COLLECTIVE EFFECTS IN SPONTANEOUS EMISSION BY TWO ATOMS

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Collective spontaneous emission by two excited atoms located at an arbitrary distance from each other is investigated. The frequency and angular dependences of the radiation and its integral intensity as a function of time are obtained. Special attention is paid to interference effects. The effect of energy splitting and Coulomb interaction on the collective features of the emission spectrum is discussed.

1. INTRODUCTION

COLLECTIVE effects in spontaneous emission of identical atoms were first considered by Dicke[1], who used the emission of atoms in a volume whose linear dimensions were much smaller than the wavelength. He determined such characteristic features of the collective radiation of atoms as the onset of metastable states and the change of the decay constants. A generalization of Dicke's problem to the case of an arbitrary distance between atoms is made difficult because of the nonconservation of the total energy spin of the system (see, for example,[2]). Therefore, for arbitrary distances, the problem could be solved only for two atoms. For an initial condition with one excited atom, the spectrum of the collective spontaneous emission and the lifetime of the excitation were considered in[3-5]. It is pointed out in[3] that the role of the Coulomb interaction is appreciable when the distance between the atoms is less than or of the order of the wavelength. The collective emission of atoms with degenerate upper level was considered in[4], and collective effects in the emission of unlike atoms (different frequencies and radiative constants) were investigated in[5]. The related problem of the spontaneous decay of an almost degenerate state was considered in[6].

Collective spontaneous emission of two excited unlike atoms was considered first in a recent paper by Varfolomeev[7], who used the Heitler-Ma method to obtain a two-photon amplitude from which the form of the spontaneous-emission spectrum can be calculated. By way of example, he presented the spectra for several concrete (not very realistic, $\gamma = \omega/20$) values of the parameters of the two-atom system. The general expression for the spectrum turns out to be two complicated and is not given in[7]. Varfolomeev calculated also the time dependence of the probability of finding only one of the atoms excited, with allowance for the Coulomb interaction and the differences between the frequencies and the radiative constants of the atoms.

In the present paper we investigated in detail the frequency and angular dependences of the spontaneous-emission spectrum of two excited atoms as a function of the ratio of the radiative widths γ, the resonance defect Δ, and the Coulomb interaction V. In particular, for the case of identical atoms we obtain an explicit expression for the form of the spectrum (19) and calculate the integrated intensity of the cross section as a function of the time (27).

The diagram method used by us for the density matrix[8] gives a clear idea of the elementary processes that were taken into account. This analysis may turn out to be useful for the investigation of collective effects in extended polyatomic systems.

2. GRAPHIC REPRESENTATION OF THE DENSITY MATRIX

We consider two-level atoms with resonant transition frequencies ω_a and ω_b; the atoms are separated by a distance **R**. The operator of the interaction of the atoms with the elctromagnetic field, for a Coulomb gauge of the vector potential **A**, is taken in the form

$$\hat{H}_I = -\sum_{i=a,b} \hat{\mathbf{A}}(\mathbf{R}_i)\hat{\mathbf{p}}_i + \hat{V} = \sum_{\mathbf{q}} \sum_{i=a,b} \{f_{\mathbf{q}} \exp(i\mathbf{q}\mathbf{R}_i)\hat{a}_{\mathbf{q}}\hat{\sigma}_i^{+} + \text{h.c.}\} + \hat{V}, \quad (1)$$

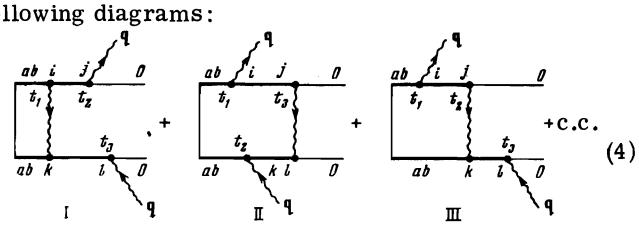
$$f_{\mathbf{q}} = -i\left(\frac{2\pi}{q}\right)^{1/2} \frac{\omega_0}{\sqrt{L^3}} \mathbf{d}\mathbf{e}_{\mathbf{q}}, \quad (2)$$

where $q = |\mathbf{q}|$ and $2\omega_0 = \omega_a + \omega_b$. Here $\hat{V}$ is the operator of the Coulomb interaction of the atoms, a_q is the annihilation operator of a photon of mode $\hat{q}$, and $\hat{\sigma}_i^{+}$ is the creation operator for the excitation of the i-th atom. The symbol q includes the wave vector and the polarization of the photon. The unit vector for the polarization of the photon q is designated by e_q. The matrix elements of the dipole-moment operator between the ground and the excited states of each atom are assumed to be identical for both atoms and equal to d.

