

COHERENT EFFECTS IN SPONTANEOUS EMISSION BY UNLIKE ATOMS

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The emission spectrum and the spontaneous-decay probability are calculated for a system of two two-level atoms, one of which is excited at the initial instant of time. The energy levels of the excited states of the atoms are assumed to be close, and the distance between the atoms is assumed fixed. Account is taken of the electromagnetic interaction of the atoms. The coherent effects in the radiation are analyzed. It is shown, in particular, that unlike the case of like atoms, no metastable states are produced in a system of unlike atoms, and all the states decay rapidly.

1. INTRODUCTION

As is well known, the presence of like oscillators close to an excited oscillator leads to a coherent effect in spontaneous emission<sup>[1-6]</sup>. This effect was first described by Dicke<sup>[1]</sup>, who considered the interaction of identical two-level molecules occupying a small volume compared with the radiation wavelength. He has shown that the interaction of the oscillators via the radiation field causes the molecules to behave like a single quantum-mechanical system. The coherence of the individual oscillators can lead both to intensification of the radiation (superradiant states) and to the suppression of the radiation (metastable states). The Dicke problem was considered later in different variants and by different methods (see, for example,<sup>[2-4]</sup>). The emission from two identical two-level atoms at a definite distance from each other was considered in<sup>[5-6]</sup>. Unlike the formulation in the Dicke problem, the Coulomb interaction of the atom was taken into account in<sup>[5]</sup>.

In the present paper we determine the characteristics of the spontaneous decay in a system of two atoms that are not identical, although they have close excited-state levels. We consider in detail coherent effects in strong Coulomb interaction between atoms (at small distances between atoms).

2. CALCULATION CONDITIONS AND RESULTS

We consider the interaction of two two-level atoms A and B with close (or identical) excitation levels  $\hbar\omega_{1A}$  and  $\hbar\omega_{1B}$ . We assume that the atoms are at a distance R from each other; this distance is comparable with the wavelength of the resonant radiation  $c/\omega_{1A}$ . We take into account the interaction of the atoms via the electromagnetic field, without neglecting retardation effects. To simplify the problem, we assume that the wave functions of the atoms do not overlap. In the emission and motion of the atoms, we shall likewise disregard the recoil energy. In principle, these phenomena can be taken into account in the final results by introducing corresponding shifts and broadenings of the levels of the individual atoms. As the initial condition we assume that the atom A is excited at the instant  $t = 0$ .

The state of the system can be described in general form by a wave function in the Schrödinger representation:

$$\Psi(t) = b_{1A}(t)\Psi_{1A} + b_{1B}(t)\Psi_{1B} + \sum_{q\mu} b_{q\mu}(t)\Psi_{q\mu}$$

where b are the coefficients of the expansion, which include exponential time-dependent factors, and the symbols designating the coefficients in the wave functions mean that the atom A or B is excited, or that a quantum with wave vector q and polarization  $\mu$  is present in the field. For simplicity we disregard states with large numbers of virtual quanta, since they can only lead to insignificant corrections to the level shifts and to the decay constants.

The Hamiltonian of the interaction of the system is written in the form

$$\mathcal{H}_{int} = H + V,$$

where H describes the interaction with the transverse field, and the operator V is determined by the electrostatic interaction (Coulomb gauge). In the nonrelativistic approximation without allowance for the two-quantum transitions we have

$$H = - \sum_i \frac{e}{mc} \mathbf{A}(\mathbf{r}_{Ai}) \mathbf{p}_{Ai} - \sum_j \frac{e}{mc} \mathbf{A}(\mathbf{r}_{Bj}) \mathbf{p}_{Bj}$$

where the standard notation is used: e is the electron charge, m the electron mass,  $\mathbf{r}_i$  are the radius vectors of the electrons in the atoms, and  $\mathbf{p}_i$  are the momentum operators. The vector potential of the field in the volume  $\Omega$  is

$$\mathbf{A}(\mathbf{r}) = \sum_{q\mu} \left( \frac{2\pi\hbar c}{q\Omega} \right)^{1/2} \mathbf{e}_{q\mu} (a_{q\mu} e^{i\mathbf{q}\cdot\mathbf{r}} + a_{q\mu}^+ e^{-i\mathbf{q}\cdot\mathbf{r}}),$$

where  $\mathbf{e}_{q\mu}$  is the polarization vector, and  $a_{q\mu}$  and  $a_{q\mu}^+$  are the annihilation and creation operators of the photon  $\mathbf{q}, \mu$ .

