Spontaneous emission of a resonance medium in a strong traveling-wave field

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The spontaneous emission by atoms (executing thermal motion) in the field of a strong progressive monochromatic wave is investigated in the case of equal relaxation constants. It is shown that the compensation of Doppler shifts, which occurs during this process, gives rise to a structure on the Doppler profile of the main spectrum and the beat spectrum, which is qualitatively similar to the spectrum due to fixed atoms.

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There is a considerable number of published papers, both Soviet and foreign, on the behavior of a resonance medium in a strong field. Most of the interest has been focused on the amplification (absorption) of a weak probe signal in the presence of a strong signal. Much less attention has been devoted to spontaneous emission. This has probably been due to experimental difficulties. Nevertheless, each of these problems (amplification and spontaneous emission) has its own particular features and may be of independent interest.

In this paper, we shall be interested in spontaneous emission. First, let us summarize existing information on this process. The spectrum of spontaneous emission of fixed atoms has been investigated[1-3] for a strong field associated with a progressive monochromatic wave. This has been done for both equal and very different relaxation constants, and for moving atoms in the case of very different relaxation constants. The emission spectrum of fixed atoms was also investigated for equal relaxation constants in the field of a standing wave. It was assumed in these calculations that the two levels involved in the process were not ground states, and decayed rapidly to lower-lying levels. An attempt to investigate the polarization characteristics of spontaneous emission in a strong progressive wave field in the case of a gaseous resonance medium was reported in[4]. However, the analysis was confined to lowest-order perturbation theory in the external field. The beat spectrum of spontaneous radiation from fixed atoms was investigated in[5] (second order correlations, according to Glauber[6]). The foregoing list of published papers cannot, of course, be regarded as exhaustive. For example, we have not mentioned the large group of papers in which one of the levels is the ground state. This leads to additional features in the spectrum. However, we have mentioned all the aspects which have been discussed in the literature on spontaneous emission in an external monochromatic field.

We shall determine the principal spectrum $I_1(\omega)$ (first-order correlations) and the beat spectrum $I_2(\omega)$ (second-order correlations) of the spontaneous emission of a gaseous resonance medium exposed to a strong monochromatic progressive wave. We shall assume that the broadening due to the motion of the atoms is much greater than the natural broadening ($\Delta\omega_D \gg \gamma_{ab}$). The external field is incident on the boundary of the resonance medium at the point $x = 0$, and propagates in the positive direction of the x axis (we shall confine our attention to the one-dimensional problem). We shall suppose that the frequency ω_f of the strong field is equal to the frequency ω_0 of the atomic transition corresponding to spontaneous emission. We shall not be interested in self-amplification or self-absorption in the resonance medium, and will therefore assume that the thickness of the medium is sufficiently small. We shall remain within the framework of the two-level model, assuming that neither of these levels is the ground state and that they decay rapidly to lower-lying states ($\gamma_1 \ll \gamma$, where γ_1 is the decay constant between the upper level a and the lower level b and γ is the decay constant to the lower-lying levels).

Rautian has shown that, in the case of a medium located in the strong field of a standing wave, the central part of the Doppler profile acquires a structure which, in the special case of equal relaxation constants ($\gamma_a = \gamma_b = \gamma_{ab}$), is due to the compensation of Doppler shifts during multiphoton processes.[3] This structure thus corresponds to the spectrum of spontaneous emission of fixed atoms in the field of the standing wave. There are no reasons to suppose that the mechanism responsible for the formation of the structure in the field of a progressive wave will be different (when the spontaneous emission spectrum is observed in the direction of a strong field). However, we must then expect the appearance of three sharp peaks instead of the one found in the case of the standing wave. This is so because the spontaneous emission spectrum due to fixed atoms consists of three profiles in this case[3] (Fig. 1a) (in the standing wave, the lateral peaks are suppressed because of spatial inhomogeneity). The correlation properties of $I_1(\omega)$ which determine the spectrum $I_2(\omega)$, are also expected to be of the same kind as for fixed atoms. In particular, the lateral peaks should be in phase with one another, and one half of the central peak should be in antiphase with the other.[5]

We shall consider two cases. In the first case, only the upper level is populated and, in the second, only the lower level is populated. The first case is important for the study of spontaneous emission of quanaum-mechanical amplifiers and generators. From the experimental point of view, this has been the only conceivable variant until quite recently. However, the appearance of lasers based on the use of dyes has altered the situation. It is now more convenient to consider the excitation of the lower level (see below for reasons).

