

RADIATIVE TRANSITIONS IN A RESONANT SYSTEM OF TWO-LEVEL MOLECULES

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It is shown that one of the reasons that a resonant system of two-level molecules cannot be described by ordinary perturbation theory is the appearance of multi-photon radiative transitions. The first terms of the corresponding perturbation theory series are calculated and the limit of applicability of the usual formula for the probability of "single-photon" radiation of molecules in the system is demonstrated.

ALEKSEEV, Vdovin and Galitskiĭ,^[1] calculated, with the help of an elegant method, the density fluctuations of photons in a resonant system of two-level molecules. In the calculation, the Hamiltonian was used in the form suggested by Dicke.^[2] It was shown that the radiation of the molecules cannot generally be described by the usual perturbation theory, since, in addition to the inapplicability of perturbation theory in the case of strong resonance fields, the "collective" effect of the interaction of the radiating molecules with one another through the radiation field also appears.

The effect can be described by means of a perturbation theory series in which each component gives a radiative transition with account of the interaction mentioned above.

A criterion will be derived below for the applicability of the ordinary formula for the probability of a "single-photon" transition in the system under consideration. The quantity obtained here coincides in the case of inversion in the system with the corresponding expression obtained in the cited reference^[1]

$$t \ll (\omega_0^2 V / 2\pi c^3 W |N_-^0|)^{1/2}, \tag{1}$$

where $|N_-^0|$ is the initial overpopulation and W is the probability of spontaneous dipole emission of an isolated molecule per unit time.

We shall compute only the first terms of this series, to make clear the qualitative features of the phenomena. Investigation of the entire series is not carried out in this work. We shall assume below that a radiative transition of frequency ω_0 takes place only between two nondegenerate states of the molecules. We shall assume the density of molecules to be so small that overlap of the electron wave functions can be neglected. Finally, the translational motion of the molecules is also neglected. If we neglect the damping of the system,

it is simplest to use for the calculation of the transition probabilities the time shift operator;^[3] here and below we shall denote this operator by the letter S , to distinguish it from the symbol for the T product, which is encountered below.

We shall write the (Schrödinger) Hamiltonian of the system in the form^[1,2]

$$H = \sum_j^n \frac{\hbar\omega_0}{2} \sigma_j^z + \sum_{\mathbf{k}, \lambda=1,2} \hbar\omega_{\mathbf{k}} c_{\mathbf{k}\lambda}^+ c_{\mathbf{k}\lambda} + g \left(\sum_{\mathbf{k}, \lambda=1,2} \left(\frac{1}{\omega_{\mathbf{k}} V} \right)^{1/2} c_{\mathbf{k}\lambda}^+ A_{\mathbf{k}\lambda} + \sum_{\mathbf{k}, \lambda=1,2} \left(\frac{1}{\omega_{\mathbf{k}} V} \right)^{1/2} c_{\mathbf{k}\lambda} A_{\mathbf{k}\lambda}^+ \right), \tag{2}$$

where $g = (2\pi\hbar)^{1/2} e/m$; the first two terms form the Hamiltonian H_0 of the unperturbed system of n molecules and of the free radiation field, and the last term is the interaction Hamiltonian H_{int} . The energies of the excited and ground states of the molecule are measured from the midpoint of the distance between the levels. The radiation field is represented in the form of a plane wave expansion which satisfies the usual periodic boundary conditions in the volume V . The operators $A_{\mathbf{k}\lambda}^+$ and $A_{\mathbf{k}\lambda}$ are described by the expressions

$$A_{\mathbf{k}\lambda}^+ = \sum_j^n (\mathbf{e}_{\mathbf{k}\lambda} \xi_{\mathbf{k}}^*) \exp(-i\mathbf{k}\mathbf{R}_j) \sigma_j^+,$$

$$A_{\mathbf{k}\lambda} = \sum_j^n (\mathbf{e}_{\mathbf{k}\lambda} \xi) \exp(i\mathbf{k}\mathbf{R}_j) \sigma_j^-;$$

$$\xi = \left(\varphi_b, \sum_i^z \exp(i\mathbf{k}\mathbf{r}_i) \mathbf{p}_i \varphi_a \right). \tag{3}$$

Here $\mathbf{e}_{\mathbf{k}\lambda}$ is the polarization vector, ξ is the transition matrix element for the isolated molecule (summation is over the electrons taking part in the transition), and \mathbf{R}_j is the coordinate of the center of mass of molecule j .

