

# Radiation from an atom located in an external electromagnetic field

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Radiation from an atom located in an external electromagnetic field in the case when the field amplitude and the level widths are arbitrarily related is considered in the resonance approximation only. The emission spectrum is determined from a system of kinetic equations for the atom-density and correlation matrices. The case of a monochromatic external field is considered in detail. The spectrum is split into coherent and noncoherent parts. The transition to the case of rapidly decaying levels, when the one-photon approximation is valid, as well as to the case of a stationary lower level (resonance fluorescence), is considered.

## 1. INTRODUCTION

In the present paper we investigate the emission spectrum of an atom located in an external electromagnetic field. We consider, using only the resonance approximation, the general case of an arbitrary relation between the field strength and the upper- and lower-level widths  $\gamma_a$  and  $\gamma_b$ . This problem has been investigated before in a number of papers<sup>[1-7]</sup>. In these investigations, however, either the one-photon approximation was used, or the strong-field limit was considered without allowance for the level widths. The one-photon approximation turns out to be valid in the case when one of the states of the atom decays sufficiently rapidly (see<sup>[3]</sup>), i.e., when

$$\gamma \ll \gamma_a^0 + \gamma_b. \quad (1)$$

The probability of emission of a resonance photon is then given by  $\gamma/(\gamma_a^0 + \gamma_b) \ll 1$ . Here it is assumed that the width  $\gamma_a$  of the upper level is the sum of the width  $\gamma$  of the resonance transition and the width  $\gamma_a^0$  of the transition to the other nonresonance states. The lower level broadens only because of the nonresonance transitions.

Below it is assumed that the relation between  $\gamma$ ,  $\gamma_a^0$ , and  $\gamma_b$  can have any form and that the number of emitted resonance photons is arbitrary. If, in particular, the lower level is the ground state and  $\gamma_b^0 = 0$ , then we have resonance fluorescence to deal with. The spectral properties of the radiation are determined from a system of kinetic equations for the atom-density and correlation matrices. With the aid of these equations, we consider in detail the radiation emitted by an atom located in a monochromatic field.

## 2. THE BASIC EQUATION

In deriving the kinetic equations, we shall, for simplicity, omit the relaxation constants  $\gamma_a^0$  and  $\gamma_b$ ; they can easily be taken into account in the final equations. The external electromagnetic field has the form

$$E(t) e^{-i\omega_0 t} + c.c., \quad (2)$$

where  $\omega_0$  is the transition frequency and  $E(t)$  is a slowly varying amplitude. The field  $E(t)$  is assumed to be sufficiently strong and will be treated below as a classical field. We shall measure the field frequencies relative to the transition frequency.

The basic Hamiltonian in the resonance approximation has the form ( $\hbar = 1$ )

$$H = H_a + H_e + H_{ac};$$

$$H_e = d(\sigma_+ E(t) + \sigma_- E^*(t)), \quad H_a = \sum_{\mathbf{k}} (\omega_{\mathbf{k}} - \omega_0) a_{\mathbf{k}}^+ a_{\mathbf{k}}, \quad (3)$$

$$H_{ac} = \sum_{\mathbf{k}} g_{\mathbf{k}} (\sigma_+ a_{\mathbf{k}} + \sigma_- a_{\mathbf{k}}^+).$$

Here  $a_{\mathbf{k}}^+$  and  $a_{\mathbf{k}}$  are the photon creation and annihilation operators, which obey the usual commutation rule

$$[a_{\mathbf{k}}, a_{\mathbf{k}'}^+] = \delta_{\mathbf{k}, \mathbf{k}'}. \quad (4)$$

The quantities  $\sigma_x$ ,  $\sigma_y$ , and  $\sigma_z$  denote the Pauli spin matrices, which obey the well-known product rule

$$\sigma_i \sigma_j = \delta_{ij} + i \epsilon_{ijk} \sigma_k, \quad (5)$$

where  $\epsilon_{ijk}$  is the antisymmetric unit tensor. In the usually employed representation  $\sigma_z$  is the excess-population operator, while

$$\sigma_{\pm} = \frac{1}{2}(\sigma_x \mp i\sigma_y), \quad \sigma_{\pm} = \frac{1}{2}(\sigma_x \pm i\sigma_y) \quad (6)$$

are the radiation-current operators with positive and negative frequencies. The transition dipole moment in (3) is denoted by  $d$ . The explicit form of the field-atom coupling constant  $g_{\mathbf{k}}$  need not be written out, since below it will enter only into the relaxation frequency  $\gamma$ .