We start with the equation for the density matrix for spontaneous emission (the formal scheme is described in the Appendix)[5]

$$\partial\rho/\partial x = -A_1(a_1a_1^+\rho - a_1^+\rho a_1 + \rho a_2a_2^+ - a_2^+\rho a_2)$$

$$-A_2(\rho a_1^+a_1-a_1\rho a_1^++a_2^+a_2\rho-a_2\rho a_2^+)$$
$$+A_3(a_1a_2\rho-a_2\rho a_1+\rho a_1^+a_2^+-a_2^+\rho a_1^+)$$
$$+A_4(\rho a_1a_2-a_1\rho a_2+a_1^+a_2^+\rho-a_1^+\rho a_2^+)+\text{h.c.}$$

where ρ is the density matrix describing two spontaneous emission waves with frequencies $\omega_1 = \omega_0 - \Delta$ and $\omega_2 = \omega_0 + \Delta$, located symmetrically relative to the atomic transition frequency (we recall that the frequency of the strong field is equal to the frequency of the atomic transition). The coefficients A_1 and A_2 describe the independent interaction of each of the waves with the resonance medium exposed to the strong classical field, whilst A_3 and A_4 are responsible for their combinational interaction involving four-photon processes. The expressions for the coefficients of the equation for fixed atoms are given in the Appendix.

When the thickness of the resonance medium is small, the main spectral profile is determined by $I_1(\omega) = 2x\mathrm{Re}A_1$. To obtain information about the beat spectrum, we shall use the separation of the main profile into AM and PM profiles[5] (the AM profile is the profile in which one half is in phase with the other and, whereas in the PM profile, one half is in antiphase with the other). Each spectral profile can be written as the sum $I_1(\omega) = I_{AM}(\omega) = I_{PM}(\omega)$. Thus, if we are only interested in beats with the central component, all the information about them is contained in the AM profile. The PM profile does not contribute to these beats because oscillations on the right of the central frequency are compensated by oscillations on the left. In our case, when there is a strong field at the central frequency, all other beats can be neglected. Thus, the problem of the beat spectrum essentially reduces to the determination of the AM profile corresponding to the main spectral profile $I_1(\omega)$. The AM profile is given by $I_{AM}(\omega) = x\mathrm{Re}(A_1 + A_4)$. The explicit expressions for $\mathrm{Re}A_1$ and $\mathrm{Re}(A_1 + A_4)$ are given in the Appendix for waves traveling along the strong field when only the upper (1) or only the lower (2) of the two levels is excited

As expected, the structure on the Doppler profile of spontaneous emission of a medium in the strong field of a progressive wave is the same as the spectrum due to fixed atoms (Fig. 1a shows the spectrum due to fixed atoms and Fig. 1b the spectrum due to atoms executing thermal motion; the analysis was performed subject to the condition $\gamma \ll G \ll \Delta\omega_D$, where G is the power associated with the external field in units of frequency). There are three clearly defined peaks against the background of the broad valley (width $\sim G$) whose relative width is of the order of unit. The central frequency peak has a height $\sim G/\gamma$ and occurs at the frequency ω_0, and the other two are found at frequencies $\omega_0 \pm 2G$ and their heights are $\sim(G/\gamma)^{1/2}$. Thus, in a sense, thermal motion facilitates the observation of the dynamic Stark effect because the distribution then has a greater contrast (for fixed atoms the height of the central peak is of the order of unity and that of the lateral peaks of the order of one-half). Of course, the Doppler "background" will produce a strong noise effect and will impede the reliable detection of the structure. However, this "background" vanishes when the lower rather than the upper level is excited. In this case, the spontaneous emission spectrum is qualitatively similar to the spectrum for fixed atoms (Fig. 2a). The absence of the Doppler background would appear to be natural because only those atoms which interact with the strong field participate in the population of the upper level.

