

We consider a system of atoms interacting with a quantized electromagnetic field and subjected to a certain external action responsible for the excitation of the atoms. The energy operator H of the system is taken in the form

$$H=H_0+V+W, \quad V=-\sum_i \mathbf{d}_i\mathbf{E}(\mathbf{r}_i), \tag{1}$$

$$\mathbf{E}(\mathbf{r})=\mathbf{E}^+(\mathbf{r})+\mathbf{E}^-(\mathbf{r}),$$

$$\mathbf{E}^-(\mathbf{r})=\sum_{\mathbf{k}\lambda} i\left(\frac{2\pi k}{L^3}\right)^{1/2} \mathbf{e}_{\mathbf{k}\lambda}a_{\mathbf{k}\lambda}{}^+e^{-i\mathbf{k}\mathbf{r}}, \tag{2}$$

where H_0 is the energy operator of the free atoms and of the quantized electromagnetic field, V is the operator of the interaction of the atoms with the quantized electromagnetic field, and W is the operator of the perturbation that leads to excitation of the atoms. Here $\mathbf{d}_i$ is the dipole-moment operator of the i-th atom, $a^+_{k\lambda}$ is the operator for the production of a photon with momentum $\mathbf{k}$ and polarization λ, $\mathbf{e}_{\mathbf{k}\lambda}$ is the unit polarization vector, and L^3 is the quantization volume. $\mathbf{E}^+(\mathbf{r})$ and $\mathbf{E}^-(\mathbf{r})$ denote respectively the positive-frequency and negative-frequency contributions to the intensity operator of the quantized electromagnetic field.

We assume a Doppler distribution in frequency with mean frequency ω_0 and with mean squared deviation ω_D. For the sake of argument we assume that the upper of the two working levels has a total angular momentum I and a radiative width γ, while the lower level represents the ground or metastable state with total angular momentum I'.

The registration of the radiation by the photodetector is described in quantum theory in the following manner. The radiation causes the atoms of the photodetector to go from the ground state to an excited state belonging to the continuous spectrum or else having a larger width than the frequency spectrum of the incident radiation (the Glauber photon-model[7]). We denote by $N(t, s)$ the operator of the number of atoms of the photodetector excited by the light by the instant of time t on a unit area with coordinate s (in the Heisenberg representation). The density of the photocurrent per unit surface is determined by the operator $i(t, s)$, while the total photocurrent by the operator $i(t)$:

$$i(t,s)=\frac{\partial}{\partial t}N(t,s),\quad i(t)=\int ds\, i(t,s), \tag{3}$$

where ds is the photodetector surface element. The radiation-intensity fluctuation spectrum $(i^2)_\omega$ is given in the stationary case by the Fourier transform with respect to the difference between the times, of the correlator $\langle\langle \frac{1}{2}[i(t_1)i(t_2)]_+\rangle\rangle$, (where $[\dots]_+$ is the anticommutator. We shall henceforth use $\langle\dots\rangle$ to denote quantum-mechanical averaging. If the excitation intensity of the atoms in the radiation source is specified as a random function of the time, it is necessary to average over the corresponding random process, an operation designated by a horizontal bar in the absence of quantum-mechanical averaging. Simultaneous averagings (both quantum-mechanical and over the random function of the time) will be designated for brevity by the double angle bracket $\langle\langle\dots\rangle\rangle$.

For isotropic photodetector atoms, the photocurrent density and its correlator are represented, in the lowest order in the interaction between the quantized electromagnetic field with the counter, in terms of the intensity operators in the following manner:

$$\langle\langle i(t,s)\rangle\rangle = q(2\pi\omega_0)^{-1}\sum_{\alpha=x,y,z}\langle\langle E_\alpha^-(t,s)E_\alpha^+(t,s)\rangle\rangle, \tag{4}$$

