Effect of Dipole-dipole Interaction on the Radiation of Two Excited Atoms

KUNG-CHEN LIU (劉公陳)

Electrophysics Department, National Chiao Tung University, Hsinchu, Taiwan

(Received 6 May 1975)

The extended Weisskopf-Wigner method developed by Ernst and Stehle is used to investigate the radiation of two initially excited atoms with dipole-dipole interaction. It is found that the lifetime for the system is the same as that without dipole-dipole interaction but the energy shift is just the dipole-dipole interaction.

I. INTRODUCTION

THE radiation from atoms has been an attracting problem since Weisskopf and Wigner[1] solved the emission of radiation by a single excited atom. Thereafter, many works *on* the systems of two- and many-atom have been successively done by many people.[2]-[6] Recently, Lee, Lee, and Chang[7] reconsidered these problems as the demonstration for the powerful applications of the so-called multiple-time-scale perturbation theory.

For two atoms, one of which is excited initially, the dipole-dipole interaction is important[4] when their distance is smaller than the wavelength of the transition from the ground state to the excited state. It was shown that in this case the emitted photon may be trapped between the two atoms, which leads to the dipole-dipole interaction, so that the lifetime of the system is equal to that for a single atom, while the energy shift is exactly the dipole-dipole interaction. In fact, it was already known that the dipole-dipole interaction is responsible for the propagation of the collective excitations such as excitons in crystals.[8]

In 1968, Ernst and Stehle[6] extended Weisskopf and Wigner theory and applied it to a system of N atoms with all of them excited initially. According to their viewpoint, the complete treatment of the two-atom problem is the framework for the understanding of the general case. Without considering the

(1) V. Weisskopf and E. Wigner, Z. Physik 63, 54 (1930).
(2) R. H. Dicke, Phys. Rev. 93, 99 (1954).
(3) F. Schwab1 and W. Thirring, Ergeb. Exakt. Naturw. 36, 219 (1964).
(4) M. J. Stephen, J. Chem. Phys. 40, 669 (1964).
(5) D. A. Hutchinson and H.F. Hameka, J. Chem. Phys. 41, 2006 (1964).
(6) V. Ernst and P. Stehle, Phys. Rev. **176,** 1456 (1968).
(7) P.S. Lee, Y. C. Lee and C. T. Chang, Phys. Rev. **AS,** 1722 (1973).
P.S. Lee and Y. C. Lee, Phys. Rev. **AS,** 1727 (1973).
(8) See, for example, A. S Davydov, *Theory of Molecular Excitons* (Plenum, New York 1971).

dipole-dipole interaction, they found that the system of two initially excited atoms has half a lifetime of a single excited atom, exactly as two independently radiating atoms. This was compared to the remarkable influence on the lifetime of a single excited atom in the presence of a nonexcited atom.(*)

In this paper, we consider a system of two initially excited atoms, with their distance smaller than the-wavelength of the transition from the ground state to the excited state, and also take into account the dipole-dipole interaction. In the next section, we shall investigate the effect of the dipole-dipole interaction on the lifetime and the energy shift of the emitted photon for our system by making use of the extended Weisskopf-Wigner theory. The results show that the system has half a lifetime of a single excited atom and there exists a shift in the emitted photon energy. The conclusions and discussions will be given in the last section.

II. FORMULATION

Consider two identical two-level atoms A_1 and A_2 which are initially excited and cited at $\mathbf{x}_1$ and $\mathbf{x}_2$ respectively, with the distance $|\mathbf{x}_1-\mathbf{x}_2|<\Delta^{-1}$(9), where Δ is the energy difference of the atomic levels. The dipole-dipole interaction of this system has the real, nonzero matrix elements of the form(4)

$$V_{12}=\langle g_1e_2;\ \mathbf{k}|\ \hat{V}(t)\ |e_1g_2;\ \mathbf{k}\rangle=\langle e_1g_2;\ \mathbf{k}|\ \hat{V}(t)\ |g_1e_2;\ \mathbf{k}\rangle, \tag{1}$$

where $\hat{V}(t)$ is the dipole-dipole interaction operator,

$$\hat{V}=\frac{(\hat{\mathbf{q}}_1\cdot\hat{\mathbf{q}}_2)\,|\mathbf{x}_1-\mathbf{x}_2|^2-3[\hat{\mathbf{q}}_1\cdot(\mathbf{x}_1-\mathbf{x}_2)][\hat{\mathbf{q}}_2\cdot(\mathbf{x}_1-\mathbf{x}_2)]}{|\mathbf{x}_1-\mathbf{x}_2|^5} \tag{2}$$

in the interaction picture. Here $\hat{\mathbf{q}}=-\sum_i e\hat{\mathbf{r}}_i$ is the dipole oprator for an atom.