Usually, in solving the quantum problem of spontaneous emission by an atom, we expand the wave function of the field in terms of states with a fixed number of photons. In the present case such an approach is inconvenient, since the number of emitted photons can be large. For example, in the case when the lower atomic level is the ground level, the number of emitted photons is proportional to the time. To find the spectral properties of the radiation, it is sufficient to know the mean radiation current  $\langle \sigma^H(t) \rangle$  and the current-current correlator  $\langle \sigma_i^H(t_1) \sigma_j^H(t_2) \rangle$ . The index H indicates that the operator is a Heisenberg operator and the angle brackets denote averaging over the initial quantum state of the field and the atom. It is assumed that there are no photons in the initial state (except the photons of the external field  $E(t)$ ).

Let us define the following mean quantities and correlators:

$$F_i(t) = \langle \sigma_i^H(t) \rangle, \quad F_{ij}(t_1, t_2) = \langle \sigma_i^H(t_1) \sigma_j^H(t_2) \rangle. \quad (7)$$

Let us recall that  $F_z$  is the difference between the populations of the levels a and b, while  $F_x$  and  $F_y$  are the real and imaginary components of the radiation current. Let us denote by  $F_0$  the total population of the levels a and b. The set of quantities  $F_i$ ,  $F_0$  is equivalent to an atom-density matrix. For the correlator  $F_{i_1 i_2}$  we have, in accordance with (5), the following boundary condition:

$$F_{i_1 i_2}(t, t_1) = F_0(t) \delta_{i_1 i_2} + i e_{i_1 i_2} F_j(t_1). \quad (8)$$

The functions  $F_i$  and  $F_{i_1 i_2}$  obey a simple system of kinetic equations. Knowing these functions, we can easily determine the spectral properties of the radiation.

Let us further introduce the matrices  $\sigma_i(t)$ ; their variation with time is determined by the Hamiltonian  $H_a(t)$ :

$$\begin{aligned} \partial \sigma_i(t) / \partial t &= 2d e_{i, \mu} E_j(t) \sigma_k(t); \\ E_x &= \text{Re } E(t), \quad E_y = -\text{Im } E(t), \quad E_z = 0. \end{aligned} \quad (9)$$

Let us go over to the interaction representation and define the S matrix corresponding to the Hamiltonian  $H_{ae}(t)$ :

$$\begin{aligned} i \partial S / \partial t &= H_{ae}(t) S; \\ H_{ae}(t) &= \sigma_+(t) f(t) + \sigma_-(t) f^*(t), \quad f(t) = \sum_{\mathbf{k}} g_{\mathbf{k}} a_{\mathbf{k}} \exp[-i(\omega_{\mathbf{k}} - \omega_0)t]. \end{aligned} \quad (10)$$

Since we are interested only in the resonance frequencies  $|\omega_{\mathbf{k}} - \omega_0| \ll \omega_0$ , the commutator  $[f, f^*]$  can be replaced by a time delta function:

$$[f(t_1), f^*(t_2)] = \gamma \delta(t_1 - t_2), \quad \gamma = \sum_{|\mathbf{k}| = \omega_0/c} |g_{\mathbf{k}}|^2. \quad (11)$$

Such a replacement corresponds to the Wigner-Weisskopf approximation, which is usually used in the theory of the spontaneous radiation emission by a free atom. Thus, the Hamiltonian (10) describes the behavior of an atom in the quantized field  $f(t)$ , which can be regarded as "white" noise. This circumstance significantly simplifies the calculations.