Equations (2) and (3) contain the operators σ_j^2 ,

σ_{γ}^{+} and σ_{γ}^{-} acting on the molecule j in the space of occupation numbers of n_{+} excited molecules and n_{-} molecules in the ground state.

The corresponding state of the molecular system can be described by a normalized function of the form

$$F(n_{+}, n_{-}) = \prod_{j'}^{n_{+}} \varphi_{j'}^{+} \prod_{j''}^{n_{-}} \varphi_{j''}^{-} \quad (4)$$

$$\equiv \varphi_{n_{+}}(\dots j' \dots) \varphi_{n_{-}}(\dots j'' \dots),$$

where $\varphi_{j'}^{+}$ and $\varphi_{j''}^{-}$ describe (in the occupation number space) the states of molecules j' and j'' . The functions (4), which correspond to the different energies of the molecular system, and which also differ in the permutations of molecules between the groups n_{+} and n_{-} for one and the same energy, form a system of orthogonal basis functions of the unperturbed Hamiltonian of the system of molecules.

The radiation field is described by wave functions satisfying the orthogonality and normalization conditions:

$$(\Phi(N_{k_1\lambda_1}, N_{k_2\lambda_2}, \dots), \Phi(N'_{k_1\lambda_1}, N'_{k_2\lambda_2}, \dots)) = \delta_{NN'} \delta_{k_1k_1'} \delta_{\lambda_1\lambda_1'} \dots \quad (5)$$

Thus, the functions

$$\Psi = F(n_{+}, n_{-}) \Phi(N_{k_1\lambda_1}, N_{k_2\lambda_2}, \dots) \quad (6)$$

form the basis system of eigenfunctions of the Hamiltonian H_0 of the unperturbed system.

The transitions induced in the system by the interaction H^{int} can be described in the interaction representation in the form of a perturbation-theory series for the operator [3]

$$S = \sum_m S^{(m)}, \quad (7)$$

which now, for real transitions with radiation (or absorption), in contrast to the case of a single isolated molecule, is not cut off at the term $S^{(1)}$, but has a finite number of terms which are uniquely determined by the number n_{+} of excited molecules (or n_{-} molecules lying in the ground state).

Transforming to the interaction representation, it is easy to obtain the following expression for the operator H^{int}

$$H^{\text{int}}(t) = g \sum_{\mathbf{k}, \lambda=1,2} \left(\frac{1}{\omega_{\mathbf{k}} V} \right)^{1/2} c_{\mathbf{k}\lambda}^{+} \sum_j^n (\mathbf{e}_{\mathbf{k}\lambda} \xi) \exp(i\mathbf{k}\mathbf{R}_j) \times \exp[i(\omega_{\mathbf{k}} - \omega_0)t] \sigma_j^{-} + g \sum_{\mathbf{k}, \lambda=1,2} \left(\frac{1}{\omega_{\mathbf{k}} V} \right)^{1/2} \times c_{\mathbf{k}\lambda} \sum_j^n (\mathbf{e}_{\mathbf{k}\lambda} \xi^{*}) \exp(-i\mathbf{k}\mathbf{R}_j) \exp[i(\omega_0 - \omega_{\mathbf{k}})t] \sigma_j^{+}. \quad (8)$$

Let us find the probability of emission of a single photon in the system. We consider the case in which there are no photons in the system at the initial moment, but in the final state, the molecule j' undergoes transition to the ground state with emission of a photon (\mathbf{k}, λ) . The initial and final wave functions of the system take the form

$$|i\rangle = F(n_{+}, n_{-}) \Phi_0,$$

$$|f\rangle = F(n_{+} - 1, n_{-} + 1) \Phi_1(\mathbf{k}, \lambda), \quad (9)$$

where Φ_0 is the amplitude of the vacuum state.