$$\langle\langle {}^1/_2[i(t_1,s_1)i(t_2,s_2)]_+\rangle\rangle = \delta(s_1-s_2)\delta(t_1-t_2)\langle\langle i(t_1,s_1)\rangle\rangle$$
$$+q^2(2\pi\omega_0)^{-2}\sum_{\alpha,\beta=x,y,z}\{\theta(t_2-t_1)\langle\langle E_\alpha^-(t_1,s_1)E_\beta^-(t_2,s_2)E_\beta^+(t_2,s_2)E_\alpha^+[t_1,s_1)\rangle\rangle$$
$$+\theta(t_1-t_2)\langle\langle E_\beta^-(t_2,s_2)E_\alpha^-(t_1,s_1)E_\alpha^+(t_1,s_1)E_\beta^+(t_2,s_2)\rangle\rangle\}, \tag{5}$$

where q is the photodetector quantum yield, and $\theta(t)$ is the step function (it differs from zero and is equal to unity at $t \geq 0$).

The term of (5) quadratic in the intensity describes the shot noise of the photocurrent, and the fourth-degree terms determine the radiation-intensity fluctuations proper. Formulas (4) and (5) are similar to those obtained by Glauber (see, e.g., [7]) for the registration of free fields. The difference is that (4) and (5) contain intensity operators that are developed in accordance with the Heisenberg representation with Hamiltonian (1).

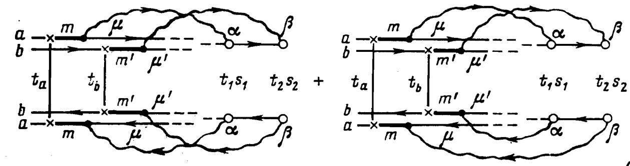
3. DIAGRAM REPRESENTATION OF THE OBSERVABLES

We represent in explicit form the correlator that determines at $t_2 > t_1$ the fluctuation intensities of the spontaneous emission

$$\sum_{\alpha,\beta=x,y,z}\langle E_\alpha^-(t_1,s_1)E_\beta^-(t_2,s_2)E_\beta^+(t_2,s_2)E_\alpha^+(t_1 s_1)\rangle$$
$$=\sum_{\alpha,\beta=x,y,z}\mathrm{Sp}\{E_\beta^+(s_2)U(t_2-t_1)E_\alpha^+(s_1)U(t_1)\rho(0)U^+(t_1)E_\alpha^-(s_1)$$
$$\times U^+(t_2-t_1)E_\beta^-(s_2)\}. \tag{6}$$

Here $U(t)$ is the evolution operator and $\rho(0)$ is the initial density matrix corresponding to the unexcited state of the radiating atoms and the vacuum state of the electromagnetic field. We expand the evolution operator in the lowest orders in the perturbation V and W and select the terms that give a nonzero contribution to (6).

The graphic representation of the mean value of (6) is



(7)

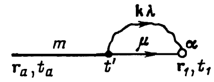
The vertices in (7) are set in correspondence with the interaction-representation matrix elements of the operators V, W, and $E_\alpha^\pm$. The circuit direction marked by the arrows (starting with the point t_2 on the lower line) corresponds to reading the expression under the spur sign in (6) from right to left. The matrix elements of the operators W, V, and the intensity $E_\alpha^\pm$ are marked by a cross, a dot, and a circle, respectively. The wavy line corresponds to the emitted photons. The atoms a and b of the radiating gas are represented by straight lines. The thick atomic line on the segment (t', t'') corresponds to the factor $\exp(-\frac{1}{2}\gamma(t''-t'))$, which describes the radiative damping of the upper level of the atom.

The aggregate of the lines going to the right in the first diagram of (7) describes the amplitude of a process in which the atoms a and b are excited into states with angular momentum projections m and m', and after radiative decay the photon emitted by the atoms a in the transition $m \to \mu$ excites the atom of the counter at the point s_1 and the instant t_1. The photon emitted by the atom b in the transition $m' \to \mu'$ excites a counter atom at the point s_2 at the instant t_2. The aggregate of the lines going to the left in the same diagram describes the complex-conjugate amplitude.

If the perturbation W that excites the atoms has a frequency spectrum that is broad in comparison with γ and ω_D, then the excitation can be described in the balance approximation. The moments of the excitation of the atoms on the upper and lower lines in (7) are then regarded as coinciding (the vertical-tie approximation). A vertical tie is set in correspondence with an excitation intensity $M(t)$ that coincides with the Poisson-averaged number of atoms excited in the volume per unit time. The intensity of the excitation can be a random function of the time.

