

Collective Radiative Processes Involving Unlike Atoms

A. A. VARFOLOMEEV

I. V. Kurchatov Atomic Energy Institute

Submitted June 24, 1971

Zh. Eksp. Teor. Fiz. 62, 111-118 (January, 1972)

Two-photon emission processes in a system of two unlike atoms having close excitation energy levels and located at a fixed distance from each other are considered within the framework of the perturbation theory for decaying states. Solutions are obtained which describe the behavior of the system during resonance fluorescence and spontaneous decay from an initial state with two excited atoms. It is shown that resonant interaction between unlike atoms involving excitation exchange leads to complete radiative decay of the system (in contrast to a system of identical atoms) and to the appearance of interference effects which considerably alter the time law of decay and the radiation energy spectra.

INTRODUCTION

THE process of radiative damping in an ensemble of identical two-level radiators was considered by many authors. In most papers it was assumed, for the sake of simplicity, that the particle interaction is effected only via the common transverse radiation field^[1-4]. The total (including the Coulomb) resonant interaction was taken into account only for the simplest systems consisting of two atoms^[5]. It was assumed that the atoms are located at fixed distances from each other and that one of the atoms is excited at the initial instant of time. An analogous problem for two unlike atoms with close excitation levels was considered in^[6]. It was shown that coherent effects become manifest in the case of unlike atoms. The Coulomb interaction leads in this case to certain specific features (the absence of excited metastable states, radiation beats). The present paper contains an analysis of two-photon radiation processes in a system of unlike atoms, namely collective spontaneous decay from the initial state with two excited atoms, and resonant fluorescence of two atoms.

COLLECTIVE SPONTANEOUS DECAY

We consider radiative damping of a system of two unlike atoms excited at the initial instant, under the following conditions.

The excitation levels of the atoms A and B, equal respectively to ω_{01} and ω_{02} (we use a system of units with $\hbar = c = 1$) are close or coincide. For simplicity we assume that the levels are nondegenerate. The distance R between atoms is assumed fixed and exceeding the dimensions of the atoms, so that the overlap of the wave functions of electrons of different atoms can be neglected. The motion of the atoms as a whole and the recoil energy of the nuclei are not taken into account. System states with more than two quanta are not considered, as a result of which nonresonant multiquantum transitions are excluded. The following states of the system are assumed possible:

$|12\rangle$ — both atoms in the excited state;

$|1\lambda\rangle$ ($|2\lambda\rangle$) — atom A (atom B) is excited, and the radiation field contains one quantum $\lambda(\mathbf{q}, \mu)$ with wave vector \mathbf{q} and polarization $\mathbf{e}_{\mathbf{q}\mu}$;

$|\lambda\lambda'\rangle$ — both atoms in the ground state, the field corresponds to two quanta λ and λ' .

We represent the interaction Hamiltonian in the form (Coulomb gauge)

$$\mathcal{H}_{int} = H + V + \mathcal{V}, \tag{1}$$

where the operator \mathcal{V} is determined by the Coulomb interaction of the atoms, and the operators H and V describe the interaction of the atoms A and B, respectively, with the transverse field. Using the perturbation theory of Heitler and Ma^[7], which makes it possible to take the damping of the states into account, we introduce the matrix U(E), which defines the Fourier components of the system state amplitudes:

$$b_{12}(t) = \frac{i}{2\pi} \int_{-\infty}^{\infty} dE G(E) \exp\{i(E_0 - E)t\},$$

$$b_{jk}(t) = \frac{i}{2\pi} \int_{-\infty}^{\infty} dE U_{jk}(E) \zeta(E - E_{jk}) G(E) \exp\{i(E_{jk} - E)t\}, \tag{2}$$

where $G(E) = [E - E_0 + i\Gamma(E)]^{-1}$, E_0 and E_{jk} are the values of the system energy in the initial state and in the intermediate or final states $|jk\rangle$, respectively, $\zeta(E - E_{jk})$ is the Dirac zeta function, and $\Gamma(E)$ is the damping function of the initial state. Introducing the matrix elements of the operators (1), we write down a system defining the matrix U(E) and the function $\Gamma(E)$:

$$U_{12}(E) = V_{01} + \sum_{\lambda} H_{10}^{\lambda} U_{1\lambda}(E) \zeta(E - E_{1\lambda}) + \mathcal{V}_{AB} U_{2\lambda}(E) \zeta(E - E_{2\lambda}),$$

$$U_{2\lambda}(E) = H_{01}^{\lambda} + \sum_{\lambda'} V_{10}^{\lambda'} U_{1\lambda'}(E) \zeta(E - E_{1\lambda'}) + \mathcal{V}_{BA} U_{1\lambda}(E) \zeta(E - E_{1\lambda}), \tag{3}$$

$$U_{1\lambda'}(E) = H_{01}^{\lambda'} U_{1\lambda}(E) \zeta(E - E_{1\lambda}) + V_{01}^{\lambda'} U_{2\lambda}(E) \zeta(E - E_{2\lambda})$$

$$+ H_{01}^{\lambda'} U_{1\lambda'}(E) \zeta(E - E_{1\lambda'}) + V_{01}^{\lambda'} U_{2\lambda'}(E) \zeta(E - E_{2\lambda'});$$

$$-i\Gamma(E) = \sum_{\lambda} H_{10}^{\lambda} U_{2\lambda}(E) \zeta(E - E_{2\lambda}) + \sum_{\lambda} V_{10}^{\lambda} U_{1\lambda}(E) \zeta(E - E_{1\lambda}). \tag{4}$$

It can be shown that the solution of the system (3) takes the form

$$U_{1\lambda}(E) \zeta(E - E_{1\lambda}) = \frac{(E - E_{2\lambda} + i\gamma_B) V_{01}^{\lambda} + \mathcal{U}_{AB} H_{01}^{\lambda}}{(E - E_{1\lambda} + i\gamma_A)(E - E_{2\lambda} + i\gamma_B) - \mathcal{U}_{AB} \mathcal{U}_{BA}}$$

$$U_{2\lambda}(E) \zeta(E - E_{2\lambda}) = \frac{(E - E_{1\lambda} + i\gamma_A) H_{01}^{\lambda} + \mathcal{U}_{AB} V_{01}^{\lambda}}{(E - E_{2\lambda} + i\gamma_B)(E - E_{1\lambda} + i\gamma_A) - \mathcal{U}_{AB} \mathcal{U}_{BA}} \tag{5}$$

We have used here the notation

$$\sum_{\lambda} |H_{01}^{\lambda}|^2 \zeta(E - E_{\beta}) = -i\gamma_{\lambda}(E), \quad \sum_{\lambda} |V_{01}^{\lambda}|^2 \zeta(E - E_{\lambda}) = -i\gamma_{\beta}(E),$$

$$\sum_{\nu} V_{10}^{\nu} H_{01}^{\nu} \zeta(E - E_{\lambda\nu}) = \mathcal{U}_{\beta A}(E - \omega_{\lambda}) - \mathcal{V}_{\beta A}(E - \omega_{\lambda}). \quad (6)$$

By definition, $2\text{Re}\gamma_A$ is equal to the decay probability of the excited atom A. Accordingly, $2\text{Re}\gamma_B$ is equal to the radiative width of the level of the atom B. The quantities $\text{Im}\gamma_A$ and $\text{Im}\gamma_B$ are connected with the radiative shifts of the levels of the atoms and can be neglected in comparison with the level width of the allowed transitions (see^[7]), putting $\text{Re}\gamma_A(\omega_{01}) = \gamma_A$ and $\text{Re}\gamma_B(\omega_{02}) = \gamma_B$.

According to (6), γ_A and γ_B are functions of the energy E. Their arguments, however, can vary within insignificant limits (on the order of the level widths). Therefore γ_A and γ_B enter in (5) as constants. Similar reasoning holds for the function \mathcal{U}_{AB} of the resonant interaction of the atoms with exchange of excitation and for the Coulomb interaction function \mathcal{V}_{AB} . (The explicit form of \mathcal{U}_{AB} for dipole-dipole and dipole-quadrupole interactions in the nonrelativistic approximation is given in^[6], see formulas (8)–(10). For the general form see^[8].) From the definition of the functions it follows that $\mathcal{U}_{AB} = \mathcal{U}_{BA}$ and $\mathcal{V}_{AB} = \mathcal{V}_{BA}$.