Then the equations equivlent to the Schrödinger equation in the interaction picture, in terms of Ernst and Stehle's notations, ") can be written as follow :

$$\begin{aligned}
i\frac{d}{dt}\alpha_0^0(t)&=\sum_{\vec{k}_1}C_{\vec{k}_1}^{(1)*}\frac{e^{i(\Delta-k_1)t}}{(2Vk_1)^{1/2}}\alpha_{\vec{k}_1}^1(t)+\sum_{\vec{k}_1}C_{\vec{k}_1}^{(2)*}\frac{e^{i(\Delta-k_1)t}}{(2Vk_1)^{1/2}}\alpha_{\vec{k}_1}^2(t),\\
i\frac{d}{dt}\alpha_{\vec{k}_1}^1(t)&=C_{\vec{k}_1}^{(1)}\frac{e^{-i(\Delta-k_1)t}}{(2Vk_1)^{1/2}}\alpha_0^0(t)+\sum_{\vec{k}_2}\in(\mathbf{k}_1\mathbf{k}_2)C_{\vec{k}_2}^{(2)*}\frac{e^{i(\Delta-k_2)t}}{(2Vk_2)^{1/2}}\alpha_{\vec{k}_1\vec{k}_2}^{1\ 2}(t)\\
&\quad+V_{12}\alpha_{\vec{k}_1}^2(t),\\
i\frac{d}{dt}\alpha_{\vec{k}_1}^2(t)&=C_{\vec{k}_1}^{(2)}\frac{e^{-i(\Delta-k_1)t}}{(2Vk_1)^{1/2}}\alpha_0^0(t)+\sum_{\vec{k}_2}\in(\mathbf{k}_1\mathbf{k}_2)C_{\vec{k}_2}^{(1)*}\frac{e^{i(\Delta-k_2)}}{(2Vk_2)^{1/2}}\alpha_{\vec{k}_1\vec{k}_2}^{1\ 2}(t)\\
&\quad+V_{12}\alpha_{\vec{k}_1}^1(t),\\
i\frac{d}{dt}\alpha_{\vec{k}_1\vec{k}_2}^{1\ 2}(t)&=\in^{-1}(\mathbf{k}_1\mathbf{k}_2)\Big\{C_{\vec{k}_1}^{(2)}\frac{e^{-i(\Delta-k_1)t}}{(2Vk_1)^{1/2}}\alpha_{\vec{k}_2}^1(t)\\
&\quad+C_{\vec{k}_2}^{(2)}\frac{e^{-i(\Delta-k_2)t}}{(2Vk_2)^{1/2}}\alpha_{\vec{k}_1}^1(t)+C_{\vec{k}_1}^{(1)}\frac{e^{-i(\Delta-k_1)t}}{(2Vk_1)^{1/2}}\alpha_{\vec{k}_2}^2(t)+C_{\vec{k}_2}^{(1)}\frac{e^{-i(\Delta-k_2)t}}{(2Vk_2)^{1/2}}\alpha_{\vec{k}_1}^2(t)\Big\},
\end{aligned} \tag{3}$$

(9) We have used the system of units in which $\hbar=c=1$.