Let us write  $F_i(t)$  in terms of the operators in the interaction representation:

$$F_i(t) = \langle S^+(t, 0) \sigma_i(t) S(t, 0) \rangle, \quad (12)$$

and let us find the small change in  $F_i$  due to the change  $\Delta t$  in  $t$ :

$$\begin{aligned} F_i(t + \Delta t) &= F_i(t) + 2d e_{i, \mu} E_j(t) F_k(t) \Delta t \\ &+ \langle S^+(t, 0) (S^+(t + \Delta t, t) - 1) \sigma_i(t) (S(t + \Delta t, t) - 1) S(t, 0) \rangle. \end{aligned} \quad (13)$$

On account of the condition (11), the photon operators entering into  $S(t, 0)$  and  $S(t + \Delta t, t)$  can be averaged independently<sup>1)</sup>. As a result, taking the limit as  $\Delta t \rightarrow 0$ , we obtain

$$\begin{aligned} \partial F_{i_1 i_2} / \partial t &= 2d e_{i_1 \mu} E_j F_k - (\hat{\gamma} F)_{i_1} - 2\gamma \delta_{i_1 i_2} F_0; \\ (\hat{\gamma} F)_{i_1} &= \begin{cases} \gamma F_{i_1} & i = x, y, \\ 2\gamma F_{i_1} & i = z. \end{cases} \end{aligned} \quad (14)$$

This equation coincides with the Bloch equation (see, for example, [9]). The equation for  $F_{i_1 i_2}$  can easily be obtained in a similar manner. Thus, for  $t_1 > t_2$  we have

$$\partial F_{i_1 i_2}(t, t_2) / \partial t_1 = 2d e_{i_1 \mu} E_j(t_1) F_{k i_2}(t_1, t_2) - (\hat{\gamma} F)_{i_1}(t_1) - 2\gamma \delta_{i_1 i_2} F_0(t_2). \quad (15)$$

For  $t_2 > t_1$ , the equation for  $F_{i_1 i_2}$  has the same form as Eq. (15), except that the variables are  $t_2$  and the index  $i_2$ . As the initial condition for the solution of Eq. (15), the relation (8) should be used.

Thus, Eqs. (14), (15), and (8) constitute a closed system of quantum-kinetic equations, from which the correlation properties of the radiation current can be determined. Notice that an equation of the type (15) was obtained in [5] for the correlation density matrix, the only difference being that  $\gamma_a^0$  and  $\gamma_b$  serve in accordance with the condition (1), as the relaxation constants in this equation.

Let us represent the correlation function in the form

$$F_{i_1 i_2}(t, t_2) = F_{ii}(t_1) F_{ii}(t_2) + \Phi_{i_1 i_2}(t, t_2). \quad (16)$$

The first term in this expression depends on the phase of the external field and describes the coherent part of the radiation, while the second term does not depend on the phase and corresponds to the incoherent part. In the case when the lower level is the ground level and the norm of the working levels is conserved ( $F_0 = 1$ ), the equation for  $\Phi_{i_1 i_2}$  has the same form as Eq. (15), except that it is a homogeneous equation. The solution of this equation can be found with the aid of the retarded Green function  $G_{i_1 i_2}(t_1, t_2)$  of the homogeneous Bloch equation:

$$\Phi_{i_1 i_2}(t, t_2) = G_{i_1 i_2}(t, t_2) [\delta_{i_1 i_2} - F_i(t_1) F_i(t_2) + i e_{i_1 i_2} F_k(t_2)], \quad t_1 > t_2. \quad (17)$$

The correlation function  $\Phi_{i_1 i_2}$  can be expressed as a bilinear form in terms of the atom-density matrix. In the following section we shall discuss the physical reason for such a nonlinear relation.

### 3. THE EMISSION SPECTRUM OF THE ATOM

Let us consider with the aid of the obtained system of kinetic equations the emission spectrum of an atom located in an external field. The emission spectrum can be determined from the correlator

$$\langle E^H(t_1) E^H(t_2) \rangle, \quad (18)$$

where  $E^H(t)$  is Heisenberg operator for the electromagnetic field at some observation point. If we disregard the free-field operator, which makes no contribution to (18), then in the wave zone  $E^H(t)$  coincides up to a constant factor with  $\sigma_-^H(t - \mathbf{r}/c)$ , where  $\mathbf{r}$  is the distance to the observation point. Therefore, to find the emission spectrum, it is necessary to compute the correlator  $F_{+-}(t_1, t_2)$ . Notice that this correlator satisfies the condition

$$F_{+-}^*(t_1, t_2) = F_{+-}(t_2, t_1), \quad (19)$$

which allows us to represent the emission spectrum  $I(\omega)$  in the following form:

$$I(\omega) = \frac{2\gamma}{\pi} \text{Re} \int_0^{\infty} dt_2 \int_{t_1}^{\infty} dt_1 \exp[-i\omega(t_1 - t_2)] F_{+-}(t_1, t_2). \quad (20)$$

