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Propagation Effect in Resonance Fluorescence: Spatial Antibunching of Photons

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We investigate spatial fluctuations of the cooperative emission of radiation by a two-level–atom system driven by a resonant electric field. For very small absorption length, a directional photon antibunching effect is expected.

Experiments of propagation of intense light pulses in near-resonant gaseous media exhibit spatial fluctuations of the scattered light in the limit of very large atomic densities. These fluctuations are visible on a screen located at the exit and relatively far from the source within a finite spatial extension, more precisely in a ring centered on the propagation axis of the electric field.[1,2]

Two different processes take part in the emission of radiation by a medium of N_c two-level atoms per cm^3. One is the isotropic and incoherent emission of fluorescence, the intensity of which is the sum of the atomic intensities and then is proportional to N_c. The other is the cooperative emission, the intensity of which is proportional to N_c^2 and spatially focused in a small solid angle about the propagation axis. At the space-time point $(\vec{R},t)$ far from the source, these intensities are given by the expectation values of

$$I_{\rm incoh}(R,t)=\frac{1}{R^2}\sum_k R_k^{\,+}(t-|\vec{R}-\vec{x}_k|/c)R_k^{\,-}(t-|\vec{R}-\vec{x}_k|/c) \tag{1}$$

for the incoherent emission and of

$$I_{\rm coop}(\vec{R},t)=\frac{1}{R^2}\sum_{k\neq l}\sum_{l} R_k^{\,+}(t-|\vec{R}-\vec{x}_k|/c)R_l^{\,-}(t-|\vec{R}-\vec{x}_l|/c) \tag{2}$$

for the cooperative emission, where $\vec{x}_k$ is the position of atom with label k in the sample and $R_k^{\,\pm}(t)$ are the raising and lowering operators associated with it, expressed in the Heisenberg picture,

$$R_k^{\,\pm}=|\pm\rangle\langle\mp| . \tag{3}$$

The correlation function of the full intensity, at equal time t and at two observation points $\vec{R}_1$ and $\vec{R}_2$, displays the spatial fluctuations which may be visible on a screen to the naked eye. It is the sum of three relevant terms: The first two terms display the correlations in the intensity of the incoherent emission; they exhibit an isotropic structure. The last term displays the correlations in the intensity of the cooperative emission and is the relevant part of the full correlation function in the small solid angle in which the cooperative emission takes place.

The first one describes the well-known bunching effect.[3,4] For spatial fluctuations it is proportional to

$$\sum_{l\neq k}\sum_{k}\langle R_k^+(t_1^k)R_l^+(t_2^l)[R_l^-(t_2^l)R_k^-(t_1^k)+R_k^-(t_2^k)R_l^-(t_1^l)]\rangle,$$

where $t_{1,2}^j = t - |\vec{R}_{1,2}-\vec{x}_j|/c$ is the retarded time.

Within the traditional far-field approximation, $|\vec{R}_1| \simeq |\vec{R}_2| = R$ and $R \gg |\vec{R}_1 - \vec{R}_2|$, we get[5]

$$G_B^{(2)}(\vec{R}_1,t;\vec{R}_2,t) = \langle I_{\rm incoh}(R,t)\rangle^2\left\{1+\left[2J_1\left(\frac{2\pi a}{\lambda}\frac{|\vec{R}_1-\vec{R}_2|}{R}\right)\left(\frac{2\pi a}{\lambda}\frac{|\vec{R}_1-\vec{R}_2|}{R}\right)^{-1}\right]^2\right\} \tag{4}$$

for a cylindric source with radius a and wavelength λ. J_1 is the first-order Bessel function. Equation (4) displays a spatial bunching of photons for separation $|\vec{R}_1 - \vec{R}_2|$ smaller than the coherence radius δ_c which is the product of the diffraction angle $\lambda/\pi a$ by R. Thus, in pulsed amplified–spontaneous-emission experiments,[6] the intensity detected at R has a granular structure, or speckles of light of dimension δ_c. These speckles are the visible manifestation that the photons bunch together on a distance δ_c.

The second term $G_A^{(2)}(\vec{R}_1,t;\vec{R}_2,t)$ is proportional to $\sum_k\langle R_k^+(t_1^k)R_k^+(t_2^k)R_k^-(t_2^k)R_k^-(t_1^k)\rangle$. It displays the isotropic antibunching effect,[7-9] which reflects the fact that a two-level atom can emit only one photon at time t when jumping from its upper state to its ground state and that any subsequent emission must begin with the atom again in the upper state. It can be measured only in the limit of a single radiating atom since the bunching effect [Eq. (4)] conceals the antibunching effect as the number of atoms increases. A recent experiment[9] confirmed that the photons tend to be separated from one another when they are emitted by a two-level atom driven by a near-resonant field. Patterns of this isotropic antibunching effect do not exist because of the low flux of the light emitted by an atom.