The Schrödinger equation for the system, with allowance for the single-quantum transitions, leads to the system

$$i\dot{b}_{1A} = \omega_{1A} b_{1A} + V_{AB} \hbar^{-1} b_{1B} - i \sum_{q\mu} P_A(\mathbf{q}\mu) b_{q\mu}, \tag{1}$$

$$i\dot{b}_{1B} = \omega_{1B} b_{1B} + V_{BA} \hbar^{-1} b_{1A} - i \sum_{q\mu} P_B(\mathbf{q}\mu) b_{q\mu}, \tag{2}$$

$$i\dot{b}_{q\mu} = \omega_{q\mu} b_{q\mu} + iP_A^*(\mathbf{q}\mu) b_{1A} + iP_B^*(\mathbf{q}\mu) b_{1B}. \tag{3}$$

Here  $V_{AB} = \langle A_1B | V | AB_1 \rangle$  and respectively  $V_{BA} = \langle AB_1 | V | A_1B \rangle$  are the matrix elements of the Coulomb-interaction operator without the time-dependent factor; these elements connect the states  $A_1B$  and  $AB_1$ ;

$$-i\hbar P_A(\mathbf{q}\mu) = -\frac{e}{m} \left( \frac{2\pi\hbar}{cq\Omega} \right)^{1/2} \sum_i \mathbf{p}_{Ai} e_{q\mu} e^{i\mathbf{q}\cdot\mathbf{r}_i}$$

is the matrix element of the interaction of the atom A with the transverse field.

We seek the solution of the system (1)–(3) in the form

$$b_{1A} = \exp\{-i(\omega_{1A} + \omega_{1B})t/2\} (Ce^{-i\omega t} + De^{-i\omega' t}), \quad (4)$$

$$b_{1B} = \exp\{-i(\omega_{1A} + \omega_{1B})t/2\} E(e^{-i\omega t} - e^{-i\omega' t}), \quad (5)$$

assuming the amplitudes C, D, and E and the frequencies  $\omega$  and  $\omega'$  to be the unknowns. Using (4) and (5), we get from (3)

$$-ib_{q\mu} = CP_A^*(\mathbf{q}\mu)f(\omega, \omega_1)e^{-i(\omega_1+\omega)t} + DP_A^*(\mathbf{q}\mu)f(\omega', \omega_1)e^{-i(\omega_1+\omega')t} + EP_B^*(\mathbf{q}\mu)f(\omega, \omega_1)e^{-i(\omega_1+\omega)t} - EP_B^*(\mathbf{q}\mu)f(\omega', \omega_1)e^{-i(\omega_1+\omega')t}, \quad (3')$$

where

$$\omega_1 = (\omega_{1A} + \omega_{1B})/2, \\ f(\omega, \omega_2) = \{1 - \exp[i(\omega_1 + \omega - \omega_2)t]\} / (\omega_1 + \omega - \omega_2).$$

We shall assume that the damping of the system is relatively small:  $|\omega'| \ll \omega_1$ ,  $|\omega| \ll \omega_1$ . We confine ourselves also to the case of close resonant frequencies,  $|\omega_{1A} - \omega_{1B}| \ll \omega_1$  and not too small time intervals  $\omega_1 t \gg 1$ . When these conditions are satisfied, we can use the well-known relation

$$f(\omega, \omega_1) = P(\omega_1 - \omega_q)^{-1} - i\pi\delta(\omega_1 - \omega_q) = f_1(\omega_1).$$

The expression obtained for  $b_{q\mu}$  makes it possible to transform (1) and (2) as follows:

$$C(-\Delta\omega_1 + \omega + i\gamma_A)e^{-i\omega t} + D(-\Delta\omega_1 + \omega' + i\gamma_A)e^{-i\omega' t} = U_{AB}\hbar^{-1}E(e^{-i\omega t} - e^{-i\omega' t}), \quad (1')$$

$$E(\Delta\omega_1 + \omega + i\gamma_B)e^{-i\omega t} - E(\Delta\omega_1 + \omega' + i\gamma_B)e^{-i\omega' t} = U_{BA}\hbar^{-1}(Ce^{-i\omega t} + De^{-i\omega' t}). \quad (2')$$