Taking (2) and (9) into account, we arrive at the following expression for the transition matrix element:

$$\langle f | S^{(1)} | i \rangle = -\frac{g}{i\hbar} \left(\frac{1}{\omega_{\mathbf{k}} V} \right)^{1/2} (\mathbf{e}_{\mathbf{k}\lambda} \xi) \exp(i\mathbf{k}\mathbf{R}_{j'}) F_{\mathbf{k}}(t), \quad (10)$$

where for brevity we have introduced the notation

$$F_{\mathbf{k}}(t) = \frac{\exp[i(\omega_{\mathbf{k}} - \omega_0)t] - 1}{i(\omega_{\mathbf{k}} - \omega_0)}. \quad (10a)$$

The square of the modulus of (10), averaged over the directions ξ and summed over all oscillators of the field (the usual transition is made from summation in \mathbf{k} to integration) gives the sought value

$$W_{j'}^{\gamma} = \frac{2}{3\pi} \frac{g^2 \bar{\xi}^2}{\hbar^2 c^3} \omega_0 t. \quad (11)$$

In the derivation of (11), the well known representation of the δ -function was used:

$$\delta(\omega) = \lim_{t \rightarrow \infty} \frac{1}{\pi} \frac{\sin^2 \omega t}{\omega^2 t}. \quad (11a)$$

Up to now, in introducing the phases $\exp(i\mathbf{k} \cdot \mathbf{R}_j)$, we have tacitly assumed that the wavelength of the radiation is smaller than the dimensions of the system. As will be seen below, the value of the wavelength begins to have a significant effect on the properties of the system only in the case of spontaneous emission, in which the order of the transitions is greater than unity.

Multiplying (11) by the number n_{+} of excited molecules, we get the probability of emission of the photon in the entire system:

$$W^{\gamma} = W_{j'}^{\gamma} n_{+} = \frac{4}{3} \frac{e^2 \bar{\xi}^2 \omega_0}{\hbar m^2 c^3} \frac{n_{+}}{V} V t. \quad (12)$$

Expressions (11) and (12) describe the radiation of isolated molecules and cannot depend on the volume of the system V and the wavelength. The dependence on the wavelength in the case of single photon transitions arises if we begin to consider "mixed" states of the system, the classification of which can be carried out according to the

scheme proposed by Dicke.^[2]

We now find the probability of emission of two coherent photons of the type (\mathbf{k}, λ) with transition of the molecules j' and j'' to the ground state. The wave function of the final state of the system has the form

$$|f\rangle = F(n_+ - 2, n_- + 2)\Phi_2(\mathbf{k}, \lambda). \quad (13)$$

With account of (2), (9), and (13), we get the following expression for the matrix element:

$$\langle f | S^{(2)} | i \rangle = -\frac{g^2}{\hbar^2} \frac{V\sqrt{2}}{\omega_{\mathbf{k}}V} (\mathbf{e}_{\mathbf{k}\lambda}\xi)^2 \exp[i\mathbf{k}(\mathbf{R}_{j'} + \mathbf{R}_{j''})] F_{\mathbf{k}}^2(t). \quad (14)$$

Taking the square of the modulus of (14), we get the factor $\omega^{-4} \sin^4 \omega t$. It is not difficult to show by successive integration by parts that

$$\int_{-\infty}^{\infty} \frac{\sin^4 \omega t}{\omega^4 t^3} d\omega = \frac{2}{3} \pi.$$

Thus the function

$$\delta(\omega) = \frac{3}{2\pi} \lim_{t \rightarrow \infty} \frac{\sin^4 \omega t}{\omega^4 t^3}$$

is the usual Dirac δ -function. Completely analogous to the δ -function is the expression

$$\delta(\omega) = \frac{20}{11\pi} \lim_{t \rightarrow \infty} \frac{\sin^6 \omega t}{\omega^6 t^5}.$$

The latter representation of the δ -function is encountered in calculation of the probability of emission of three coherent photons. In the calculation of radiative transitions of higher order, the introduction of exact δ -functions becomes cumbersome, and one can use approximate expressions obtained from the equations used to define the δ -function in the derivation of (11), namely:

$$\lim_{t \rightarrow \infty} \frac{\sin^4 \omega t}{\omega^4} \approx \lim_{t \rightarrow \infty} \frac{1}{\pi} \frac{\sin^2 \omega t}{\omega^2 t} \pi t^3,$$

$$\lim_{t \rightarrow \infty} \frac{\sin^6 \omega t}{\omega^6} \approx \lim_{t \rightarrow \infty} \frac{1}{\pi} \frac{\sin^2 \omega t}{\omega^2 t} \pi t^5$$

and so forth. Averaging the square of the modulus of (14) over the directions ξ and summing over the field oscillators, we get

$$W_{j'j''}^{2\gamma} = \frac{8}{15\pi} \frac{g^4 (\bar{\xi}^2)^2}{\hbar^4 c^3 V} t^3. \quad (15)$$