Here we are dealing with the only term of the intensity correlation function which prescribes a preferential direction for the fluxtuations. It is

$$G_{\rm coop}^{(2)}(\vec{R}_1,t;\vec{R}_2,t) = \langle I_{\rm coop}(\vec{R}_1,t)I_{\rm coop}(\vec{R}_2,t)\rangle \tag{5}$$

and it gives a measure of the spatial fluctuations of the cooperative emission of radiation. To date, these fluctuations have been ignored.[7,10] They are actually negligible in the beam core where $\langle I_{\rm coop}\rangle$ is maximum. However, when $\vec{R}_1$ and $\vec{R}_2$ deviate from the central region, the correlation function (5) may become larger than $\langle I_{\rm coop}(\vec{R}_1,t)\rangle\langle I_{\rm coop}(\vec{R}_2,t)\rangle$ for still appreciable values of the intensity. We may develop Eq. (5) further by using Eq. (2) so that

$$G_{\rm coop}^{(2)}(\vec{R}_1,t;\vec{R}_2,t) = \langle I_{\rm coop}(\vec{R}_1,t)\rangle\langle I_{\rm coop}(\vec{R}_2,t)\rangle + \frac{1}{R^4}\sum_{\substack{p\\ p\neq k\neq l}}\left(\sum_{\substack{k,l\\ k\neq l}}[\langle R_k^+(t_1^k)R_l^+(t_2^l)R_p^-(t_2^p)R_p^-(t_1^p)\rangle + {\rm c.c.}] + \sum_k\langle R_k^+(t_1^k)R_k^+(t_2^k)R_p^-(t_2^p)R_p^-(t_1^p)\rangle\right). \tag{6}$$

The last two terms of Eq. (6) give, before the summation over the index p, the probability for atom p to emit two photons separated by a time interval $|t_1^p - t_2^p|$. For the same reasons as for the one-atom antibunching effect, this probability is zero at time coincidence and the photons emitted in such a way tend to be separated from one another. The main feature of this effect lies in the fact that the photons involved in the anitbunching are emitted in a well-defined solid angle because of the cooperation between the atoms. This directivity tends to rule out the spatial antibunching of the photons.

In order to estimate these fluctuations we make some simplifying assumptions. The atoms are in resonance with the driving pulse of duration τ_p. We also assume that $\tau_p \gg T_2$, where T_2 is the inverse of the homogeneous broadening of the line shape, due to the collisions between the atoms and a buffer gas, so that we can only consider stationary solutions. The propagation effect is analyzed in the limit of a coherent plane wave

$$\vec{E} = \vec{x}\mathcal{E}(z)\cos(\omega_0 t - k_0 z), \tag{7}$$

where $\vec{E}$ propagates in the Oz direction. The slowly varying envelope $\mathcal{E}(z)$ is determined by its initial value $\mathcal{E}_0$ and by the reduced Maxwell equation.[11] An antibunching term may then be written as

$$\langle R_k^+(t_1^k)R_k^+(t_2^k)\rangle = \mathfrak{A}_k(t_1^k,t_2^k)\exp[i\omega_0(t_1^k+t_2^k) - 2ik_0 z_k] \tag{8a}$$

with

$$\mathcal{A}(t_1^{\,k},t_2^{\,k}) = -\Omega^2(z_k)\int_{t_2^k}^{t_1^k} dt \int_0^{t_2^k} dt' \exp\left(\frac{t+t'-t_1^{\,k}-t_2^{\,k}}{T_2}\right)\langle R_{3k}(t)R_{3k}(t')\rangle . \tag{8b}$$

The factor $\mathcal{A}_k$ which vanishes at time coincidence ($t_1^{\,k}=t_2^{\,k}$) gives a measure of the antibunching of photons coherently emitted by the atoms. The correlation function of the atomic energy R_{3k} is given by

$$\langle R_{3k}(t)R_{3k}(t')\rangle = \frac{1}{4}\frac{1}{1+[\Omega(z_k)T_2]^2}\{1+[\Omega(z_k)T_2]^2\exp(-|t-t'|/T_2)\cos[\Omega(z_k)|t-t'|] + \Omega(z_k)T_2\exp(-|t-t'|/T_2)\sin[\Omega(z_k)|t-t'|]\} \tag{9}$$

when using the Heisenberg equations and adding phenomenologically the relaxation terms. $\Omega(z)$ is the Rabi frequency at penetration z, defined as the product of the electric dipole moment p with $\mathcal{E}(z)\hbar$.