where $\alpha_0^0(t)$ is the probability amplitude of the system with both atoms excited at t, $\alpha^1_{\vec{k}_1}(t)$ ($\alpha^2_{\vec{k}_1}(t)$) is that for the system with $A_1(A_2)$ in its ground state while $A_2(A_1)$ still excited, and a photon being emitted with wave vector $\mathbf{k}_1$, and $\alpha^{1\ 2}_{\vec{k}_1\vec{k}_2}(t)$ is the probability amplitude for the system with both atoms deexcited and two photons being emitted with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$, respectively.[10] Hence, the assumption that the two atoms are in the excited state at $t=0$ can be replaced by the initial conditions:

$$\mathrm{d}(0) = 1,\ \alpha^1_{\vec{k}_1}(0) = \alpha^2_{\vec{k}_1}(0) = \alpha^{1\ 2}_{\vec{k}_1\vec{k}_2}(0) = 0. \tag{4}$$

From Equation (3). we note that due to the selection rules of the dipole-dipole interaction, the time evolution of the probability for one atom in the ground state by emitting a photon of mode k is additive to that for another atom in the ground state by emitting a photon of the same mode through the dipole-dipole interaction. But the probabilities for finding the two atoms both in the excited states or in the ground states evolve in the same ways as though there were no dipole-dipole interaction.

In order to simplify the summations of all photon modes in Equation (3), we observe that the occurence of dipole-dipole interaction does not involve the summations of the photon modes. Thus we are still able to use the ansatz[11] as

$$\begin{aligned}
&\alpha_0^0(t) = \beta_0(t),\\
&\alpha^1_{\vec{k}_1}(t) = C^{(1)}_{\vec{k}_1}\frac{e^{-i(\Delta-k_1)t}}{(2Vk_1)^{1/2}}[\beta^{(+)}_{k_1}(t) + e^{i\vec{k}_1\cdot(\vec{x}_1-\vec{x}_2)}\beta^{(-)}_{k_1}(t)],\\
&\alpha^2_{\vec{k}_1}(t) = C^{(2)}_{\vec{k}_1}\frac{e^{-i(\Delta-k_1)t}}{(2Vk_1)^{1/2}}[\beta^{(+)}_{k_1}(t) + e^{i\vec{k}_1\cdot(\vec{x}_2-\vec{x}_1)}\beta^{(-)}_{k_1}(t)],\\
&\alpha^{1\ 2}_{\vec{k}_1\vec{k}_2}(t) = \epsilon^{-1}(\mathbf{k}_1\mathbf{k}_2)(C^{(1)}_{\vec{k}_1}C^{(2)}_{\vec{k}_2} + C^{(2)}_{\vec{k}_1}C^{(1)}_{\vec{k}_2})\frac{e^{-i(\Delta-k_1)t}}{(2Vk_1)^{1/2}}\\
&\qquad\frac{e^{-i(\Delta-k_2)t}}{(2Vk_2)^{1/2}}\Big\{\beta^{(+)}_{k_1k_2}(t) + [(e^{i\vec{k}_1\cdot(\vec{x}_1-\vec{x}_2)} + e^{i\vec{k}_2\cdot(\vec{x}_1-\vec{x}_2)})^{-1}\\
&\qquad + (e^{i\vec{k}_1\cdot(\vec{x}_2-\vec{x}_1)} + e^{i\vec{k}\cdot(\vec{x}_2-\vec{x}_1)})^{-1}]\beta^{(-)}_{k_1k_2}(t)\Big\}.
\end{aligned} \tag{5}$$

The functions $\beta^{(\pm)}_{k_1}(t)$ are assumed to depend on the energy k_1 only and are appreciably different from zero only in small neighborhoods of $k_1=\Delta$ of dimensions Γ_0, where Γ_0 is the half natural linewidth of Weisskopf and Wigner. Similarly, $\beta^{(\pm)}_{k_1k_2}(t)$ are assumed to depend on the energy k_1k_2 only and appreciably different from zero only in small neighborhoods of $k_1=\Delta$ and $k_2=\Delta$ of dimensions Γ_0^2.

Inserting Equation (5) into Equation (3) and applying the same procedures as in Ref. 6, we wil have the coupled set of equations for the functions $\beta^{(\pm)}_{k_1}(t)$ and $\beta^{(\pm)}_{k_1k_2}(t)$.