Assume that both atoms are excited at the initial instant. If we know the density matrix $\hat{\rho}$ of the system, we can calculate the emission spectrum

$$\langle \hat{n}_{\mathbf{q}} \rangle = \lim_{t\to\infty} \mathrm{Sp}(\hat{n}_{\mathbf{q}}\hat{\rho}(t)). \quad (3)$$

Expression (3) can be set in correspondence with the following diagrams:

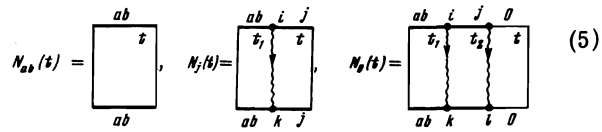


(4)

Here $|a\rangle$ ($|b\rangle$) is a state in which only one atom a (b) is excited; $|0\rangle$ is the ground state of the atomic system and $|ab\rangle$ is the fully excited state.

We formulate briefly the rules for setting the diagrams in correspondence with analytic expressions. The upper straight line corresponds to the propagators $G_{ab,ab}(t)$ and $G_{i,j}(t)$ of the atomic system. The vertex on the upper line corresponds to the factor $-if_q$; the lower line corresponds to analogous complex-conjugate expressions. Summation is over all the intermediate-state indices, and the nonresonant transitions are disregarded. The internal photon line corresponds to pairing of the creation and annihilation operators and to summation over all the radiation oscillators. The approximation in which the internal photon lines are vertical corresponds to discarding terms of order γ/ω_0, where γ is the radiative constant of the atom. Consecutive integration with respect to the times is carried out.

The state populations in the time dependence for the atomic subsystem can be set in correspondence with the diagrams



(5)

They yield in obvious fashion the average number of photons as a function of the time

$$n(t) = \mathrm{Sp}\,(\hat{n}\hat{\rho}(t)) = N_a(t) + N_b(t) + 2N_0(t). \tag{6}$$

The normalization condition is

$$N_0 + N_a + N_b + N_{ab} = 1. \tag{7}$$

To calculate the diagrams it is necessary to know the atomic propagators. From relations such as Dyson's equations it follows that the Laplace transforms of the propagators satisfy the system

$$\begin{aligned} G_{ab,ab} &= G^0_{ab,ab} + G^0_{ab,ab}\,(\Sigma_{aa} + \Sigma_{bb})\,G_{ab,ab}, \\ G_{a,a} &= G^0_{a,a} + G^0_{a,a}\,\Sigma_{a,a}G_{a,a} + G^0_{a,a}(\Sigma_{ab} - iV)\,G_{b,a}, \\ G_{b,a} &= G^0_{b,b}\Sigma_{bb}G_{b,a} + G^0_{b,b}(\Sigma_{ba} - iV)\,G_{a,a}, \end{aligned} \tag{8}$$

where, for example, $G^0_{a,a}(p)$ is the free propagator of the state $|a\rangle$, V is the matrix element of the Coulomb interaction, and $\Sigma(p)$ is the self-energy part;

$$\begin{aligned} G^0_{a,a}(p) &= (p + i\omega_a)^{-1}, \qquad V = \langle a|\hat{V}|b\rangle, \\ \Sigma_{ij}(p) &= \sum_q \frac{|f_q|^2 \exp(iqR_{ij})}{-p - iq}. \end{aligned} \tag{9}$$

As usual, we neglect the dependence of $\Sigma_{ij}(p)$ on the Laplace variable p, putting $p = -i\omega_0 - \epsilon$, $\epsilon \to 0$. The radiative-decay constant of an individual atom is equal to $\gamma/2 = -\mathrm{Re}\,\Sigma_{aa} = -\mathrm{Re}\,\Sigma_{bb}$. Neglecting the Lamb shift, we put $\mathrm{Im}\,\Sigma_{aa} = 0$, $\mathrm{Im}\,\Sigma_{bb} = 0$ (see[9]). For the off-diagonal self-energy parts we introduce the notation

$$\Sigma_{ab} = \Sigma_{ba} = {}^1\!/_2\tilde{\gamma} + iu. \tag{10}$$

The real part (10) as a function of R and of the angle θ between R and d is (see, for example,[10])

$$\tilde{\gamma} = \frac{3}{2}\gamma\left\{\sin^2\theta\,\frac{\sin qR}{qR} + (2\cos^2\theta - \sin^2\theta)\left(\frac{\sin qR}{(qR)^3} - \frac{\cos qR}{(qR)^2}\right)\right\}. \tag{11}$$