The shape of the AM profiles in Figs. 1d and 2b can

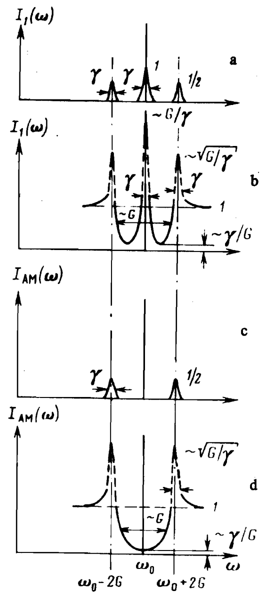

FIG. 1

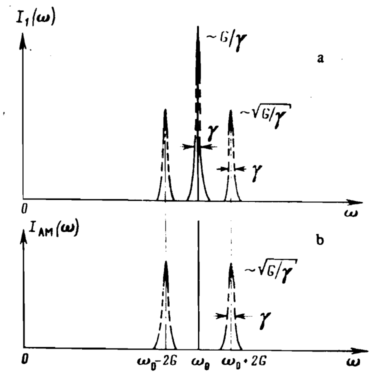

FIG. 2

FIG. 1. Spectral characteristics of spontaneous emission of atoms in a strong field when only the upper state is excited.

FIG. 2. Spectral characteristics of spontaneous emission of atoms in a strong field when only the lower state is excited.

be used to obtain information about the correlations in the main profiles $I_1(\omega)$ between different Fourier components, due to the combinational interaction between them. It is clear that when either the upper or the lower level is populated, the central peak is absent from the AM profiles but the lateral peaks remain unaltered. This means that the lateral components are correlated in phase and one half of the central peak is in antiphase with the other (cf. with fixed atoms, Fig. 1c).

Finally, a few words about emission in other directions relative to the strong field. Here, there is no point in discussing the beat spectrum in addition to the main spectrum $I_1(\omega)$. The absence of combinational coupling between waves means that the appearance of additional correlations between different Fourier components of the main spectrum cannot be considered within the framework of the above model. Moreover, for emission in backward directions, multiphoton processes do not lead to compensation of Doppler shifts. The result is that the spectrum $I_1(\omega)$ does not exhibit any well-defined structure. When the upper level is excited, there is simply a Doppler profile with the Bennett dip (width $\sim G$, relative depth $\sim 1/2$), and when the lower level is excited we have the Lorentz profile of width 2G.

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APPENDIX

Golubev[5] has given a prescription that can be used to construct the kinetic equation for the density matrix describing spontaneous emission:

$$\begin{aligned}\partial\rho/\partial x = &-A_1(a_1a_1^+\rho - a_1^+\rho a_1) - A_1'(\rho a_2a_2^+ - a_2^+\rho a_2)\\ &-A_2(\rho a_1^+a_1 - a_1\rho a_1^+) - A_2'(a_2^+a_2\rho - a_2\rho a_2^+)\\ &+A_3(a_1a_2\rho - a_2\rho a_1) + A_3'(\rho a_1^+a_2^+ - a_2^+\rho a_1^+)\\ &+A_4(\rho a_1a_2 - a_1\rho a_2) + A_4'(a_1^+a_2^+\rho - a_1^+\rho a_2^+) + \text{h.c.};\end{aligned}$$

where ρ is the density matrix describing two spontaneous emission waves of frequency $\omega_1 = \omega_f - \Delta$ and $\omega_2 = \omega_f + \Delta$, located symmetrically relative to the frequency ω_f of the strong classical field. All the coefficients A_i and A_i' depend on Δ and on the power G associated with the strong external field $[G^2 = |g|^2 n(0)$, $n(0)$ is the mean number of photons of the strong external field at the point $x = 0]$. Although, in the present paper, we assume that the frequency of the strong field is equal to the frequency of the atomic transition, to take into account the thermal motion of the atoms one must write down the equation for $\omega_{0f} = \omega_0 - \omega_f \neq 0$.

We shall now write down the coefficients of the equation for fixed atoms.