Here we have introduced for the brevity the notation

$$\omega_{1A} - \omega_{1B} = 2\Delta\omega_1, \\ -i\gamma_A = \sum_{q\mu} P_A(\mathbf{q}\mu)P_A^*(\mathbf{q}\mu)f_1(\omega_1), \quad -i\gamma_B = \sum_{q\mu} P_B(\mathbf{q}\mu)P_B^*(\mathbf{q}\mu)f_1(\omega_1) \quad (6)$$

$$U_{AB} = \sum_{q\mu} \hbar P_A(\mathbf{q}\mu)P_B^*(\mathbf{q}\mu)f_1(\omega_1) + V_{AB},$$

$$U_{BA} = \sum_{q\mu} \hbar P_B(\mathbf{q}\mu)P_A^*(\mathbf{q}\mu)f_1(\omega_1) + V_{BA}. \quad (7)$$

Using the asymptotic expression for  $f_1(\omega_1)$  and carrying out the corresponding summation in (6), we can readily verify that  $2\text{Re } \gamma_A$  and  $2\text{Re } \gamma_B$  represent the natural level widths of the respective individual atoms, while  $2\text{Im } \gamma_A$  and  $2\text{Im } \gamma_B$  are small radiative shifts of these levels<sup>[7]</sup>, which will henceforth be assumed equal to zero.

The calculations connected with the summation in expressions (7) are somewhat more cumbersome. We note, however, that  $U_{AB}$  and  $U_{BA}$  constitute matrix elements of the retarded interaction of two atoms, corresponding to the exchange of a photon between them. According to<sup>[8]</sup>, we can write in a nonrelativistic approximation

$$U_{AB}(\omega_1, R) = \left\langle A_1 B \left| e^2 \sum_{ij} \frac{\exp\{i\omega_1|\mathbf{r}_{A_i} - \mathbf{r}_{B_j}|c^{-1}\}}{|\mathbf{r}_{A_i} - \mathbf{r}_{B_j}|} \left(1 - \frac{\mathbf{p}_{A_i}\mathbf{p}_{B_j}}{m^2c^2}\right) \right| AB \right\rangle. \quad (8)$$

If both atoms have nonzero electric dipole moments

$$\mathbf{d}_A = \left\langle A_1 \left| -\sum_i e\mathbf{r}_i \right| A \right\rangle, \quad \mathbf{d}_B = \left\langle B_1 \left| -\sum_j e\mathbf{r}_j \right| B \right\rangle,$$

then the matrix element  $U_{AB}$  takes the form

$$U_{dd}(\omega_1, R) = q^3 e^{iqR} \left\{ \frac{1}{qR} \left[ \frac{(\mathbf{R}\mathbf{d}_A)(\mathbf{R}\mathbf{d}_B)}{R^2} - \mathbf{d}_A\mathbf{d}_B \right] + \left( \frac{1}{iq^2R^2} + \frac{1}{q^2R^3} \right) \left[ \mathbf{d}_A\mathbf{d}_B - 3 \frac{(\mathbf{R}\mathbf{d}_A)(\mathbf{R}\mathbf{d}_B)}{R^2} \right] \right\}. \quad (9)$$

If  $qR \ll 1$ , then

$$\text{Re } U_{dd} = \frac{\mathbf{d}_A\mathbf{d}_B R^2 - 3(\mathbf{d}_A\mathbf{R})(\mathbf{d}_B\mathbf{R})}{R^3} = V_{dd}, \quad \text{Im } U_{dd} \approx \hbar\overline{\gamma_A\gamma_B}. \quad (9')$$

The interaction of two atoms, one of which has an allowed transition and the other (for example the atom A) has a nonzero electric quadrupole moment

$$Q_{\alpha\beta} = \left\langle A_1 \left| -\sum_i e(x_{i\alpha}x_{i\beta} - r_i^2/3) \right| A \right\rangle,$$

i.e., the dipole-quadrupole interaction, is described by the matrix element

$$U_{Qd}(\omega_1, R) = \frac{1}{2} e^{iqR} \sum_{\alpha\beta} Q_{\alpha\beta} \left\{ R_\alpha R_\beta \frac{(\mathbf{R}\mathbf{d})}{R^2} [15(1 - iqR) - 6q^2R^2 + iq^3R^3] - \frac{R_\beta d_\alpha + R_\alpha d_\beta}{2R^2} [6(1 - iqR) - 3q^2R^2 + iq^3R^3] \right\}, \quad (10)$$

$$\text{Re } U_{Qd} = \sum_{\alpha\beta} Q_{\alpha\beta} \left( \frac{15}{2} R_\alpha R_\beta \frac{\mathbf{R}\mathbf{d}}{R^2} - \frac{3}{2} \frac{R_\beta d_\alpha + d_\beta R_\alpha}{R^2} \right) = V_{Qd}, \\ \text{Im } U_{Qd} = 0. \quad (10')$$