The imaginary part of (10) describes interaction via a transverse electromagnetic field and can be included in V, as can be seen from the structure of Eqs. (8). The solution of the system (8) is

$$\begin{aligned} G_{ab,ab}(p) &= (p + 2i\omega_0 + 2\gamma)^{-1}, \\ G_{a,a}(p) &= \frac{p + i\omega_b + \gamma/2}{(p - p_1)(p - p_2)}, \qquad G_{b,a}(p) = \frac{-(\tilde{\gamma}/2 + iV)}{(p - p_1)(p - p_2)}, \end{aligned} \tag{12}$$

where

$$p_{1,2} = -i\omega_0 - {}^1\!/_2\gamma \pm [({}^1\!/_2\tilde{\gamma} + iV)^2 - \Delta^2]^{1/2}, \qquad 2\Delta = \omega_a - \omega_b. \tag{13}$$

At $|\Delta| \gg \tilde{\gamma}$, V the poles of the propagators correspond to non-interacting atoms, and there are no collective effects. However, if the Coulomb interaction is large, $|V| \gg |\Delta|$, then the radiative constant in (13) are equal to $\gamma \pm \tilde{\gamma}$ independently of the ratio of the quantities Δ and γ. The reason is that in this case the damping matrix ($\gamma_{aa} = \gamma_{bb} = \gamma$, $\gamma_{ab} = \gamma_{ba} = \tilde{\gamma}$) and the energy matrix are simultaneously diagonalized.

3. FORM OF THE EMISSION SPECTRUM

It can be assumed that a characteristic relation for real systems is $|\Delta| > \gamma$ (the inhomogeneity is larger than the radiative width). Therefore the collective effects will come into play, according to (13), only under the condition $|V| > |\Delta|$, and the frequency detuning can be neglected when $|V| \gg |\Delta|$. We consider first the spectrum of the spontaneous emission of identical atoms ($\omega_a = \omega_b$), and discuss the results pertaining to the general case later on. The most natural for the case of identical atoms is a representation using the symmetrized basis

$$|S\rangle = \frac{|a\rangle + |b\rangle}{\sqrt{2}}, \qquad |A\rangle = \frac{|a\rangle - |b\rangle}{\sqrt{2}}, \tag{14}$$

in which both the damping matrix and the energy matrix reduce to diagonal form. The atomic propagators in this basis are also diagonal

$$G_{S,S} = (p - p_2)^{-1}, \quad G_{A,A} = (p - p_1)^{-1}, \quad G_{A,S} = G_{S,A} = 0; \tag{15}$$

$$p_{1,2} = -i\omega_0 - {}^1\!/_2\gamma \pm ({}^1\!/_2\tilde{\gamma} + iV). \tag{16}$$

It is now necessary to sum in the diagrams of (4) over i, j, k, l = S, A. Since the propagators are diagonal and some of the interconnections vanish, the intermediate indices S and A do not appear simultaneously in any one of the diagrams of (4), so that the spectrum is a sum of two groups of terms, one of which corresponds to decay via the "symmetrical" channel and the other via the "antisymmetrical" one.

We calculate diagram I of expression (4) for the intermediate state S:

$$\begin{aligned} \langle n_q\rangle^{(I)} &= (\gamma + \tilde{\gamma})\,(1 + \cos\mathbf{qR})\,|f_q|^2 \int_0^\infty dt_3 \int_0^{t_3} dt_2 \int_0^{t_2} dt_1 \cdot \\ &\cdot \exp(-2\gamma t_1 + iq(t_2 - t_3))\,G_{SS}(t_2 - t_1)\,G_{SS}^*(t_3 - t_1) + \text{c.c.}. \end{aligned} \tag{17}$$

It is convenient to regard this expression as the Laplace transform as $p \to 0$; using (15), we obtain

$$\begin{aligned} \langle n_q\rangle^{(I)} = (\gamma + \tilde{\gamma})\,(1 + \cos\mathbf{qR})\,|f_q|^2 [2\gamma(\gamma + \tilde{\gamma})\{i(q - \omega_0 + V) \\ + {}^1\!/_2(\gamma + \tilde{\gamma})\}]^{-1} + \text{c.c.} \end{aligned} \tag{18}$$