We obtain the total probability of emission of two coherent photons by multiplying (15) by the number of combinations of n_+ elements taken two at a time:

$$W^{2\gamma} = C_{n_+}^2 W_{j'j''}^{2\gamma} = \frac{4}{15\pi} \frac{g^4 (\bar{\xi}^2)^2}{\hbar^4 c^3} t^3 \frac{n_+}{V} \frac{n_+ - 1}{V} V. \quad (15')$$

The last formula can also be represented in the

form

$$W^{2\gamma} = \frac{2}{3\pi} \frac{g^2 \bar{\xi}^2 \omega_0}{\hbar^2 c^3} \frac{n_+}{V} V t \frac{3\pi c^3 (n_+ - 1) t}{10\omega_0^2 V} \frac{4}{3\pi} \frac{g^2 \bar{\xi}^2 \omega_0}{\hbar^2 c^3} t. \quad (16)$$

The second factor is regarded as the probability of "collision" between a photon emitted by one of the n_+ molecules and one of the $(n_+ - 1)$ remaining molecules, with the subsequent radiation induced by the latter. One can define the "collision" time τ :

$$\tau = 10\omega_0^2 V / 3\pi c^3 (n_+ - 1), \quad (17)$$

where for $(n_+ - 1)/V \rightarrow 0$ we have $\tau \rightarrow \infty$, i.e., the "collisions" vanish for the isolated molecule.

Finally, let us find the probability of coherent emission of three photons (\mathbf{k}, λ) . Again let there be no photons in the initial state; in the final state, the molecules j' , j'' and j''' undergo transition to the ground state with emission of three coherent photons of the species (\mathbf{k}, λ) .

The matrix element of transition is defined by the equation

$$\langle f | S^{(3)} | i \rangle = \frac{1}{3! (i\hbar)^3} \times T \left\langle f \left| \int_0^t H^{int}(t_1) H^{int}(t_2) H^{int}(t_3) dt_1 dt_2 dt_3 \right| i \right\rangle.$$

Obviously, only the components of $H^{int}(t)$ containing creation operators of photons contribute to the result. Since the matrix elements that correspond to the various intermediate (according to the indices of the molecules) states correspond to equivalent non-connective graphs, the matrix element must be multiplied by their number, which is equal to $3!$. The T product symbol can be omitted, and we finally obtain

$$\langle f | S^{(3)} | i \rangle = \frac{(3!)^{1/2}}{(i\hbar)^3} g^3 (\mathbf{e}_{\mathbf{k}\lambda} \xi)^3 \left(\frac{1}{\omega_{\mathbf{k}} V} \right)^{3/2} \times \exp[i\mathbf{k}(\mathbf{R}_{j'} + \mathbf{R}_{j''} + \mathbf{R}_{j'''})] F_{\mathbf{k}}^3(t).$$

The probability of emission, after carrying out all summations, takes the form

$$W^{3\gamma} = \frac{11}{70\pi} \frac{g^6 (\bar{\xi}^2)^3}{\hbar^6 \omega_0 c^3} \frac{n_+}{V} \frac{n_+ - 1}{V} \frac{n_+ - 2}{V} V t^5. \quad (18)$$