From the structure of expressions (8)–(10) it follows that  $U_{AB} = U_{BA}$  ( $\mathbf{d}$  and  $Q_{\alpha\beta}$  are real quantities).

After determining (6) and (7), the solution of the system (1'), (2'), entails no difficulty. Equating the coefficients of like exponents and using the initial conditions  $C + D = 1$ , we obtain a system of algebraic equations, which leads to the solution

$$2i\omega = \gamma_A + \gamma_B - \beta, \quad 2i\omega' = \gamma_A + \gamma_B + \beta, \quad E = U_{AB}/\hbar\beta, \\ 2C = 1 - (\gamma_A + i\omega_{1A} - \gamma_B - i\omega_{1B})/\beta, \\ 2D = 1 + (\gamma_A + i\omega_{1A} - \gamma_B - i\omega_{1B})/\beta; \\ \beta^2 = (\gamma_A + i\omega_{1A} - \gamma_B - i\omega_{1B})^2 - 4U_{AB}^2\hbar^{-2}.$$

The form of the functions (4) and (5) enables us to draw certain general conclusions concerning the behavior of the system of atoms in time. The probability of radiation dragging by the system can be defined as

$$|b_{1A}|^2 + |b_{1B}|^2 = \exp\{-(\gamma_A + \gamma_B)t\} \{ (|C|^2 + |E|^2) \exp\{\text{Re}\beta t\} + (|D|^2 + |E|^2) \exp\{-\text{Re}\beta t\} + 2[\text{Re}(CD^*) - |E|^2] \cos \text{Im}\beta t - 2\text{Im}(CD^*) \sin \text{Im}\beta t \}. \quad (12)$$

Thus, the real part of  $\beta$  determines, in addition to  $\gamma_A$  and  $\gamma_B$ , also the damping of the system, while the imaginary part determines the frequency of the oscillations and the excitations between the atoms, and together with it also the frequency of the oscillations of the excited state of the entire system. Accordingly, besides the exponential decrease of the radiation intensity, one should observe, generally speaking, radiation beats analogous to those observed in coherent radiation with close energy levels<sup>[9]</sup>. Characteristically, the radiation beats vanish in the case of identical atoms.

The coherent effects significantly alter the spectrum of the spontaneous emission. The radiation line shape can easily be obtained from (3') by summing over the polarizations and averaging over the angles

$$\sum_{\mu \neq \nu} |b_{\mu\nu}|^2 = \frac{2\pi c}{\Omega q} \left\{ \frac{|C|^2 \gamma_A - 2 \operatorname{Re}(CE^*) \operatorname{Im} U_{AB} \hbar^{-1} + |E|^2 \gamma_B}{(\Delta\omega)^2 + \Gamma^2/4} + \frac{|D|^2 \gamma_A + 2 \operatorname{Re}(DE^*) \operatorname{Im} U_{AB} \hbar^{-1} + |E|^2 \gamma_B}{(\Delta\omega')^2 + \Gamma'^2/4} + 2 \operatorname{Re} \frac{CD^* \gamma_A - (ED^* - E^*C) \operatorname{Im} U_{AB} \hbar^{-1} - |E|^2 \gamma_B}{(\Delta\omega - i\Gamma/2)(\Delta\omega' + i\Gamma'/2)} \right\},$$

where

$$2\Delta\omega = \omega_{1A} + \omega_{1B} - 2\omega_0 - \operatorname{Im}\beta, \quad \Gamma = \gamma_A + \gamma_B - \operatorname{Re}\beta, \\ 2\Delta\omega' = \omega_{1A} + \omega_{1B} - 2\omega_0 + \operatorname{Im}\beta, \quad \Gamma' = \gamma_A + \gamma_B + \operatorname{Re}\beta.$$

We see that the radiation line shape becomes Lorentzian in the case of identical atoms, and also if the atoms do not interact. The latter occurs if the level difference  $\omega_{1A} - \omega_{1B}$  is large (see below), and also if the distance between atoms is large ( $qR \gg 1$ ) so that the interaction vanishes ( $U_{AB} = 0$ ), and therefore the general solution takes a simple form corresponding to independent behavior of the atoms:

$$b_{1A} = \exp\{-i\omega_{1A}t - \gamma_A t\}, \quad b_{1B} = 0.$$

Let us now examine the behavior of the system under certain concrete assumptions regarding its parameters.