The remaining diagrams are calculated analogously. After simple transformations we obtain the spectrum

$$\begin{aligned} \langle n_q\rangle = \pi\gamma^{-1}|f_q|^2\{(1 + \cos\mathbf{qR})\,[(1 - \alpha)L_{\gamma(3+\delta)/2}(\nu + V) \\ + (1 + \alpha)L_{\gamma(1+\delta)/2}(\nu - V) + \beta(X_{\gamma(3+\delta)/2}(\nu + V) - X_{\gamma(1+\delta)/2}(\nu - V))] \end{aligned}$$

$$+ (1-\cos \mathbf{qR})\,[(1-\alpha')L_{\gamma(3-\delta)/2}(\nu - V) + (1+\alpha')L_{\gamma(1-\delta)/2}(\nu + V) - \beta'(X_{\gamma(3-\delta)/2}(\nu - V) - X_{\gamma(1-\delta)/2}(\nu + V))]\}. \quad (19)$$

We have introduced here the following notation:

$$L_\gamma(\omega) = \frac{\gamma}{\pi(\gamma^2+\omega^2)}, \quad X_\gamma(\omega) = \frac{\omega}{\pi(\gamma^2+\omega^2)},$$
$$\beta = \frac{2v(1+\delta)}{1+4v^2}, \quad \beta' = \frac{2v(1-\delta)}{1+4v^2}, \quad (20)$$
$$\alpha = \frac{1+\delta}{1+4v^2}, \quad \alpha' = \frac{1-\delta}{1+4v^2}, \quad \delta = \frac{\tilde{\gamma}}{\gamma}, \quad v = \frac{V}{\gamma}, \quad \nu = q - \omega_0.$$

To interpret the spectrum, we consider the level scheme of the system (see Fig. 1). The terms with the factor $(1 + \cos \mathbf{q}\cdot\mathbf{R})$ in (19) corresponds to the decay of the system via the symmetrical channel, with transitions 1 and 2 of Fig. 1 corresponding to Lorentzians with widths $\gamma(3+\delta)/2$ and $\gamma(1+\delta)/2$, respectively. The positions and the widths of the Lorentzians are determined by the splitting and the widths of the corresponding levels. The quantum-mechanical interference of the transitions, connected with the equidistant character of the system, is described by the diagram III of (4) and leads to an increase of the weight of the narrow Lorentzian, to a decrease of the weight of the broad Lorentzian, and to the appearance of interference terms X, which bring the lines closer together and disturb their symmetry. We note that as $V \to 0$ the contribution of the interference terms X tends to zero, and in this case the role of the interference reduces to a redistribution of the weights of the Lorentzians. A similar interference narrowing in Dicke's problem was considered in[2]. Our result agrees in this limit with the corresponding formula of[2]. The terms in the spectrum with the factor $(1 - \cos \mathbf{q}\cdot\mathbf{R})$ corresponds to decay via the "antisymmetrical" channel. The positions and widths of the lines correspond to transitions 3 and 4 (see Fig. 1). The interference of the transitions in the "antisymmetrical" channel plays a role analogous to the interference in the "symmetrical" channel.

The angular dependence of the spectrum (19) is also easy to interpret. The photons interacting most effectively with the symmetrical intermediate state are those for which the phase difference $\mathbf{q}\cdot\mathbf{R}$ is equal to $0, \pm 2\pi, \ldots$ In the case of the antisymmetrical intermediate state, these are the photons for which $\mathbf{q}\cdot\mathbf{R} = \pm\pi, \pm 3\pi \ldots$. The spectrum observed in an arbitrary direction is a sum of two independent spectra with weights given by (19). In the limit as $qR \ll 1$ (the Dicke problem), the decay from a symmetrical doubly-excited state is possible only via the symmetrical channel, since the interaction H_I becomes symmetrical with respect to the atoms. The characteristic form of the spectrum (19), averaged over the radiation direction at $|V| \gg \gamma$ and $\delta \sim 1$, is shown in Fig. 2.

In the more general case ($\omega_a \neq \omega_b$) the emission spectrum is also characterized by the presence of four Lorentz curves, two each at two frequencies corresponding to the imaginary part of the poles (13). Each Lorentz curve is accompanied by an interference increment X. The coefficients of the spectral components are quite cumbersome in the general case, and we therefore discuss here limiting cases that provide, together with (19), a sufficiently full picture of the spectrum. When the atoms are separated by a small

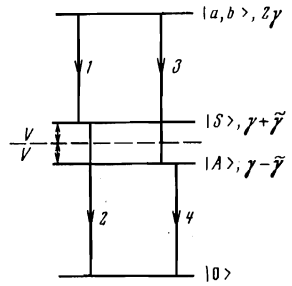


FIG. 1. Possible transitions in a system of two atoms.