The results (12), (15'), and (18) for coherent emission do not depend on the wavelength. This dependence, as was noted above, arises in the (spontaneous) emission of noncoherent photons. In the case of spontaneous emission of two noncoherent photons with wavelength much smaller than the dimensions of the system, we have for the matrix element of the transition with emission of the photons (\mathbf{k}, λ) and (\mathbf{k}', λ') (at the initial

instant there are no photons)

$$\begin{aligned} \langle f | S^{(2)} | i \rangle &= \frac{g^2}{(i\hbar)^2} \frac{2!}{2!} T \left\langle f \left| \int_0^t H^{int}(t_1) H^{int}(t_2) dt_1 dt_2 \right| i \right\rangle \\ &= \frac{g^2}{(i\hbar)^2} \frac{2!}{2!} \left(\frac{1}{\omega_{\mathbf{k}\omega_{\mathbf{k}'}} V^2} \right)^{1/2} (\mathbf{e}_{\mathbf{k}\lambda} \boldsymbol{\xi}) (\mathbf{e}_{\mathbf{k}'\lambda'} \boldsymbol{\xi}) \\ &\times [\exp(i\mathbf{k}\mathbf{R}_j) \exp(i\mathbf{k}'\mathbf{R}_{j'}) \\ &+ \exp(i\mathbf{k}'\mathbf{R}_j) \exp(i\mathbf{k}\mathbf{R}_{j'})] F_{\mathbf{k}}(t) F_{\mathbf{k}'}(t). \end{aligned}$$

Here we have introduced the factor 2! which takes into account the number of combinations which differ only in a permutation of the indices of molecules j and j' .

We obtain for the transition probability, after averaging over the electron motion and the phases:

$$\begin{aligned} W_{if} &= 2 \frac{g^4}{\hbar^4} \frac{\pi^2 t^2}{\omega_{\mathbf{k}\omega_{\mathbf{k}'}} V^2} \frac{(\bar{\xi}^2)^2}{9} \frac{1}{\pi} \frac{\sin^2[(\omega_{\mathbf{k}} - \omega_0)t/2]}{(\omega_{\mathbf{k}} - \omega_0)^2 t/4} \\ &\times \frac{1}{\pi} \frac{\sin^2[(\omega_{\mathbf{k}'} - \omega_0)t/2]}{(\omega_{\mathbf{k}'} - \omega_0)^2 t/4}. \end{aligned}$$

Since photons with different polarization vectors are taken, averaging over the directions ξ gives

$$|\overline{\mathbf{e}_{\mathbf{k}\lambda} \boldsymbol{\xi}}|^2 = \frac{1}{3} \bar{\xi}^2, \quad |\overline{\mathbf{e}_{\mathbf{k}'\lambda'} \boldsymbol{\xi}}|^2 = \frac{1}{3} \bar{\xi}^2.$$

We can write for the total probability of emission in the system

$$W_{\lambda \ll L}^{\gamma\gamma} = \frac{2}{3\pi} \frac{g^2 \bar{\xi}^2 \omega_0}{\hbar^2 c^3} \frac{n_+}{V} Vt \frac{2}{3\pi} \frac{g^2 \bar{\xi}^2 \omega_0}{\hbar^2 c^3} \frac{n_+ - 1}{V} Vt, \quad (19)$$

i.e., each molecule radiates independently. If the wavelength is much greater than the dimensions of the system [$\exp(i\mathbf{k} \cdot \mathbf{R}_j) \sim 1$], we get for the transition matrix element

$$\langle f | S^{(2)} | i \rangle = 2 \frac{2! g^2}{2! (i\hbar)^2} \left(\frac{1}{\omega_{\mathbf{k}\omega_{\mathbf{k}'}} V^2} \right)^{1/2} (\mathbf{e}_{\mathbf{k}\lambda} \boldsymbol{\xi}) (\mathbf{e}_{\mathbf{k}'\lambda'} \boldsymbol{\xi}) F_{\mathbf{k}}(t) F_{\mathbf{k}'}(t),$$

so that in place of (19) we have a quantity that is twice as large:

$$\begin{aligned} W_{\lambda \gg L}^{\gamma, \gamma} &= 2W_{\lambda \ll L}^{\gamma, \gamma} \\ &= 2 \frac{4}{3} \frac{e^2 \bar{\xi}^2 \omega_0}{\hbar m^2 c^3} \frac{n_+}{V} Vt \frac{4}{3} \frac{e^2 \bar{\xi}^2 \omega_0}{\hbar m^2 c^3} \frac{n_+ - 1}{V} Vt, \quad (20) \end{aligned}$$

i.e., the probability of spontaneous emission of each molecule increases under the action of the radiation field of the neighbors.