1) Interaction of atoms with non-overlapping levels at small distances ensuring a strong interaction of the atoms,  $\omega_{1A} = \omega_{1B} = \omega_1$ ,  $|\operatorname{Re} U_{AB}| > \hbar(\gamma_A + \gamma_B)$ .

Using expansions (9') and (10'), we can easily show that the condition  $|\operatorname{Re} U_{AB}| > \hbar(\gamma_A + \gamma_B)$  in the case of dipole-dipole interaction of the atoms is equivalent to the condition

$$(qR)^6 < \gamma_A \gamma_B / (\gamma_A + \gamma_B)^2,$$

and in the case of quadrupole-dipole interaction is equivalent to the condition

$$(qR)^8 < \gamma_A / \gamma_B,$$

where  $\gamma_B$  is the constant of the allowed decay. From (9') and (10') it follows also that  $|\operatorname{Re} U_{AB}| > \operatorname{Im} U_{AB}$ . If these inequalities are satisfied, the solution can be written in the form

$$b_{1A} = \frac{1}{2} \exp\left\{-i\omega_1 t - \frac{(\gamma_A + \gamma_B)t}{2}\right\} \left[ \left(1 + i \frac{\gamma_A - \gamma_B}{2U_{AB}} \hbar\right) \exp\left\{i \frac{U_{AB}t}{\hbar}\right\} + \left(1 - i \frac{\gamma_A - \gamma_B}{2U_{AB}} \hbar\right) \exp\left\{-i \frac{U_{AB}t}{\hbar}\right\} \right], \\ b_{1B} = \frac{1}{2} \exp\left\{-i\omega_1 t - \frac{(\gamma_A + \gamma_B)t}{2}\right\} \left[ \exp\left\{-i \frac{U_{AB}t}{\hbar}\right\} - \exp\left\{i \frac{U_{AB}t}{\hbar}\right\} \right].$$

If the transitions become allowed for both atoms, then the damping constants of the different components at  $qR \ll 1$  are equal to

$$\gamma_A \pm 2|\operatorname{Im} U_{AB}| + \gamma_B \approx (\gamma_A \pm \gamma_B)^2,$$

and this can be interpreted as a result of interference between the radiations from the individual atoms. The coherence of the states of the atoms is the result of the interaction of the atoms via the electromagnetic field.

If the multipolarities of the transitions in atoms A and B do not coincide, no interference of the radiation takes place. In particular, for the dipole-quadrupole interaction we have according to (10')  $\operatorname{Im} U_{AB} = 0$ . The entire system decays with a total constant  $\gamma_A + \gamma_B$ , re-

gardless of which of the atoms was initially excited.

A similar situation also obtains if the difference between the decay constants of the like allowed transitions is large, for in this case  $|\operatorname{Im} U_{AB}| \approx \hbar \sqrt{\gamma_A \gamma_B}$   $qR \rightarrow 0$

$\ll \hbar(\gamma_A + \gamma_B)$ . The system then actually decays with the larger of the constants  $\gamma_A$  or  $\gamma_B$ , even if the weakly-decaying atom was initially excited. No metastable states are produced in this case.

It follows from the obtained solution that the probability of radiation dragging  $|b_{1A}|^2 + |b_{1B}|^2$  oscillates with a very small amplitude ( $\sim \operatorname{Im} U_{AB} / \operatorname{Re} U_{AB}$ ), but the probabilities for the excited state for the individual atoms with strongly differing radiative-decay constants contain only the oscillating terms

$$|b_{1A}|^2 = \exp\{-(\gamma_A + \gamma_B)t\} \cos^2(\operatorname{Re} U_{AB}t / \hbar), \\ |b_{1B}|^2 = \exp\{-(\gamma_A + \gamma_B)t\} \sin^2(\operatorname{Re} U_{AB}t / \hbar).$$

A direct electromagnetic interaction between the atoms leads to a periodic energy transfer from one atom to the other and back. The frequency of the oscillations of the excitations is determined by the interaction energy  $\operatorname{Re} U_{AB}$ , and the depth of the oscillations is maximal at  $\omega_{1A} = \omega_{1B}$ . This corresponds fully to interaction between a system with two states (levels) and a resonant perturbation (see<sup>[10]</sup>).