Let L be the full length of the cylinder and let α^{-1} be the absorption length defined by $\alpha L = T_2\tau_R$. (τ_R is the characteristic time for cooperative emission of radiation and is defined as the inverse of the product of the radiative linewidth, Γ, by $N_c\lambda^2 L$, i.e., $\tau_R^{-1}=N_c\Gamma\lambda^2L$.) As we will show below, large absorption, i.e., $\mathcal{E}(z)=\mathcal{E}_0\exp(-\frac{1}{2}\alpha z)$ with $\alpha L \gg 1$, is required in order that appreciable fluctuations may exist. This implies that αL satisfies the condition[12] $\alpha L \gg (\Omega_0 T_2)^2$ or equivalently $\tau_R T_2 \ll \Omega_0^{-2}$, where Ω_0 stands for $\Omega(0)$.

The spatial antibunching depends on the sum of (8) over the atoms, which becomes in the limit $|Z| \gg L$ with $\vec{R}_{1,2}=(|\vec{X}_{1,2}|\cos\varphi_{1,2}, |\vec{X}_{1,2}|\sin\varphi_{1,2}, Z)$

$$\sum_p \exp[-2ik_0|Z-z_p| - 2ik_0z_p + i(k_0z_p/2Z^2)(\vec{X}_1^{\,2}+\vec{X}_2^{\,2}) - ik_0(\vec{X}_1+\vec{X}_2)\cdot\vec{x}_p/Z]\mathcal{A}_k(\vec{X}_1-\vec{X}_2, Z). \tag{10}$$

The relevant part of $\mathcal{A}_k$ is shown to be only a function of the difference $\vec{X}_1-\vec{X}_2$ when using Eqs. (8b) and (9). It is

$$\mathcal{A}_k = \tfrac{1}{4}\{1-\cos[\Omega(z_k)(\vec{X}_1-\vec{X}_2)\cdot\vec{x}_k/cZ]\} \tag{11}$$

in the limit $\Omega(z_k)T_2 \gg 1$ and

$$\mathcal{A}_k = \tfrac{1}{4}[\Omega(z_k)T_2]^2\{1-\exp[-|(\vec{X}_1-\vec{X}_2)\cdot\vec{x}_k/cT_2Z|]\} \tag{12}$$

for $\Omega(z_k)T_2 \ll 1$.

The phase $2k_0|Z-z_k| + 2k_0z_k$ is independent of the atomic positions only for detection in the direction of propagation of the driving field ($Z>0$). Possible detection in the opposite direction is inhibited by interferences. The antibunching effect in the backward direction will be negligible if $8k_0/\alpha \gtrsim 1$. The high directivity of the cooperative emission of radiation provides the condition $|\vec{X}_1+\vec{X}_2| < \lambda Z/a$, which cancels the antibunching factor $\mathcal{A}_k$ since ω_0 is much larger than $\Omega(z)$ and T_2^{-1}. The only conjecture which holds simultaneously for the directivity and the antibunching is $\vec{X}_1 = -\vec{X}_2$. The last phase factor $\exp(ik_0z_kX^2/Z^2)$, with $|\vec{X}_{1,2}|=X$, would prescribe in the absence of attenuation of the incident field that the fluctuations may arise only in solid angle approximately $(k_0L)^{-1}$ so that the condition $X/Z \gtrsim cT_2/2a$ or $X/Z \gtrsim c/2a\Omega_0$ required by $\mathcal{A}_k$ would be met only for $\pi a^2/\lambda L$ much larger than ω_0/Ω_0 or ω_0T_2, i.e., for extremely large Fresnel numbers. This circumstance explains why large fluctuations of the cooperative emission may be important only for large absorption, i.e., generally for high atomic densities. Indeed, if $\alpha^{-1} \ll L$ is the effective length, the condition $X/Z < (k_0/\alpha)^{-1/2}$ may be consistent with the other conditions required for X/Z. Finally we get

$$G_{\rm coop}^{(2)}(X,Z) = \langle I_{\rm coop}(X,Z)\rangle^2[1+\eta(X/Z)/2\mathcal{R}(X/Z)]^2 . \tag{13}$$