A. Excitation of the upper atomic level:

$$A_{1a}=|g|^2N_a\frac{\omega_{0f}^2+\Gamma_1^2}{\omega_{0f}^2+\Gamma^2}\frac{1}{i\omega_{10}+\gamma}+B,$$
$$B=|g|^2N_a\frac{\omega_{0f}^2+\Gamma_1^2}{\omega_{0f}^2+\Gamma^2}\frac{1}{i\omega_{10}+\gamma} \quad (1)$$
$$\times\left[-\frac{G^2}{i\omega_{1f}+\gamma}\frac{\omega_{0f}^2+(\gamma+i\omega_{1f})^2}{\omega_{0f}^2+(\gamma+i\omega_{1f})^2+4G^2}\left(\frac{2}{i\omega_{10}+\gamma}+\frac{-i\omega_{0f}+\gamma}{\omega_{0f}^2+\Gamma^2-2G^2}\right)\right],$$
$$A_{2a}=|g|^2N_a\frac{2G^2}{\omega_{0f}^2+\Gamma^2}\frac{1}{i\omega_{10}+\gamma}$$
$$\times\left[-\frac{G^2}{i\omega_{1f}+\gamma}\frac{\omega_{0f}^2+(\gamma+i\omega_{1f})^2}{\omega_{0f}^2+(\gamma+i\omega_{1f})^2+4G^2}\left(\frac{2}{i\omega_{10}+\gamma}-\frac{-i\omega_{0f}+\gamma}{2G^2}\right)\right],$$
$$A_{3a}=|g|^2N_a\frac{2G^2}{\omega_{0f}^2+\Gamma^2}\frac{1}{i\omega_{10}+\gamma}$$
$$\times\left[-\frac{G^2}{i\omega_{1f}+\gamma}\frac{\omega_{0f}^2+(\gamma+i\omega_{1f})^2}{\omega_{0f}^2+(\gamma+i\omega_{1f})^2+4G^2}\left(\frac{2}{i\omega_{02}+\gamma}-\frac{i\omega_{0f}+\gamma}{2G^2}\right)\right],$$
$$A_{4a}=|g|^2N_a\frac{\omega_{0f}^2+\Gamma_1^2}{\omega_{0f}^2+\Gamma^2}\frac{1}{i\omega_{10}+\gamma}$$
$$\times\left[-\frac{G^2}{i\omega_{1f}+\gamma}\frac{\omega_{0f}^2+(\gamma+i\omega_{1f})^2}{\omega_{0f}^2+(\gamma+i\omega_{1f})^2+4G^2}\left(\frac{2}{i\omega_{02}+\gamma}+\frac{i\omega_{0f}+\gamma}{\omega_{0f}^2+\Gamma_1^2}\right)\right].$$

The coefficients $A_1'A_2'A_3'$ and A_4' are obtained, respectively, from $A_1A_2A_3$ and A_4 by introducing the substitution $\omega_1 \rightleftarrows \omega_2$ into these equations.

The notation is: N_a—stationary density of atoms in the upper state in the absence of the strong field, $g = (2\omega)^{-1/2} d_{ab}\epsilon e^{-i\mathbf{k}\cdot\mathbf{r}}$—coupling constant between the $\mathbf{k}$-th wave of the electromagnetic field and the atom at the point $\mathbf{r}$, and

$$\omega_{0f}=\omega_0-\omega_f,\ \ \omega_{1f}=\omega_1-\omega_f,\ \ \omega_{10}=\omega_1-\omega_0,$$
$$\omega_{02}=\omega_0-\omega_2,\ \ \Gamma^2=\gamma^2+4G^2,\ \ \Gamma_1^2=\gamma^2+2G^2.$$

B. Excitation of the lower atomic state:

$$A_{1b}=|g|^2N_b\frac{2G^2}{\omega_{0f}^2+\Gamma^2}\frac{1}{i\omega_{10}+\gamma}+A_{2a}(N_a\to N_b),$$
$$A_{2b}=B(N_a\to N_b),\ \ A_{3b}=A_{4a}(N_a\to N_b),\ \ A_{4b}=A_{3b}(N_a\to N_b); \quad (2)$$

where N_b is the density of atoms in the lower state in the absence of the strong field.