In the case of intense excitation exchange between the atoms, the system decays with the summary decay constant. This effect does not depend on the interference between the radiations from the atoms, an interference that depends, as already indicated, on  $\operatorname{Im} U_{AB}$  and vanishes in the case of transitions of different multipolarity (when  $qR \ll 1$ ).

2) Interaction of identical atoms  $\omega_{1A} = \omega_{1B} = \omega_1$ ,  $\gamma_A = \gamma_B = \gamma$ .

The solution for the expansion coefficients takes a form analogous to the result obtained in<sup>[5]</sup>:

$$b_{1A} = \exp\{-i(\omega_1 + \gamma)t\} [\exp\{iU_{AB}t/\hbar\} + \exp\{-iU_{AB}t/\hbar\}], \\ b_{1B} = \exp\{-i(\omega_1 + \gamma)t\} [\exp\{iU_{AB}t/\hbar\} - \exp\{-iU_{AB}t/\hbar\}].$$

When  $qR \ll 1$ , we can use the approximate relation (9'). The states of the system are either stable or decaying with twice the rate  $|b_{1A}|^2 + |b_{1B}|^2 = (e^{-4\gamma t} + 1)/2$ .

There are no radiation oscillations. If  $|\operatorname{Re} U_{AB}| > \gamma$ , oscillations of the excited state of each of the atoms set in:

$$|b_{1A}|^2 = 1/4 [e^{-4\gamma t} + 2e^{-2\gamma t} \cos(\operatorname{Re} U_{AB}t/\hbar) + 1].$$

The amplitude of the oscillations at the initial instant of time is equal to unity, and the excitation is completely transferred from one atom to the other and back at a frequency  $\operatorname{Re} U_{AB}/\hbar$ .

If we neglect the Coulomb interaction of the atoms, then  $\operatorname{Re} U_{AB} \rightarrow 0$  and the oscillations disappear, but the interference effect in the radiation still remains, owing to the interaction of the atoms via the transverse field. This is precisely the case corresponding to the conditions of the Dicke problem<sup>[4]</sup>. We note incidentally that a similar formulation of the problem for unlike atoms ( $\operatorname{Re} U_{AB} = 0$ ,  $\operatorname{Im} U_{AB} \neq 0$ ) leads to the presence of metastable states with a statistical weight  $\gamma_{\min}^2 / (\gamma_A + \gamma_B)^2$ , where  $\gamma_{\min}$  is the smaller of the

two quantities  $\gamma_A$  and  $\gamma_B$ . Thus, neglect of  $\text{Re } U_{AB}$  in the case of unlike atoms is not permissible, in view of the strong influence of the oscillations of the excitations on the radiation process.

When  $qR > 1$ , the interaction of like atoms decreases, and the stable states vanish with increasing distance (see<sup>[6]</sup>).

3) Interaction of atoms with non-overlapping levels,  $|\omega_{1A} - \omega_{1B}| > (\gamma_A + \gamma_B)$ .

The behavior of the system is determined in the general case by the solution (11)–(12). With increasing difference between the levels  $\omega_{1A}$  and  $\omega_{1B}$ , the interference effect in the radiation decreases ( $\text{Re } \beta$  decreases). At the same time, the frequency of exchange of excitations between the atoms (which is determined by  $\text{Im } \beta$ ) increases. But the increase of the frequency of modulation of  $|b_{1A}|^2$  and  $|b_{1B}|^2$  is accompanied by a decrease of the depth of modulation. It is easily seen, for example, that  $|E|^2$  decreases with increasing  $|\beta|$  (these results agree fully with the solution of the corresponding problem in the book of Landau and Lifshitz<sup>[10]</sup>), if the frequency of the external field is set equal to zero in its conditions).