Since the angular dependence has the standard form, we consider only the total radiation intensity integrated over the angle. The normalization constant has been chosen such that the total radiated energy

$$J = \int_{-\infty}^{+\infty} d\omega I(\omega) \quad (21)$$

is measured in units of the photon energy  $\omega_0$ . Therefore,  $J$  is the average number of resonance photons emitted. In particular, when the condition (1) is fulfilled,  $J$  is small and determines the probability of emission of a photon of frequency  $\omega_0$ .

The spectral function  $I(\omega)$  can be split into its coherent  $I_c(\omega)$  and incoherent  $I_{inc}(\omega)$  radiation components, which are determined by the mean value of, and the fluctuations in, the radiation current. Using the relation (16), we have:

$$I(\omega) = I_c(\omega) + I_{inc}(\omega); \quad (22)$$

$$I_c = \frac{\gamma}{\pi} \left| \int_0^{\infty} dt e^{-i\omega t} F_+(t) \right|^2,$$

$$I_{inc}(\omega) = \frac{\gamma}{2\pi} \text{Re} \int_0^{\infty} dt \int_0^{\infty} d\tau e^{-i\omega\tau} \Phi_{+-}(t + \tau, t).$$

Let us note some general properties of the integrated intensities of the coherent and incoherent com-

ponents of the radiation  $J_c$  and  $J_{inc}$ . With the aid of the formula (8), we find

$$J_c = \int_0^{\infty} dt |F_+(t)|^2, \quad J_{inc} = \int_0^{\infty} dt \left( \frac{F_0(t) + F_+(t)}{2} - |F_+(t)|^2 \right). \quad (23)$$

These relations have a simple physical meaning. The quantity  $\frac{1}{2}(F_0 + F_+)$  is the population of the upper working level. The total radiated energy can be expressed as a time integral of this quantity. The distribution of the radiated energy between the coherent and incoherent components is determined by  $|F_+(t)|^2$ , the square of the modulus of the mean current.

Nevertheless, it is possible to distinguish experimentally between the coherent and incoherent parts of the radiation only in certain cases. Thus, in laser radiation we deal with radiation emitted by atoms in a selected mode of a resonator, i.e., in the case of a traveling wave, with forward radiation scattering. The mean strength of the electromagnetic field of the mode is then expressible in terms of the mean radiation current  $F_+$  from the atom, while the fluctuations in the intensity of the mode are expressible in terms of the quantity  $J_{inc}$ . The correction to  $J_{inc} = J - J_c$  due to the subtraction of  $J_c$  was noted earlier<sup>[10]</sup> in connection with the quantum theory of the laser.

The quantity  $J_c/J$  can be regarded as the stimulated-emission probability,  $J_{inc}/J$  as the spontaneous-emission probability. As a function of the external-field intensity,  $J_c$  has a maximum in the vicinity of the saturating field.  $J_c$  is small in weak and in strong fields (because of the saturation effect). Therefore, the spontaneous-emission probability has in the vicinity of the saturating field a dip of relative depth  $\sim 25\%$ <sup>[10]</sup>. If we consider the radiation emitted at some angle to the incident field, then the coherent component can be distinguished by its spectrum in the case of a metastable or stable lower level. In this case at low external-field energies  $J \approx J_c$ , while at high field energies  $J \approx J_{inc}$ .

We shall carry out the subsequent concrete computations for the case of a monochromatic external field.

#### 4. RADIATION IN A MONOCHROMATIC FIELD

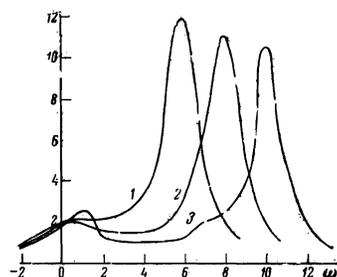
Let us consider the case of the monochromatic field:

$$E(t) = Ee^{-i\omega t}, \quad (24)$$

where  $\Delta$  is the field-frequency detuning relative to the transition frequency. We can get rid of the time dependence of the field by going over into a rotating coordinate system. In this coordinate system, there appears in the expression for  $F_{+-}$  an additional factor  $\exp[i\Delta(t_1 - t_2)]$ .