We integrate in the diagrams integration over the intermediate times, average over the frequencies, coordinates, and angular-momentum projections m and m' over the atoms a and b (henceforth designated $\langle\dots\rangle_{\omega,r}$), and sum over the final angular-momentum projections μ and μ', over the momenta, and over the polarizations of the photons.

Let us separate in the quantum expressions a function analogous to the radiation wave of a classical damped oscillator. To this end, we consider a fragment of the diagram in the form



$\equiv (\mathscr{E}^{\mu m}(t_{1a}, \mathbf{r}_{1a}))_\alpha$, (8)

where $\mathscr{E}^{\mu m}(t_{1a}, \mathbf{r}_{1a})$ is a certain vector, $\mathbf{r}_{1a} = \mathbf{r}_1 - \mathbf{r}_a$, and

$t_{1a} = t_1 - t_a$. A diagram with oppositely directed time corresponds to the complex-conjugate vector.

It follows from (8) that

$$\mathscr{E}^{\mu m}(t_{1a}, \mathbf{r}_{1a}) = \sum_{\mathbf{k}\lambda} \int_{t_a}^{t_1} dt' \, \mathbf{e}_{\mathbf{k}\lambda}(\mathbf{e}_{\mathbf{k}\lambda}\mathbf{d}_{\mu m}) \frac{2\pi k}{L^3}$$

$$\times i \exp\left\{ i\mathbf{k}(\mathbf{r}_1 - \mathbf{r}_a) - \left(\frac{\gamma}{2} + i\omega_a\right)(t' - t_a) - ik(t_1 - t')\right\}. \quad (9)$$

After changing over in (9) to integration in momentum space, it is convenient to use the residue theorem, extending the integration with respect to k over the entire real axis.[1)] As a result we obtain

$$\mathscr{E}^{\mu m}(t_{1a}, \mathbf{r}_{1a}) = \left\{ \mathbf{d}_{\mu m}\left(\frac{k^2}{r_{1a}} + \frac{ik}{r_{1a}^2} - \frac{1}{r_{1a}^3}\right) + \mathbf{n}_{1a}(\mathbf{d}_{\mu m}\mathbf{n}_{1a})\left(-\frac{k^2}{r_{1a}} - \frac{3ik}{r_{1a}^2} + \frac{3}{r_{1a}^3}\right)\right\}$$

$$\times \exp\left\{\left(-i\omega_a - \frac{\gamma}{2}\right)(t_{1a} - r_{1a})\right\}\theta(t_{1a} - r_{1a}), \quad (10)$$

$$k = \omega_a - i\gamma/2, \quad \mathbf{n}_{1a} = \mathbf{r}_{1a}/r_{1a}.$$

It follows therefore that $\mathscr{E}^{\mu m}(t_{1a}, \mathbf{r}_{1a})$ coincides in form with the dipole-radiation wave in classical electrodynamics (see[5]).

If the experimental conditions are such that: a) the linear dimension of the radiating volume is small in comparison with the distance R to the spherical cathode of the counter, b) all the distances are much less than the coherence length ω_D^{-1}, c) the damping γ is much smaller than the carrier frequency ω_a, then it is convenient to choose the following approximation for the wave (10) on the photodetector surface:

$$\mathscr{E}^{\mu m}(t_{1a}, \mathbf{r}_{1a}) = \omega_0^2 R^{-1}\{\mathbf{d}_{\mu m} - \mathbf{n}_{1a}(\mathbf{d}_{\mu m}\mathbf{n}_{1a})\}\exp\{-i\omega_a t_{1a} + i\omega_0 r_{1a} - {}^1/_2\gamma t_{1a}\}\theta(t_{1a}). \quad (11)$$