The solution (5) makes it easy to obtain the decay probability of the initial state of the system (4). The integration with respect to the frequencies reduces to a sum of the residues of the functions (5). Introducing, in analogy with^[2,7], a cutoff limit for the spectrum of the virtual quanta, we can show that, accurate to interactions of the order of $(\gamma_A + \gamma_B)/(\omega_{01} + \omega_{02})$, the following result holds true:

$$\Gamma(E_0) = \gamma_A(E_0) + \gamma_B(E_0). \quad (7)$$

Just as in the case of identical atoms, the decay probability of the system is equal to the sum of the decay probabilities of the individual atoms. The presence of a resonant interaction between the atoms in the intermediate state (one of the atoms is excited) does not influence the decay probability of the doubly-excited state.

When the amplitude of the state with one excited atom is determined with the aid of formulas (2) and (5), the integrals reduce to the sum of the residues at the poles $E_{1,2} = \omega_{\lambda} + [E_0 - i\Gamma(E_0)]/2 \pm [(\omega_{01} - \omega_{02} - i\gamma_A + i\gamma_B)^2/4 + \mathcal{U}_{AB}\mathcal{U}_{BA}]^{1/2}$, and $E_3 = E_0 - i\Gamma(E_0)$. In the general case, the expression for the square of the modulus of the amplitude of the state will contain terms with different exponentials, and also interference terms, some of which may lead to radiation beats (see^[6,9]). Let us note some particular cases that follow from the general solution.

Putting $\mathcal{U}_{AB} = -i(\gamma_A\gamma_B)^{1/3}$ (see^[6]) we obtain a solution for a system of two atoms with overlapping levels, separated by a small distance, without allowance for their Coulomb interaction. For sufficiently large time intervals ($t \gg \omega_{02}^{-1}$) the total probability of the atom B remaining excited at the instant of time t is given by

$$\sum_{\lambda} |b_{2\lambda}|^2 = \gamma_A(\gamma_B - \gamma_A)^2 \Gamma^{-3} (1 - e^{-\Gamma t})^2 + 4\gamma_A\gamma_B \Gamma^{-2} e^{-2\Gamma t} \left[2\gamma_B t + \frac{\gamma_A - \gamma_B}{\Gamma} (1 - e^{-\Gamma t}) \right]. \quad (8)$$

We see that the interference of the radiation from the different states of the system greatly complicates the time dependence of the decay of the system. Attention is called to the presence of a stable component in the case of unlike atoms (dragging of the radiation). This result, however, is a consequence of neglecting the contribution of the Coulomb term to the resonant interaction between the atoms, and is not valid in the general case. This can easily be verified by noting that when account is taken of the total resonant interaction ($\text{Re}\mathcal{U}_{AB} \neq 0$) the condition $|\text{Im}(E_1 - E_2)| = \Gamma$, which leads in the foregoing case to cancellation of the real parts in the arguments of the exponentials, is not satisfied.

In the limiting case of strongly interacting atoms ($\text{Re}\mathcal{U}_{AB} \gg \Gamma$) with overlapping levels ($\omega_{01} \approx \omega_{02}$), the probability of the excited state of one of the atoms changes with time like

$$\sum_{\lambda} |b_{2\lambda}|^2 = e^{-\Gamma t} \text{ch}(2 \text{Im}\mathcal{U}_{AB} t) - e^{-2\Gamma t}. \quad (9)$$

For identical atoms ($\gamma_A = \gamma_B$) the results that follow from (8) and (9) coincide. Allowance for the strong Coulomb interaction does not lead to a change in the decay probabilities of a system of identical atoms. In the case of unlike atoms, allowance for the total resonant interaction is necessary in principle, since it leads to a qualitatively different solution (to a complete decay of the system without dragging of the radiation). This result can easily be explained. The initial state of the identical atoms is symmetrical with respect to permutation of the atoms, and the decay proceeds to its conclusion to a symmetrical ground state. In the case of unlike atoms, there is no such symmetry, and if the interaction is effected only through the transverse radiation field, then the decay is not necessarily possible from all the states of the system. However, complete resonant interaction of the atoms can lead to exchange of excitations between them^[6], thereby shifting the states of the system as a whole. As a result, the system undergoes radiative decay regardless of the form of the initial state.