(10) For the definitions of the other notations, see Sec. II. in Ref. 6.
(11) Equation (4.3) in Ref. 6.

$$
\begin{aligned}
&i\frac{d}{dt}\beta_0(t)=\frac{2\Gamma_0}{\pi}\int_{-\infty}^{\infty}dk_1\beta_{k_1}^{(+)}(t)+\frac{2\Gamma_{12}}{\pi}\int_{-\infty}^{\infty}dk_1\beta_{k_1}^{(-)}(t),\\
&(\Delta-k_1+i\frac{d}{dt})\beta_{k_1}^{(+)}(t)\\
&\quad=\beta_0(t)+\frac{\Gamma_0}{\pi}\int_{-\infty}^{\infty}dk_2\beta_{k_1k_2}^{(+)}(t)+\frac{\Gamma_{12}}{\pi}\int_{-\infty}^{\infty}dk_2\beta_{k_1k_2}^{(-)}(t)\\
&\qquad+V_{12}\beta_{k_1}^{(-)}(t),\qquad (6)\\
&(\Delta-k_1+i\frac{d}{dt})\beta_{k_1}^{(-)}(t)\\
&\quad=\frac{\Gamma_{12}}{\pi}\int_{-\infty}^{\infty}dk_2\beta_{k_1k_2}^{(+)}(t)+\frac{\Gamma_0}{\pi}\int_{-\infty}^{\infty}dk_2\beta_{k_1k_2}^{(-)}(t)\\
&\qquad+V_{12}\beta_{k_1}^{(+)}(t),\\
&(\Delta-k_1+\Delta-k_2+i\frac{d}{dt})\beta_{k_1k_2}^{(\pm)}(t)\\
&\quad=\beta_{k_1}^{(\pm)}(t)+\beta_{k_2}^{(\pm)}(t).
\end{aligned}
$$

Note that in Equation (6), Γ_{12} is the only function concerned with the distance between the two atoms, $|\mathbf{x}_1-\mathbf{x}_2|$. It will approach to Γ_0 if $|\mathbf{x}_1-\mathbf{x}_2|<\frac{\Delta^{-1}}{2}$.

The exact solution of Equation (6) is easily to be found, and the results are as follow:

$$
\begin{aligned}
&\beta_0(t)=e^{-2\Gamma_0 t},\\
&\beta_{k_1}^{(\pm)}(t)=\frac{1}{2}\Big(e^{-(\Gamma_0+\Gamma_{12})t}\,\frac{e^{-(\Gamma_0-\Gamma_{12})t}-e^{i(\Delta-k_1-V_{12})t}}{\Delta-k_1-V_{12}-i(\Gamma_0-\Gamma_{12})}\\
&\quad\pm e^{-(\Gamma_0-\Gamma_{12})t}\,\frac{e^{-(\Gamma_0+\Gamma_{12})t}-e^{i(\Delta-k_2+V_{12})t}}{\Delta-k_1+V_{12}-i(\Gamma_0+\Gamma_{12})}\Big),\qquad (7)\\
&\beta_{k_1k_2}^{(\pm)}(t)=\frac{1}{2}\Big[\frac{1}{\Delta-k_1-i(\Gamma_0-\Gamma_{12})}\Big(\frac{e^{-2\Gamma_0 t}-e^{i(\Delta-k_1+\Delta-k_2)t}}{\Delta-k_1+\Delta-k_2-2i\Gamma_0}-\\
&\quad-e^{i(\Delta-k_1)t}\,\frac{e^{-(\Gamma_0+\Gamma_{12}+iV_{12})t}-e^{i(\Delta-k_2)t}}{\Delta-k_2+V_{12}-i(\Gamma_0+\Gamma_{12})}\Big)+\\
&\quad\frac{1}{\Delta-k_2-V_{12}-i(\Gamma_0-\Gamma_{12})}\Big(\frac{e^{-2\Gamma_0 t}-e^{i(\Delta-k_1+\Delta-k_2)t}}{\Delta-k_1+\Delta-k_2-2i\Gamma_0}\\
&\quad-e^{i(\Delta-k_2)t}\,\frac{e^{-(\Gamma_0+\Gamma_{12}+iV_{12})t}-e^{i(\Delta-k_1)t}}{\Delta-k_2+V_{12}-i(\Gamma_0+\Gamma_{12})}\Big)\Big]\pm\\
&\quad\frac{1}{2}\Big[\frac{1}{\Delta-k_1+V_{12}-i(\Gamma_0+\Gamma_{12})}\Big(\frac{e^{-2\Gamma_0 t}-e^{i(\Delta-k_1+\Delta-k_2)t}}{\Delta-k_1+\Delta-k_2-2i\Gamma_0}\\
&\quad-e^{i(\Delta-k_1)t}\,\frac{e^{-(\Gamma_0-\Gamma_{12}-iV_{12})t}-e^{i(\Delta-k_2)t}}{\Delta-k_2-V_{12}-i(\Gamma_0-\Gamma_{12})}\Big)+\\
&\quad\frac{1}{\Delta-k_2+V_{12}-i(\Gamma_0+\Gamma_{12})}\Big(\frac{e^{-2\Gamma_0 t}-e^{i(\Delta-k_1+\Delta-k_2)t}}{\Delta-k_1+\Delta-k_2-2i\Gamma_0}
\end{aligned}
$$