Assuming, for the sake of simplicity, that the number of excitations N_0 is the same for all field oscillators, one can write down the formulas for coherent absorption of one, two, or more photons in the presence of the radiation field:

$$\begin{aligned} W_{-1}^{\gamma} &= \frac{2}{3\pi} \frac{g^2 \bar{\xi}^2 N_0 \omega_0}{\hbar^2 c^3} \frac{n_-}{V} Vt, \\ W_{-2}^{\gamma} &= \frac{4}{15\pi} \frac{g^4 (\bar{\xi}^2)^2 N_0 (N_0 - 1)}{2\hbar^4 c^3} \frac{n_-}{V} \frac{n_- - 1}{V} Vt^3 \quad (21) \end{aligned}$$

and so forth. In place of (12), (15'), and (18) we get the expressions

$$\begin{aligned} W^{\gamma} &= \frac{2}{3\pi} \frac{g^2 \bar{\xi}^2 \omega_0}{\hbar^2 c^3} \frac{n_+}{V} Vt (N_0 + 1), \\ W^{2\gamma} &= \frac{4}{15\pi} \frac{g^4 (\bar{\xi}^2)^2}{2\hbar^4 c^3} \frac{n_+}{V} \frac{n_+ - 1}{V} Vt^3 (N_0 + 1) (N_0 + 2), \\ W^{3\gamma} &= \frac{11}{70\pi} \frac{g^6 (\bar{\xi}^2)^3}{\hbar^6 \omega_0 c^3} \frac{(N_0 + 1) (N_0 + 2) (N_0 + 3)}{3! V^3} \\ &\times n_+ (n_+ - 1) (n_+ - 2) Vt^5. \quad (22) \end{aligned}$$

We have calculated above the first large perturbation-theory series terms that contribute to the corresponding order of the radiative transition. It is not difficult to establish the fact that, for example, in addition to the term $S^{(1)}$, all odd pairs of the form $S^{(2m+1)}$ also make a contribution to photon transition. This contribution is small in comparison with the contribution made by $S^{(1)}$ only when the observation time is short in comparison with the time of radiation of the single molecule. Thus the element $S^{(3)}$ gives the following probability of emission of a single photon in the system (at the initial moment there were no photons):

$$W_{(3)}^{\gamma} = (W^{\gamma})^3 / (3!)^2, \quad (23)$$

and the corresponding contribution will be small if the following inequality is satisfied:

$$W_{(3)}^{\gamma} / W^{\gamma} = (W^{\gamma})^2 / (3!)^2 \ll 1. \quad (24)$$

Equations (21) and (22), being referred to unit volume, show that the probabilities of radiative transitions of a given order are proportional to the corresponding powers of the densities. If $n_+ \gg n_-$, that is, inversion has been effected in the system, then the system radiates strongly; conversely, when $n_- \gg n_+$, the system absorbs strongly.

The condition for the applicability of the "one-photon" probability of radiative transitions from an initial state ($N_0 = 0$) is the inequality:

$$\frac{W^{2\gamma}}{W^{\gamma}} = \frac{2}{5} \frac{g^2 \bar{\xi}^2 t^2}{\hbar^2 \omega_0} \frac{n_+ - 1}{V} \ll 1, \quad (25)$$

and in the presence of photons in the system

$$\frac{W^{2\gamma}}{W^{\gamma}} = \frac{1}{5} \frac{g^2 \bar{\xi}^2 t^2}{\hbar^2 \omega_0} (N_0 + 2) \frac{n_+ - 1}{V} \ll 1. \quad (25')$$

The inequality (25') is quickly violated as the density of photons increases. Taking for our estimates $\bar{\xi}^2 \sim 10^{-38}$ and $\omega_0 \sim 10^{15}$, we get the following inequality from (25):

$$t^2 n_+ / V \ll 10^{-9},$$

that is, if $t \sim 10^{-10}$ sec, then the maximum density of excited particles is $\sim 10^{11}$ cm³; if $t \sim 10^{-8}$ sec, then the corresponding value is $\sim 10^5$ cm³. Thus, in a system with sufficiently high molecular densities (both excited and in the ground state) many-photon transitions become most probable; the probability of these transitions increases rapidly upon satisfaction of an inequality which is the opposite of (25'), so that the system radiates in

an extremely strongly bound state.

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