Using the corresponding expressions, we can obtain from (2), (5), and (3) the limiting value of the amplitude of the final state of the system

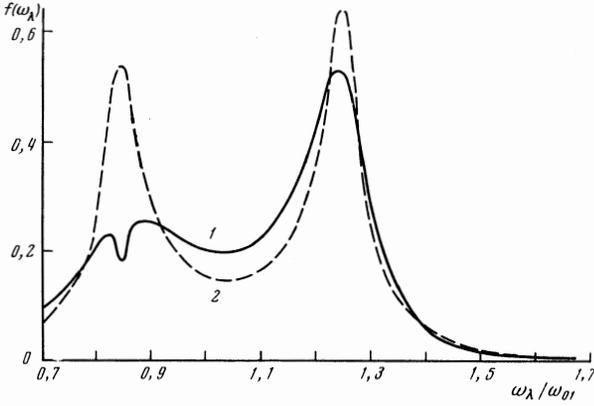
$$b_{\lambda\nu}(\infty) = \beta(\lambda, \lambda') + \beta(\lambda', \lambda);$$

$$\beta(\lambda, \lambda') = \frac{H_{01}^{\nu} V_{01}^{\lambda} \Delta_2' + V_{01}^{\nu} H_{01}^{\lambda} \Delta_1' + V_{01}^{\lambda} V_{01}^{\nu} \mathcal{U}_{BA} + H_{01}^{\lambda} H_{01}^{\nu} \mathcal{U}_{AB}}{(\Delta_1 + \Delta_2')(\Delta_1' \Delta_2' - \mathcal{U}_{AB} \mathcal{U}_{BA})} \quad (10)$$

where

$$\Delta_{1,2} = \omega_{\lambda} - \omega_{01,2} + i\gamma_{A,B}; \quad \Delta_{1,2}' = \omega_{\lambda'} - \omega_{01,2} + i\gamma_{A,B}.$$

The form of the spectrum of the collective spontaneous radiation is determined by the square of the modulus of the amplitude (10), summed over the polarizations and emission angles of the quanta and over the frequency of one of the quanta. The integration with respect to frequency reduces to the sum of the residues at the poles $\omega_k = E_k - \omega_{\lambda}$ ($k = 1, 2, 3$), and the summation over the polarizations and angles is taken into account, as above, by introducing the widths γ_A , γ_B , and $\text{Im}\mathcal{U}_{AB}$. From the structure of (10) we see that besides the emission lines of the individual atoms there are also different interference terms that complicate the form of the



Emission spectra of interacting unlike atoms: $1 - |\text{Im } \mathcal{U}_{AB}| = 0.7 \gamma_A$, $2 - |\text{Im } \mathcal{U}_{AB}| = 0.7 \gamma_A$; $\gamma_A = 0.05 \omega_{01}$, $\gamma_B = 0.5 \gamma_A$, $\omega_{02} = 1.1 \omega_{01}$, $\text{Re } \mathcal{U}_{AB} = 4 \gamma_A$.

emission spectrum of the system. The resultant expressions for the form of the spectrum are cumbersome.

The figure shows by way of illustration the forms of the emission spectrum of interacting atoms with close but not identical levels at certain concrete parameters of the system. For comparison, the figure also shows the dispersion curves of the emission of the isolated atoms. From the figure and from (9) and (10) we see that the emission-line shape changes significantly when account is taken of the resonant interaction both as a result of the level shift (determined by the value of $\text{Re } \mathcal{U}_{AB}$) and as a result of the radiation interference due to the fact that the intermediate level of the system has a sublevel structure. The radiative damping of the atoms has a collective character.