$$-e^{i(\Delta-k_2)t}\frac{e^{-(\Gamma_0-\Gamma_{12}-iV_{12})t}-e^{i(\Delta-k_1)t}}{\Delta-k_2-V_{12}-i(\Gamma_0-\Gamma_{12})}\Big)\Big].$$

Combining Equations (5) and (7), we have

$$\alpha_0^0(t)=e^{-2\Gamma_0 t}, \tag{8}$$

which means that the initial state $|e_1e_2;0\rangle$ will decay out like $e^{-2(2\Gamma_0)t}$.

III. DISCUSSIONS AND CONCLUSIONS

As shown above, our system will decay out with a lifetime $(4\Gamma_0)^{-1}$, one half for that of a single excited atom, as the case without considering the dipole-dipole interaction beween two atoms.[6] This is very easily to be understood from the fact that the introduction of the dipole-dipole interaction does not affect the equation of the time evolution for the probability amplitude of the system in the initial state. Each atom has the same probability to emit a photon. Hence there is doubly probable for the system to decay out and thus the lifetime is reduced by one half.

This can also be understood from the viewpoint of symmetry. As we know, the system of the two initially excited atoms is quite different from the system containing two atoms with only one excited initially as the exchange symmetry of the two atoms is considered. The former is an exactly symmetrical state with respect to exchange of the two atoms. In that case the symmetrical state is capable of emitting photons and decaying to the ground state with two unexcited atoms which is also symmetrical with respect to exchange of the two atoms. Hence it is natural for the symmetrical state of two initially excited atoms to emit photons with a probability of unit. In this situation the emitted photons will not be trapped between the two atoms, so there exists no Dicke's superradiant state.[2] Thus for the system of the two excited atoms, there is a lifetime doubly shorter than that for a single excited atom.

Futher investigation shows that Equation (7) may be obtained by replacing Γ_{12}, the only factor concerned with the distance between two atoms, by $\Gamma_{12}+iV_{12}$ in the resultant equation for the case without considering dipole-dipole interaction.[12] The apperance of V_{12} in the imaginary part means that the dipole-dipole interaction will introduce energy shift by V_{12} to the emission of radiation.

For a three-dimensional crystals, such as insultors, containing N all excited atoms arrayed in lattices with dipole-dipole interaction, then from the above discussions, it is ready to expect that

$$\alpha_0^0(t)=e^{-N\Gamma_0 t}. \tag{9}$$

This means that it has a lifetime $(2N\Gamma_0)^{-1}$ in contrast to the case in which the number of the excited atoms is much less than N. In fact, for the latter case, Hopfield[13] has shown that the emitted photons will be trapped in the crystal to form a mixture of photons and excitons as the eigenstates of the system of crystal and radiation field.

(12) i.e. Equation (4.5) in Ref. 6.

(13) J. J. Hopfield, Phys. Rev. **112,** 1555 (1958).

ACKNOWLEDGEMENTS

The author is indebted very much to Professor Y. C. Lee of State University of New York at Buffalo and Professor Y. Shan of National Tsing Hua University for their stimulating suggestions and enlightening discussions. He also wishes to thank Dr. D. S. Chuu for his critical reading of the manuscript.