Here $\mathcal{R}$ is the ratio between the intensity of the cooperative emission and the intensity of the incoherent emission at location $\vec{R}=(\vec{X}, Z\vec{z})$,

$$\mathcal{R} = 8\bar{N}[J_1(k_0aX/Z)/(k_0aX/Z)]^2 , \tag{14}$$

where $\bar{N}=N_cL\alpha^{-1}=a^2/\Gamma T_2\lambda^2$ is the effective number of atoms. η is the sum of $\mathcal{A}_k$ over the atoms located on a section, with

$$\eta \simeq 1 - 2J_1(2Xa\Omega_0/cZ)/(2Xa\Omega_0/cZ), \qquad \Omega_0T_2 \gg 1; \tag{15}$$

$$\eta = 1 - (\pi a^2)^{-1} d\vec{s}\exp(-2|\vec{X}\cdot\vec{s}|/cT_2Z), \qquad \Omega_0T_2 \ll 1. \tag{16}$$

Let us summarize the conditions for $G_{\rm coop}^{(2)}$ to be larger than $\langle I_{coop}\rangle^2$:

The conditions for large absorption are

(I) $(\Omega_0\tau_R)^2 \ll \tau_R/T_2 \ll 1$.

The positions of the two detectors are subject to

(IIa) $Z > 0$ (no backward detection for $8k_0/\alpha \gtrsim 1$),

(IIb) $\vec{X}_1 = -\vec{X}_2$,

(IIc) $X/Z \lesssim (k_0/\alpha)^{1/2}$,

(IId) $X/Z < k_0 a(8N/\pi)^{1/3}$,

for $\mathcal{R}$ to be larger than unity.

In the limit for large N_c as required by (I), the last inequality (IId) is stronger than (IIc).

The condition (III) $X/Z > c/a\Omega_0$ for $\Omega_0 T_2 \gg 1$ or $X/Z \rangle cT_2/a$ for $\Omega_0 T_2 \ll 1$ ensures that $\eta(X/Z)$ is of order unity. It implies that the distance $2X$ between the two detectors has to be greater than the antibunching length $2\delta_a$ defined by the relation

$$\delta_a = Zc/a\Omega_0, \quad \Omega_0 T_2 \gg 1; \qquad \delta_a = ZcT_2/a, \quad \Omega_0 T_2 \ll 1. \tag{17}$$

Note that δ_a is much larger than the coherence radius δ_c for the photon bunching ($\delta_a/\delta_c = \omega_0/\Omega_0$ or $\omega_0 T_2$). Then the possible spatial antibunching effect can be never masked by the bunching effect.

Finally in the limit $\Omega_0 T_2 \gg 1$ we find that a spatial antibunching of photons can be expected in a ring with radii $Zc/a\Omega_0$ and $k_0 aZ\ (8\bar{N}/\pi)^{1/3}$. The normalized correlation function

$$g_{coop}^{(2)} = [1 + 1/2\mathcal{R}(X/Z)]^2 \tag{18}$$

increases from a value slightly above unity to approximately 2 when X increases from $Zc/a\Omega_0$ to $k_0 aZ(8\bar{N}/\pi)^{1/3}$. For example, with $\lambda = 5\times10^{-5}$ cm, $a = 5\times10^{-2}$ cm, $L = 3$ cm, $\Omega_0 = 6\times10^{12}$ s^{-1}, $T_2 = 10^{-11}$ s, $N_c = 5\times10^{15}$ cm^{-3}, and $\Gamma = 10^8$ s^{-1}, we get $\bar{N} = 10^9$, $\tau_R^{-1} = 5\times10^{15}$ s^{-1}, and $\delta_a = 0.1Z$. From these values it follows that the conditions (I) and (II) are met. $g_{coop}^{(2)}$ varies from 1.04 to approximately 2 as X/Z increases from 0.1 to 0.17 and $\mathcal{R}$ decreases from 10 to 1.

In conclusion, spatial fluctuations of the intensity of the cooperative emission are expected when $\langle I_{coop}\rangle$ is quantatively nearer to $\langle I_{incoh}\rangle$ than to $\langle I_{coop}\rangle_{max}$ and then loses its quasicoherent character. Such fluctuations would be also a test of quantum electrodynamics.[7-9]

These fluctuations are very sensitive to the atomic density and the power of the exciting field. The assumptions $T_2^* = \infty$, $\tau_p \gg T_2$ and the coherent plane-wave assumption are not necessary.[12] For example, for incoherent Gaussian driving fields, the above discussion remains valid. The effect of the beam profile of the driving field and the transverse effects in the medium will be treated elsewhere.

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