RESONANT FLUORESCENCE OF A TWO-ATOM CENTER

Using the procedure described above, let us show that collective effects can also take place in resonant fluorescence. For simplicity, the atomic levels are assumed to be nondegenerate, and the external scattered field is assumed weak compared with the saturating field, so that the probability of the simultaneous absorption of two quanta from the primary beam can be neglected. We regard the following states of the system as possible:

$|00\rangle$ —ground state of the system prior to the interaction; $|\lambda\rangle$, $|\lambda'\rangle$ —the atom A or the atom B is excited by absorption of a quantum λ ;

$|\lambda\lambda'\rangle$ —both atoms in the ground state after the absorption of the quantum λ and emission of the quantum λ' .

The system equation for the matrix U takes in this case the form

$$U_{20}^{\lambda}(E) = V_{10}^{\lambda} + \mathcal{Y}_{nA} U_{10}^{\lambda}(E) \zeta(E - E_1^{\lambda}) + \sum_{\lambda'} V_{10}^{\lambda'} U_{10}^{\lambda'}(E) \zeta(E - E_{\lambda'}^{\lambda}),$$

$$U_{10}^{\lambda}(E) = H_{10}^{\lambda} + \mathcal{Y}_{AB} U_{20}^{\lambda}(E) \zeta(E - E_2^{\lambda}) + \sum_{\lambda'} H_{10}^{\lambda'} U_{10}^{\lambda'}(E) \zeta(E - E_{\lambda'}^{\lambda}),$$

$$(11)$$

$$U_{10}^{\lambda}(E) = V_{01}^{\lambda} U_{20}^{\lambda}(E) \zeta(E - E_2^{\lambda}) + H_{01}^{\lambda} U_{10}^{\lambda}(E) \zeta(E - E_1^{\lambda});$$

$$-i\Gamma(E) = \sum_{\lambda} H_{01}^{\lambda} U_{10}^{\lambda}(E) \zeta(E - E_1^{\lambda}) + \sum_{\lambda} V_{01}^{\lambda} U_{20}^{\lambda}(E) \zeta(E - E_2^{\lambda}),$$

$$(12)$$

where

$$E_{\lambda}^{\lambda} = E_0 + \omega_{\lambda} - \omega_{\lambda}; \quad E_k^{\lambda} = E_0 + \omega_{0k} - \omega_{\lambda}, \quad k = 1, 2;$$

E_0 is the energy of the initial state of the system. The summation over the quanta λ should be carried out with allowance for the parameters of the primary quantum beam.

From the mathematical point of view, the system (11) is analogous to (3), so that its solution is similar to (5). Using the solution of the system (11), we can determine the necessary characteristics of the resonant-fluorescence process. Omitting the intermediate calculations, we present some results for two types of the energy spectrum of the primary beam, namely wide and narrow compared with the radiative widths of the atomic levels.

The total probability of absorbing the quantum ω_{λ} from a beam with a wide energy spectrum, determined from (12) by integrating with respect to the frequency of the primary-beam quanta, is

$$\Gamma(E_0) = \pi \omega_{\lambda}^{-1} I(\omega_{\lambda}) (|H_{01}|^2 + |V_{01}|^2), \quad (13)$$

where $I(\omega_{\lambda})$ is the intensity density of the primary beam in a unit frequency interval in the region of the resonant frequencies ω_{01} and ω_{02} . Here, as above, the radiative corrections are small and neglected, so that $\text{Im } \Gamma(E_0) = 0$. As seen from (13), the total width is equal to the sum of the probabilities of quantum absorption by each of the atoms separately. There are no interference effects in this case. The intensity of total absorption from an external beam with a broad energy spectrum is proportional to the number of active atoms, just as in the case of the transition between two levels of atoms in an external field without allowance for the direct interaction between the atoms (see^[2]).

We note that the assumed condition that the intensity of the external field be small compared with the saturating field is equivalent, as follows from (13), to the condition $\Gamma(E_0) \ll \gamma_A = \gamma_B$. Thus, the broadening of the initial state by the interaction with the external field is small compared with the radiative width of the excited state.