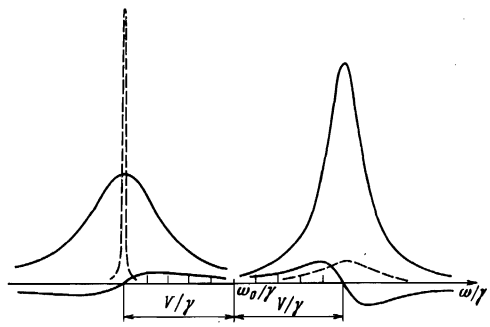


FIG. 2. Components of emission spectrum averaged over the directions at $V/\gamma = 5$ and $\delta = 0.9$. The dashed lines represent the contribution of the "antisymmetrical" channel.

distance ($qR \ll 1$), so that $\tilde{\gamma} = \gamma$ and $|V| \gg \gamma$, the difference between the frequencies of the atoms leads to insignificant changes in the form of the spectrum in comparison with (19). If the condition $|V| \gg |\Delta|$ is satisfied, then the positions and widths of the spectral components, and also the coefficients of the latter, differ from the spectrum (19) only starting with the second order of smallness in the parameter $|\Delta/V| \ll 1$. The only significant difference between the spectrum in this case and the spectrum (19) is the presence of an additional narrow component, which in second order in Δ/V is given by

$$\frac{\pi}{\gamma}|f_q|^2\left(\frac{\Delta}{V}\right)^2 L_{\gamma(\Delta/V)^2/2}\left[\nu + V\left(1+\frac{1}{2}\left(\frac{\Delta}{V}\right)^2\right)\right]. \quad (21)$$

Although the system decays at $qR \ll 1$ only via the "symmetrical" channel, the difference between the frequencies of the atoms leads to quantum oscillations between the states $|A\rangle$ and $|S\rangle$. Since the state $|A\rangle$ is stationary with respect to the radiative decay, the relaxation of the excitation in the system slows down, and this leads to addition of a narrow component to the spectrum.

We consider now the spectrum for medium distances between the atoms ($qR \gtrsim 1$). We set V equal to zero, since the Coulomb interaction between the atoms decreases with distance more rapidly than $\tilde{\gamma}$ (for $\tilde{\gamma}$, as seen from (11), there is a characteristic decrease like $(qR)^{-1}$). The spectrum averaged over the directions takes the following form at $V = 0$ and $|\Delta| > \gamma/2$:

$$\langle n_q\rangle_\Omega = \pi\gamma^{-1}\langle|f_q|^2\rangle_\Omega\{\alpha_1(L_{\gamma/2}(\gamma+\Omega_0) + L_{\gamma/2}(\nu-\Omega_0)) - \alpha_2(L_{3\gamma/2}(\nu+\Omega_0) + L_{3\gamma/2}(\nu-\Omega_0)) + \beta_1(X_{\gamma/2}(\nu+\Omega_0) - X_{\gamma/2}(\nu-\Omega_0)) + \beta_2(X_{3\gamma/2}(\nu+\Omega_0) - X_{3\gamma/2}(\nu-\Omega_0))\}, \quad (22)$$

where

$$\Omega_0 = \sqrt{\Delta^2 - (\tilde{\gamma}/2)^2}, \quad \lambda = \Omega_0/\gamma,$$
$$\alpha_1 = 2 + \frac{\delta^2(1-\delta^2)}{1+4\lambda^2}, \quad \alpha_2 = \frac{\delta^2(1-\delta^2)}{1+4\lambda^2},$$

$$\beta_1=\frac{\delta^2}{2\lambda}\left(2+\frac{1-\delta^2}{1+4\lambda^2}\right),\quad \beta_2=\frac{\delta^2}{2\lambda}\left(1+\frac{\delta^2+4\lambda^2}{1+4\lambda^2}\right). \quad (23)$$

As follows from (22), the Lorentz curves are shifted in this case towards each other relative to the atomic frequencies. The broad Lorentzians, which enter with a negative weight, ensure a faster decrease of the lines on the wings. The interference terms X lead to a mutual approach and distortion of the lines. When the inequality $|\Delta| \gg \gamma$ is satisfied, the spectrum (22) goes over into the emission spectrum of independent atoms.