Under the condition  $\hbar|\omega_{1A} - \omega_{1B}| \gg |\text{Re } U_{AB}|$ , we can show (using also the inequality  $\hbar|\gamma_A - \gamma_B| > |\text{Im } U_{AB}|$ ) that the approximate equality  $\beta \approx \gamma_A - \gamma_B + i(\omega_{1A} - \omega_{1B})$  holds. As expected, the atoms behave independently if the level difference is large.

If  $\hbar|\omega_{1A} - \omega_{1B}| < |\text{Re } U_{AB}|$ , then approximately  $\beta \approx 2iU_{AB}$ , just as in the case of coinciding levels. Thus, the results obtained for overlapping levels remain valid also in the case of non-overlapping levels, provided the level splitting due to the electromagnetic interaction of the atoms exceeds the difference between the unperturbed levels of the atoms.

### 3. DISCUSSION

The calculation shows that coherent effects should appear in the spontaneous emission in a system of two-level atoms interacting via the electromagnetic field of the radiation even in the general case of unlike oscillators. In the limiting case when the resonant frequencies and the radiation widths of the levels of the atoms become identical, the general solution takes a form coinciding with the results of<sup>[1,6]</sup>, in which the condition that the properties of the atoms are identical was used. In accordance with the point of view advanced in<sup>[11]</sup>, we can state that there is no fundamental difference between the behavior of identical and different atoms. The condition that the atoms be identical is not obligatory for coherence effects.

A distinguishing feature of a system of unlike atoms is the absence of metastable states. Moreover, if the distance between the atoms is sufficiently small, then the decay of the entire system has a constant equal to the arithmetic mean of the radiative widths of the individual atoms. If at least one of the atoms has an allowed transition at a given frequency, then the entire system will decay with the probability of the allowed transition. This is due to the fact that at a small distance the atoms exchange excitations intensively (the levels of the system as a whole are split in this case). For such virtual excitation-transfer processes it is not

necessary to have strict resonance, i.e., overlap of the atomic levels. It suffices only to have the splitting of the levels exceed the difference between the atomic levels themselves. Thus, under certain conditions, non-resonant transfer of excitation (with subsequent emission of an electromagnetic quantum) is possible without participation of phonons or excitons.

A certain parallel can be drawn between our results and the data of Rautian and Sobel'man<sup>[12]</sup>, who considered the spontaneous decay from two levels in the presence of a strong resonant field. The role of such an external field is played in our case by the interaction between the atoms, and the equality of the emission frequencies of the atoms leads to additional coherence effects. The joint motion of the atoms was disregarded in the calculations. It is possible, however, to use the results also in the case of moving atoms. If this motion does not lead to an essential change of  $R$ , the Doppler frequency shifts can be readily taken into account by introducing the corresponding frequency difference  $\omega_{1A} - \omega_{1B}$  with subsequent averaging of the result over the Doppler width.

In principle it is possible to extend the results also to the case of decay of the atoms of the impurity centers in a solid, when the interaction with the phonon field leads to an appreciable broadening of the level compared with the radiative width and to a decrease of the lifetime  $\tau$  at the corresponding sublevels. If the coefficients  $b_{1A}$  and  $b_{1B}$  pertain to such states, then the widths  $2\gamma_A$  and  $2\gamma_B$  should be replaced by the corresponding values  $\tau_{1A}^{-1}$  and  $\tau_{1B}^{-1}$ . This follows directly from the fact that  $\gamma_A$  and  $\gamma_B$  determine the lifetimes of the atoms interacting with the surrounding field. Since we are considering here a purely electromagnetic interaction of the atoms, the expression for  $U_{AB}$  remains unchanged. The presence of a dense medium should also be taken into account by introducing a corresponding dielectric constant (it is assumed that there are no absorption bands near the impurity levels under consideration).

In the Forster-Dexter sensitized-luminescence theory<sup>[13]</sup>, the process of resonant energy transfer between impurity centers is described in first order of perturbation theory, and only the Coulomb interaction ( $V_{AB}$ ) is taken into account. This approach is justified if the interaction of the atoms is relatively weak:  $\text{Re } U_{AB} < (\tau_{1A}^{-1} + \tau_{1B}^{-1})$ , and the interaction via the transverse field is smaller than the Coulomb interaction:  $|\text{Im } U_{AB}| < |\text{Re } U_{AB}|$ , just as in the case of interaction between different multipoles. In the general case, however, the interaction of the oscillators is determined by the matrix element  $U_{AB}$  and has a more complicated character, owing to the possible coherence effects.

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