4. INTENSITY-FLUCTUATION SPECTRUM

The first diagram of (7) determines the spontaneous-emission intensity fluctuations connected with the excitation fluctuations $M(t)$. Using (11), we obtain for the corresponding contribution to the photocurrent correlator

$$\left\langle\!\!\left\langle \frac{1}{2}[i(t_1)i(t_2)]_+ \right\rangle\!\!\right\rangle^{(1)} = \frac{q^2(2\pi\omega_0)^{-2}}{(2I+1)^2}\int ds_1\, ds_2$$

$$\times \int_{-\infty}^{t_1} dt_a \int_{-\infty}^{t_2} dt_b\, \overline{M(t_a)M(t_b)}\left(\sum_{m,\mu}|\mathscr{E}^{\mu m}(t_{1a}, \mathbf{r}_{1a})|^2\right)\left(\sum_{m',\mu'}|\mathscr{E}^{\mu' m'}(t_{2b}, \mathbf{r}_{2b})|^2\right). \quad (12)$$

The sum over the projections of the angular momentum in the usual notation (see[9]) is given by

$$\sum_{m,\mu}|\mathbf{d}_{\mu m} - \mathbf{n}_{1a}(\mathbf{d}_{\mu m}\mathbf{n}_{1a})|^2 = \frac{2}{3}|(I\|d\|I')|^2, \quad (13)$$

and we obtain for the contribution to the photocurrent correlator the final form

$$\left\langle\!\!\left\langle \frac{1}{2}[i(t_1)i(t_2)]_+ \right\rangle\!\!\right\rangle^{(1)} = \frac{q^2 S^2}{(4\pi R^2)^2}\overline{L(t_1)L(t_2)}. \quad (14)$$

We have taken here into account the fact that

$$\gamma = \frac{4}{3}\frac{\omega_0^3}{2I+1}|(I\|d\|I')|^2. \quad (15)$$

S denotes the surface area of the photodector and $L(t)$ denotes the atom excitation intensity smoothed out by the law governing the decay of the atomic excitation:

$$L(t_1) = \int_{-\infty}^{t_1} dt_a M(t_a)\gamma\exp(-\gamma t_{1a}). \quad (16)$$

The smoothing means that the fluctuations of the photocurrent follow the slow (in the scale of γ^{-1}) fluctuations of the excitation and do not respond to the fast excitations, a fact on which the method proposed in[2] for measuring the level width is based.

The second diagram in (7) (with "nondiagonal" arrangement of the photon lines) describes the radiation wave noise produced when the intensities from the different radiators are added on the surface of the photodetector. The analytic expression for this contribution to the photocurrent correlator is

$$\left\langle\!\!\left\langle \frac{1}{2}[i(t_1)i(t_2)]_+ \right\rangle\!\!\right\rangle^{(2)} = \frac{q^2(2\pi\omega_0)^{-2}}{(2I+1)^2}\int ds_1\, ds_2 \int_{-\infty}^{t_1} dt_a \int_{-\infty} dt_b \overline{M(t_a)M(t_b)}$$

$$\times \sum_{m,m',\mu,\mu'}\langle(\mathscr{E}^{\mu' m'}(t_{1b}, \mathbf{r}_{1b})\mathscr{E}^{\mu m}(t_{1a}, \mathbf{r}_{1a}))(\mathscr{E}^{\mu m}(t_{2a}, \mathbf{r}_{2a})\mathscr{E}^{\mu' m'}(t_{2b}, \mathbf{r}_{2b}))\rangle_{\omega, r}, \quad (17)$$

where the averaging over the frequencies and coordinates of the radiating atoms is carried out in the following manner:

$$\langle\exp\{i(\omega_a - \omega_b)(t_2 - t_1)\}\rangle_\omega = \exp\{-(t_2 - t_1)^2\omega_D^2\}, \quad (18)$$

$$\int ds_1\, ds_2\langle\exp\{i\omega_0(r_{2b} - r_{1b} + r_{1a} - r_{2a})\}\rangle_r = S\sigma. \quad (19)$$