4. DYNAMICS OF THE EMISSION

We now obtain the time and angle dependences of the number of emitted photons. We consider identical atoms ($\omega_a = \omega_b$) with allowance for the Coulomb interaction. It is obvious that the angular dependence for a decay via the "symmetrical" and "antisymmetrical" channels is the same as for the spectrum (19). Therefore, if $n^{S(A)}(t)$ is the average number of photons in the field as a result of decay via the "symmetrical" ("antisymmetrical") channel, then

$$W^{s(A)}(\Omega, t)=n^{s(A)}(t)\frac{(1\pm\cos \mathbf{q}_0\mathbf{R})\,(\mathbf{de_q})^2}{\langle(1\pm\cos \mathbf{q}_0\mathbf{R})\,(\mathbf{de_q})^2\rangle_\Omega} \quad (24)$$

is the respective number of photons emitted by the instant t into a unit solid angle. Here $\mathbf{q}_0$ is the vector of the given direction; $|\mathbf{q}_0| = \omega_0$. The averaging over the directions is designated by the angle brackets. The quantities $n^{S(A)}(t)$ are determined from the relations

$$n^s(t)=N_s(t)+2N_0^{(s)}(t),\quad n^A(t)=N_A(t)+2N_0^{(A)}(t), \quad (25)$$

where $N_S(t)$ is the probability of finding the system in the intermediate state $|S\rangle$, and $N_0^{(S)}(t)$ is the probability of decay to the ground state via the "symmetrical" channel.

For $N_i(t)$ (see (5)) at i = S we obtain

$$N_S(t)=(\gamma+\tilde\gamma)\int_0^t dt_1 e^{-2\gamma t_1}|G_{S,S}(t-t_1)|^2=\frac{1+\delta}{1-\delta}e^{-2\gamma t}\,(e^{(\gamma-\tilde\gamma)t}-1). \quad (26)$$

Calculating analogously the expressions for $N_A(t)$, $N_0^{(S)}(t)$, $N_0^{(A)}(t)$ and using (25), we get

$$\begin{aligned} n^s(t) &= (1+\delta)\{1-(1-\delta)^{-1}e^{-2\gamma t}(e^{(\gamma-\tilde\gamma)t}-\delta)\},\\ n^A(t) &= (1-\delta)\{1-(1+\delta)^{-1}e^{-2\gamma t}(e^{(\gamma+\tilde\gamma)t}+\delta)\}. \end{aligned} \quad (27)$$

from the obtained relations it follows that the dynamics of the decay is independent of the Coulomb interaction at $\omega_a = \omega_b$. If $qR \ll 1$, then the "antisymmetrical" channel is "closed" and the total number of the photons emitted by the instant of time t is then given by

$$n(t)=2\{1-e^{-2\gamma t}(1+\gamma t)\}, \quad (28)$$

which agrees with the corresponding result of[2]. At $qR \gg 1$, i.e., as $\delta \to 0$, we obtain the decay of independent atoms.

5. CONCLUSION

Collective effects in the emission of two atoms can come into play when the distances between them are smaller than or comparable with the radiation wavelength. At distances greatly exceeding the wavelength, the off-diagonal damping-matrix elements tend to zero, and the atoms emit independently. The collective effect depends then on the mutual orientation of the polarizations of the excitations. It is maximal when the polarizations are parallel and is zero for perpendicular polarizations. If the Coulomb interaction between the atoms is small, as is the case for relatively large distances, then the collective effect depends on the degree of resonance, namely, the difference between the frequencies of the atoms must not exceed the natural width of the level. This condition is too stringent for real systems, owing to the inhomogeneous broadening. If, however, the Coulomb interaction is larger than the inhomogeneity, then the energy eigenstates will coincide with the collective radiative states. This case is apparently the most realistic for the manifestation of collective radiative states.

The spectrum of the spontaneous emission of two identical excited atoms consists of two lines, the positions of which are determined by the magnitude of the interaction (see Fig. 1). Each of the lines is a superposition of two Lorentzians with widths $3\gamma + \tilde\gamma$, $\gamma - \tilde\gamma$, $3\gamma - \tilde\gamma$, $\gamma + \tilde\gamma$ ($0 \le \tilde\gamma \le \gamma$). The interference of the corresponding transitions distorts somewhat the shapes of the Lorentzians. If the Coulomb interaction exceeds the natural width, this distortion is small. However, in the absence of interaction (the Dicke problem), the interference causes the spectrum to contain only one line with characteristic width 2γ. This quantity determines also the law governing the decay of the excitation.

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