In order to somewhat simplify the computations, let us assume that

$$\gamma_a^0 = \gamma_0 = \gamma_0. \quad (25)$$



Plots of the function  $10\psi(\omega)$  for the case when  $\gamma_0 = \gamma$ . As the unit of frequency, we have used  $\gamma$ . The curve 1 corresponds to the value  $\Delta = 6$ , the curve 2 to  $\Delta = 8$ , and the curve 3 to  $\Delta = 10$ .

The emission spectrum can be expressed in terms of the Laplace transform of the function  $F_{+-}(t_1, t_2)$ , which transform we denote by  $\mathcal{F}_{+-}(pq)$ :

$$I(\omega) = \frac{2\gamma}{\pi} \text{Re} \mathcal{F}_{+-}(\gamma_0 + i(\omega - \Delta), \gamma_0),$$

$$\mathcal{F}_{+-}(pq) = \int_0^{\infty} dt \int_0^{\infty} d\tau e^{-p\tau - q\tau} F_{+-}(t + \tau, t). \quad (26)$$

The solution of the Bloch equation in the monochromatic field in the general form has a fairly unwieldy form. Therefore, we shall consider the following characteristic limiting cases.

**Weak fields.** Let us set  $F = 2|dE|$ . Then in a weak external field

$$V \ll \Gamma = \gamma + \gamma_0 \quad (27)$$

we have the following emission spectrum:

$$I(\omega) = \gamma V^2 \psi(\omega) / 2\pi\gamma_0(\Gamma^2 + \Delta^2), \quad (28)$$

$$\psi(\omega) = \text{Re} \left\{ \frac{1}{(\gamma + i\Delta)} \left[ \frac{\gamma_0(i\Delta - \Gamma)}{(\Gamma + \gamma)(\Gamma + i\omega)} + \frac{\Gamma + i\Delta}{\gamma_0 + i(\omega - \Delta)} \right] \right\}.$$

Here it is assumed that the atom is in the lower state at the initial moment of time. The emission spectrum is a superposition of two dispersion contours with complex weights. The first term in the square brackets in (28) corresponds to spontaneous emission: the line width is  $\Gamma = \gamma_0 + \gamma$  and the peak is located in the vicinity of the line center. The second term corresponds to stimulated emission: the line width is  $\gamma_0$  and the peak is located near the field frequency  $\omega \approx \Delta$ . As an example, we show in the figure plots of the function  $\psi(\omega)$  for several values of  $\Delta$ . It can be seen from the figure that the peak corresponding to the spontaneous emission becomes pronounced only at fairly large  $\Delta$ . At  $\Delta \gg \Gamma$ , the contours do not overlap, and we have independent contributions from the spontaneous and stimulated emissions:

$$I(\omega) = \frac{\gamma V^2}{2\pi\gamma_0\Delta^2} \text{Re} \left[ \frac{\gamma_0}{(\Gamma + \gamma)} \frac{1}{(\Gamma + i\omega)} + \frac{1}{\gamma_0 + i(\omega - \Delta)} \right]. \quad (29)$$

The relative weight of the spontaneous emission is  $\gamma_0/(\Gamma + \gamma)$ , so that at small values of  $\gamma_0$  the intensity of the spontaneous emission is low, while at  $\gamma_0 \gg \gamma$  the spontaneous and stimulated emissions have the same intensity.

At  $\Delta = 0$  the dispersion contours in (28), generally speaking, overlap, and the contributions from the spontaneous and stimulated emissions cannot be considered to be independent:

$$I(\omega) = \frac{\gamma V^2}{2\pi\Gamma(\Gamma + \gamma)} \frac{\omega^2 + \Gamma(2\Gamma + \gamma_0)}{(\omega^2 + \gamma_0^2)(\omega^2 + \Gamma^2)}. \quad (30)$$

It can be seen from (30) that the emission spectrum differs somewhat from the Lorentz spectrum, and that the larger the ratio  $\gamma_0/\gamma$ , the greater this difference. At small  $\gamma_0$  we have the Lorentz contour, which corresponds to stimulated emission.