Absorption from a beam with a narrow energy spectrum can be formally described by putting $I(\omega_{\lambda}) = I_0 \delta(\omega - \omega_{\lambda})$, where I_0 is the total intensity of the beam. Integration with respect to the frequency is equivalent to exchange of variables in (12). The resultant absorption probability is

$$\text{Re } \Gamma(E_0) = I_0 \omega_{10}^{-1} \Phi(\omega_{10});$$

$$\Phi(\omega_{10}) = \frac{|H_{01}^{\lambda}|^2 \text{Im } \omega_{\alpha}^*}{(\omega_{10} - \text{Re } \omega_{\alpha})^2 + (\text{Im } \omega_{\alpha})^2} + \frac{|V_{01}^{\lambda}|^2 \text{Im } \omega_{\beta}^*}{(\omega_{10} - \text{Re } \omega_{\beta})^2 + (\text{Im } \omega_{\beta})^2}$$

$$- \text{Im} \frac{|H_{01}^{\lambda}|^2 (\omega_{\beta} - \omega_{02} + i\gamma_{\beta}) + |V_{01}^{\lambda}|^2 (\omega_{\alpha} - \omega_{01} + i\gamma_{\alpha}) + 2\mathcal{U}_{AB} \text{Re}(V_{01}^{\lambda} H_{01}^{\lambda})}{(\omega_{10} - \omega_{\alpha})(\omega_{10} - \omega_{\beta})}$$

$$(14)$$

where

$$\omega_{\alpha, \beta} = (\omega_{01} + \omega_{02}) / 2 - i(\gamma_A + \gamma_B) / 2$$

$$\pm [(\omega_{01} - \omega_{02} - i\gamma_A + i\gamma_B)^2 / 4 + \mathcal{U}_{AB} \mathcal{U}_{BA}]^{1/2}$$

are the poles of the solutions of the system (11) at an energy $E = E_0$.

Thus, in the general case, the expression for the total probability of absorption of quanta from a beam with a narrow spectrum contains two terms of the dis-

persion type, which are connected with the amplitudes for absorption by each of the atoms separately, and also a term due to the interference of different coherent sublevel states of the system. The occurrence of the interference terms can also be attributed to the fact that there are two channels of excitation for each of the atoms, namely direct interaction with the field and transfer of excitation from the neighboring atom after the latter has absorbed a quantum. Both amplitudes are coherent, so that interference terms should appear in the quantum-absorption reaction cross section. It is easy to ascertain that there is no interference in the case of independent atoms ($\mathcal{U}_{AB} = 0$) and in the case of identical atoms at small distances ($\text{Im } \mathcal{U}_{AB} = -\gamma$).

Direct interaction of the atoms leads to a shift of the maximum of the resonant absorption curve and to its broadening, but does not change the total absorption cross section (integrated over the frequency ω_{λ_0}). The maximum is shifted in only one direction, in full accord with the results of the investigation of the decay of a system of identical atoms (see also^[6]). The collective level $\omega_{01} + \text{Re } \mathcal{U}_{AB}$ corresponds to a symmetrical state, has a finite lifetime with respect to radiative decay, and is excited upon interaction with the external field. The level $\omega_{01} - \text{Re } \mathcal{U}_{AB}$ corresponds to an asymmetrical nondecaying state and accordingly is not excited upon interaction with a resonant field.

The emission line shape and the absorption spectrum can be determined from the limiting value of the state amplitude as $t \rightarrow \infty$:

$$b_{\lambda^{\lambda}}(\infty) = U_{\lambda^{\lambda}}(E) [E_{\lambda^{\lambda}} - E_0 + i\Gamma(E_{\lambda^{\lambda}})]^{-1}.$$

The absorption line shape is determined by the sum $\sum_{\lambda} |b_{\lambda^{\lambda}}|^2$ over the quanta of the free-radiation field. The dispersion factor connects, in δ -fashion, the energies of the absorbed and emitted quanta. Just as in resonant fluorescence by a single atom, the energy is conserved within the limits of one act of absorption and emission (accurate to within the width Γ). On the whole, the probability of the resonant-fluorescence process depends on the energy of the primary quantum. It can be shown that this dependence, which determines the line shape for absorption from a beam with a broad energy spectrum, is given by

$$\sum_{\lambda} |b_{\lambda^{\lambda}}(\infty)|^2 = \Phi(\omega_{\lambda})/\Gamma(E_0). \quad (15)$$

Here $\Gamma(E_0)$ is defined in accordance with (13), and $\Phi(\omega_{\lambda})$ is analogous to the function (14). Just as in the case of resonant fluorescence by a single atom (see^[7]), the absorption spectrum coincides in shape with the dependence of the cross section for the absorption of a monochromatic beam on the quantum energy.