To take into account the thermal motion of the atoms, we must introduce the substitutions $\omega_1 \to \omega_1 - k_1v$, $\omega_2 \to \omega_2 - k_2v$ into the above expressions and average the result over the velocities v (assuming a Maxwellian velocity distribution). In the case of the limiting Doppler broadening $(\Delta\omega_D^2 \gg \gamma^2 + 4G^2 = \Gamma^2)$, all the integrals can readily be evaluated in terms of the residues. For spontaneous emission in the direction of the strong field, we have

$$\operatorname{Re}A_{1a}=|g|^2N_a\pi D(\Delta)\operatorname{Re}\left[\frac{G^2}{i\Delta}\left(\frac{1}{\Gamma}-\frac{1}{\tilde\Gamma}\right)+\frac{1}{\gamma-i\Delta}\left(-\frac{G^2}{\Gamma}-\frac{2G^2}{\tilde\Gamma}+\tilde\Gamma\right)\right],$$
$$\operatorname{Re}(A_{1a}+A_{4a})=|g|^2N_a\pi D(\Delta)\operatorname{Re}\left[\frac{2G^2}{i\Delta}\left(\frac{1}{\Gamma}-\frac{1}{\tilde\Gamma}\right)+\frac{1}{\gamma-i\Delta}\left(-\frac{4G^2}{\tilde\Gamma}+\tilde\Gamma\right)\right]; \quad (3)$$

$$\operatorname{Re}A_{1b}=|g|^2N_b\pi D(\Delta)\operatorname{Re}\left[-\frac{G^2}{i\Delta}\left(\frac{1}{\Gamma}-\frac{1}{\tilde\Gamma}\right)+\frac{G^2}{\Gamma}\frac{1}{\gamma-i\Delta}\right],$$
$$\operatorname{Re}(A_{1b}+A_{4b})=|g|^2N_b\pi D(\Delta)\operatorname{Re}\left[-\frac{2G^2}{i\Delta}\left(\frac{1}{\Gamma}-\frac{1}{\tilde\Gamma}\right)\right]. \quad (4)$$

In these expressions

$$\Delta=-\omega_{10}=\omega_0-\omega_1,\ \ \tilde\Gamma=\pm[(\gamma-i\Delta)^2+4G^2]^{1/2}$$

(the sign must always be chosen so that the real part of $\tilde\Gamma$ is positive: $\operatorname{Re}\tilde\Gamma > 0$) and

$$D(\Delta)=\frac{1}{\pi^{1/2}\Delta\omega_D}\exp\left(-\frac{\Delta^2}{\Delta\omega_D^2}\right)$$

is the Doppler profile centered on the frequency of the atomic transition.

The difficulties encountered in the analysis of (3) and (4) are connected with the quantity $\tilde\Gamma$. In the most interesting case, $G \gg \gamma$, the frequency scale splits into four characteristic regions: near the center of the atomic transition $\Delta \lesssim \gamma$, near the points $\omega_0 \pm 2G$ $(\Delta^2 - 4G^2 \lesssim \gamma^2)$, and two intermediate regions. The following approximate expressions are convenient for the analysis of spectra:

$$\tilde\Gamma\approx 2G\left(1+\frac{\gamma^2-\Delta^2}{8G^2}-\frac{i\gamma\Delta}{4G^2}\right),\quad \Delta\leqslant\gamma;$$
$$\tilde\Gamma\approx 2G\left(1-\frac{\Delta^2}{8G^2}-\frac{i\gamma\Delta}{4G^2}\right),\quad \gamma\ll\Delta\ll 2G;$$
$$\tilde\Gamma\approx(\gamma\Delta)^{1/2}\left(1+i-\frac{\Gamma^2-\Delta^2}{4\gamma\Delta}+i\frac{\Gamma^2-\Delta^2}{4\gamma\Delta}\right),\quad \Delta\sim 2G;$$
$$\tilde\Gamma\approx\gamma-i\Delta;\quad \Delta\gg 2G.$$

For example, near the center of the profile, we have the approximate expressions

$$\operatorname{Re}A_{1a,b}=|g|^2N_{a,b}\pi D(\Delta)\frac{G}{2}\frac{\gamma}{\gamma^2+\Delta^2},$$
$$\operatorname{Re}(A_{1a,b}+A_{4a,b})\sim\gamma/G\ll 1,$$

and at the points $\omega_0 \pm 2G$:

$$\operatorname{Re}A_{1a,b}={}^1/_4|g|^2N_{a,b}\pi D(2G)(G/2\gamma)^{1/2},$$
$$\operatorname{Re}(A_{1a,b}+A_{4a,b})={}^1/_2|g|^2N_{a,b}\pi D(2G)(G/2\gamma)^{1/2}.$$

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