**Strong field.** In a strong external field for which

$$V + |\Delta| \gg \Gamma \quad (31)$$

the emission spectrum assumes the following form:

$$I(\omega) = \frac{\gamma V^2}{4\pi\gamma_0\Omega^2(\Omega^2 + \varepsilon\Delta^2)} \text{Re} \left\{ \frac{(\Omega - \Delta)(\Omega + \varepsilon\Delta)}{\gamma_1 + i(\omega - \Delta - \Omega)} + \frac{(\Omega + \Delta)(\Omega - \varepsilon\Delta)}{\gamma_1 + i(\omega - \Delta + \Omega)} \right\} + \frac{2}{\Omega^2 + \Delta^2} \left[ \frac{V^2(\Omega^2 - \varepsilon\Delta^2)}{\gamma_2 + i(\omega - \Delta)} + \frac{2(1 + \varepsilon)\Delta^2\Omega^2}{\gamma_0 + i(\omega - \Delta)} \right];$$

$$\Omega^2 = V^2 + \Delta^2, \quad \gamma_1 = \Gamma + \gamma V^2/\Omega^2, \quad \gamma_2 = \gamma_1 - \gamma, \quad \varepsilon = \gamma/\Gamma.$$

Because of the Stark splitting of the levels in the ex-

ternal field, the emission spectrum has three peaks at the frequencies  $\omega = \Delta \pm \Omega$  and  $\omega = \Delta$ . The first three terms in the curly brackets in (32) correspond to spontaneous emission, the last term to stimulated emission. Notice that the statistical weights of the individual components depend not only on the field and the detuning, but on the ratio  $\gamma/\Gamma$  as well. At  $\Delta = 0$  we have

$$I(\omega) = \frac{2\gamma}{\pi\gamma_0} \operatorname{Re} \left[ \frac{1}{\gamma + \Gamma + i(\omega + \Omega)} + \frac{1}{i(\omega - \Omega) + \gamma + \Gamma} + \frac{2}{\Gamma + i\omega} \right]. \quad (33)$$

The component corresponding to the stimulated emission then vanishes. For  $\gamma_0 \gg \gamma_j$  (33) goes over into a formula obtained by other authors<sup>[3,4]</sup>. For large detunings  $\Delta \gg V$  we return to the case of perturbation theory, to which corresponds the formula (29).

## 5. RESONANCE FLUORESCENCE

When the lower level is a stable level, we must drop the common factor  $\gamma_0^{-1} \sim t \rightarrow \infty$  in the above formulas, and consider the radiation intensity per unit time. Let us give the expressions for the integrated intensities of the coherent and incoherent emissions per unit time  $\dot{J}_c$  and  $\dot{J}_{inc}$  for arbitrary values of  $V$  and  $\Delta$ :

$$\dot{J}_c = 2\gamma |F_+|^2 = \frac{\gamma A}{(1+A)^2}, \quad A = \frac{2V^2}{\Delta^2 + \gamma^2}, \quad (34)$$

$$\dot{J}_{inc} = \gamma(1+F_+) = \frac{\gamma A^2}{(1+A)^2}$$

In a weak field the dominant role is played by the coherent emission, the intensity of the incoherent radiation being of the order of  $V^4$ . On the contrary, in a strong field the radiation is primarily incoherent, the coherent component being of the order of  $V^{-2}$ . This agrees with results obtained by other authors<sup>[6,11]</sup>.

In conclusion, let us note that we do not pause to compare the theory with experiment<sup>[12,13]</sup>, since to do this would require the consideration of a three-level scheme of atomic levels and allowance for the Doppler broadening. Here, however, we considered only the homogeneous broadening. The generalization of the kinetic equation for the correlation density matrix to the case of several levels presents no difficulty; it has the same form as the equation for the density matrix,

and the boundary conditions are determined by relations of the type (5).

Note added in proof (February 25, 1974). Resonance fluorescence in a strong magnetic field was recently investigated by E. V. Baklanov (Zh. Eksp. Teor. Fiz. 65, 2203 (1973) [Sov. Phys.-JETP 38, No. 6 (1974)]).

<sup>1)</sup>Notice that in the diagrammatic description there are in this case no diagrams with intersecting interaction lines<sup>[8]</sup>.

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