The emission line shape can be obtained by summing over the quanta of the primary beam $\sum_{\lambda} |b_{\lambda^{\lambda}}|^2$. If in addition we average over the directions of the wave vectors of the quanta and their polarizations, then the obtained emission line shape coincides in form with the absorption line. The agreement with formula (15) is complete if $\Gamma(E_0)$ stands for the width (7).

An analysis of the absorption and emission spectra in the case of a quasimonochromatic beam ($\Delta\omega_{\lambda} \ll \gamma_A + \gamma_B$) leads to the same results as in the case of ordin-

ary resonant fluorescence. The emission spectrum is determined by the shape of the primary-beam spectrum or has a dispersion shape with width $2\text{Re}\Gamma$, if the latter exceeds the frequency scatter in the beam.

CONCLUSION

Thus, the considered emission processes in a system of atoms separated by distances comparable with the emission wavelength have a collective character. In a system of unlike atoms with close energy levels, exchange of excitations leads to a total decay of the system with the probability equal to the sum of the radiative decay probabilities of each of the atoms. Spontaneous decay takes place from each initial state (either one^[6] or both atoms excited), and the decay proceeds to conclusion without formation of metastable states. An exception is a system of identical atoms separated by short distances ($R < \omega_{01}^{-1}$). In this case allowance for the total resonant interaction does not change the main results obtained without allowance for the Coulomb interaction of the atoms at short distances^[1-5].

Both in spontaneous decay and in resonant fluorescence, the intermediate states of the system constitute a mixture of coherent sublevel states; this leads to interference effects in emission (see^[9]). These effects are revealed by a nonexponential decay law (the presence of several exponentials, beats of radiation), and by a more complicated emission spectrum as compared with the dispersion form of the emission spectrum of noninteracting atoms.

In conclusion, we note that our results, in analogy with the results of^[1-6], can pertain to systems of arbitrary radiators (atoms, molecules, nuclei). Real systems, in which the considered effects can, in principle, appear are complex molecules, activated crystals, and under certain conditions also a mixture of rarefied gases with close excitation levels. The condition that the system of two radiators be isolated was introduced to simplify the problem and is not a fundamental one, since the Coulomb interaction, which leads to the exchange of excitations, is significant only at distances on the order of the emission wavelength. In addition, resonant interaction with exchange of excitations is also possible between groups of different atoms (molecules) and makes the emission process collective.

¹R. H. Dicke, Phys. Rev. **93**, 99 (1954).

²V. M. Faïn and Ya. I. Khanin, Kvantovaya radiofizika (Quantum Radiophysics), Sov. Radio, 1965.

³A. N. Oraevskii, Usp. Fiz. Nauk **91**, 181 (1967) [Sov. Phys. Usp. **10**, 45 (1967)].

⁴V. A. Alekseev, A. V. Vinogradov, and I. I. Sobel'man, Usp. Fiz. Nauk **102**, 43 (1970) [Sov. Phys. Usp. **13**, 576 (1971)].

⁵M. J. Stephen, J. Chem. Phys. **40**, 669 (1964); D. A. Hutchinson and H. F. Hamerka, J. Chem. Phys. **41**, 2006 (1964); Yu. A. Vdovin and V. M. Ermachenko, in Voprosy teorii atomnykh stolknovenii (Problems in the Theory of Atomic Collisions) Atomizdat, 1970, p. 111.

⁶A. A. Varfolomeev, Zh. Eksp. Teor. Fiz. **59**, 1702 (1970) [Sov. Phys. JETP **32**, 926 (1971)].

⁷W. Heitler, The Quantum Theory of Radiation, Oxford, 1954.

⁸A. I. Akhiezer and V. B. Berestetskii, Kvantovaya elektrodinamika (Quantum Electrodynamics), Nauka, 1969.

⁹L. N. Novikov, V. G. Pokazan'ev and G. V. Skrotskii, Usp. Fiz. Nauk **101**, 273 (1970) [Sov. Phys. Usp. **13**, 384 (1970)].