The last equation is the usual definition of the coherence area σ on the photodetector surface. If the length of the radiating volume is much larger than the wave length of the light, then $\sigma \ll S$. The contribution to the photocurrent correlator from the counter atoms that are separated by a much larger distance than the linear dimension of the coherence area is then equal to zero. Using this circumstance, we sum over the projections of the angular momentum at coinciding directions from the radiating atoms to the counter atoms, $\mathbf{n}_{1a} = \mathbf{n}_{2a} = \mathbf{n}_{1b} = \mathbf{n}_{2b} = \mathbf{n}$:

$$\sum_{m,m',\mu,\mu'}|(\mathbf{d}_{\mu' m'} - \mathbf{n}(\mathbf{d}_{\mu' m'}\mathbf{n}))(\mathbf{d}_{\mu m} - \mathbf{n}(\mathbf{d}_{\mu m}\mathbf{n}))|^2 = \frac{2}{9}|(I\|d\|I')|^4. \quad (20)$$

Neglecting the damping of the atomic excitation over the small time interval $(t_2 - t_1) \sim \omega_D^{-1}$, we obtain

$$\left\langle\!\!\left\langle \frac{1}{2}[i(t_1)i(t_2)]_+ \right\rangle\!\!\right\rangle^{(2)} = \frac{q^2 S\sigma}{(4\pi R^2)^2}\frac{1}{2}\overline{L^2(t)}\exp(-(t_2 - t_1)^2\omega_D^2). \quad (21)$$

To calculate the contribution of the shot noise it suffices to note that the first diagram of (7) breaks up into the product of two diagrams of lower order, each of which determines the average photocurrent intensity.

The foregoing quantum-mechanical calculation leads to the following form of the spontaneous-emission intensity fluctuation spectrum:

$$\overline{(i^2)}_\omega = qF\left\{1+q\Omega\frac{\gamma^2}{\gamma^2+\omega^2}\frac{\overline{(M^2)}_\omega}{\overline{M}}+q\delta\exp\left(-\frac{\omega^2}{4\omega_D^2}\right)\frac{\overline{L^2(t)}}{2\overline{M}^2}\right\}, \qquad (22)$$

$$\delta=\frac{\sqrt{\pi}}{\omega_D}\frac{\sigma\overline{M}}{4\pi R^2} \qquad \Omega=\frac{S}{4\pi R^2}, \quad \overline{F}=\overline{M}\Omega,$$

where $(\overline{M^2})_\omega$ is the excitation-intensity noise spectrum, Ω is the angle from which the radiation is gathered, $\overline{F}$ is the average flux of the quanta through the photodetector surface, and δ is the spontaneous-emission degeneracy parameter.

The spectrum (22) contains the following: a) the frequency-independent contribution of the shot noise; b) the excitation-intensity fluctuation contribution, which makes it possible to measure the width γ of the atomic level; c) the broad ($\sim\omega_D$) spectral contour of the radiation wave noise. A probability calculation of the intensity-fluctuation spectrum (see[2]) is less general, in the sense that no account is taken there of a fraction of the elementary processes (in the language of our paper, the "interference" second diagram in (7) is neglected). This leads to a loss of the wave properties of the radiation.

Of fundamental interest is the discrepancy between the noise spectrum (22) and the semiclassical-theory result given in[2]. For a comparison we must assume in (22) that the excitation intensity is constant, and omit the fact $\frac{1}{2}$, which is a result of allowance for the wave character of the radiation field, and the contribution of the wave noise. The semi-classical result contains an excessive radiation noise that carries information on the width of the atomic level in the case of a rigorously constant excitation intensity. In quantum theory there is no excess noise, a fact confirmed by experiment with high accuracy.[2]

The excess noise is apparently due to the fact in the semiclassical theory the spontaneous-emission wave of the atom can participate in a number of successive absorption acts and lead to a correlation in the emission of the photoelectrons in times on the order of γ^{-1}. The quantum theory makes it possible to separate a relation similar to the atoms spontaneous-emission wave (see (7) and (8)), but in the latter case the wave must be taken to mean the photon-registration probability amplitude. If the corresponding event (absorption of a photon by the photodetector) is realized, then this amplitude no longer contributes to the succeeding acts of radiation registration.

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[1]If the interaction is chosen in the form $-\mathbf{d}\cdot\mathbf{E}$ and not $-\mathbf{p}\cdot\mathbf{E}$, then the single pole is located at $k=\omega_a-i\gamma/2$, and there is no $k=0$ pole.

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