Retarded interaction of two nonidentical atoms*

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We extend our recent work on the causal aspects of two-atom spontaneous decay to the case of nonidentical atoms.

In a recent paper,[1] we considered the problem of spontaneous emission from a system of two *identical* atoms a fixed distance r apart, one of which is excited at $t = 0$. Our solutions for the various probability amplitudes exhibit the effects of all the retardation times nr/c ($n = 0, 1, 2, \ldots$) due to the causal exchange of the excitation between the atoms. These solutions reduce to the familiar results for the resonant, or "cooperative," interaction after a large number of retardation times have elapsed. Earlier work[2] on the problem showed the dependence of the probability amplitudes only on the first retardation time r/c, and did not make the connection with the allied problem of cooperative decay. The causal aspect of this simple system has played a venerable heuristic role in the development of the nonrelativistic quantum theory of the interaction of light with atoms.[3]

Interest in this problem seems to have been revived after Arecchi and Courtens[4] treated the retardation problem in an appendix to a recent paper. In a recent study of superradiance[5] it was found that the retardation times nr/c appear naturally if the Markov approximation is not made (the memory implied by a non-Markovian process is reflected in the dependence of each atom's evolution upon the dynamics, at an earlier time r/c, of the other atom). In addition, the results we derived in Ref. 1 were later obtained independently by Schuurmans.[6] In view of this interest, we feel it worthwhile to present a straightforward extension of the results of Ref. 1 in order to compare these more recent results with the much earlier, and frequently quoted results of Fermi[7] and Breit[8] on the causal exchange of excitation between *nonidentical* atoms.

All of the treatments referred to use the two-level approximation: if atom 1 is initially in its first excited state and atom 2 in its ground state, then it is assumed that the only remaining states of importance are the ground state of atom 1 and the first excited state of atom 2. This approximation is meaningful only if the atoms have a near resonance of their respective transition frequencies, or are identical. Recent work[1,4-6] has been concerned with cooperative decay and assumed the

two atoms to be identical. On the other hand, both Fermi[7] and Breit,[8] in what have become the "standard" treatments of the problem, assumed that the two two-level atoms may have different transition frequencies and natural linewidths.

We therefore consider here the case of nonidentical atoms. Let atoms 1 and 2 have (circular) transition frequencies ω_1 and ω_2, respectively, and corresponding free-atom natural linewidths β_1 and β_2. Following the notation of Ref. 1, let $b_1(t)$ denote the probability amplitude for the state with atom 1 in its first excited state, atom 2 in its ground state, and the radiation field in the vacuum state, and let $b_2(t)$ be the corresponding amplitude for the state with atom 1 in its ground state and atom 2 in its first excited state. Our initial condition will be $b_1(0) = 1$, $b_2(0) = 0$.

The Hamiltonian is

$$H = H_A + H_F - i\hbar \sum_{l=1,2} \sum_{\vec{k}\lambda} [g_l(\vec{k}_\lambda)\sigma_l^- + g_l^*(\vec{k}_\lambda)\sigma_l^+]$$
$$\times [a(\vec{k}_\lambda) - a^\dagger(\vec{k}_\lambda)] ,$$

where the notation follows Ref. 1, to which the reader is referred for fuller explanation. We will use Fermi's choice of orientation for the dipoles: The interatomic axis defines the $\hat{z}$ axis, and both transition dipoles are oriented along the $\hat{x}$ axis. We choose to work with the same "essential states" as those in Ref. 1, with the obvious generalization to different transition frequencies for the atoms.

Following the same approach and approximations used previously,[1] the coupled equations of motion for the probability amplitudes derived from the Schrödinger equation can be solved by Laplace transform. We find that

$$b_1(t) = \sum_{n=0}^{\infty} (-1)^n F^{2n}(\bar{k}_0 r) I_n^{(1)}[t - (2nr/c)] , \qquad (1)$$

$$b_2(t) = \sum_{n=0}^{\infty} (-1)^n F^{2n+1}(\bar{k}_0 r) I_n^{(2)}(t - (2n+1)r/c) ,$$
$$(2)$$

where

$$F(x) \equiv -\frac{3i}{2} (\beta_1\beta_2)^{1/2}\left(\frac{1}{x} - \frac{1}{x^3} + \frac{i}{x^2}\right) e^{ix} , \qquad (3)$$

$$\bar{k}_0 \equiv \bar{\omega}_0/c = (\omega_1 + \omega_2)/2c . \tag{4}$$

We have used the approximation[9] $(\omega_1 + \omega_2)^2 \approx 4\omega_1\omega_2$ in deriving Eq. (3); the integrals $I_n^{(1)}$ and $I_n^{(2)}$ are

$$I_n^{(1)}(\tau) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{dz\, e^{iz\tau}}{(z - i\Omega_1)^{n+1}(z - i\Omega_2)^n} , \tag{5}$$

$$I_n^{(2)}(\tau) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{dz\, e^{iz\tau}}{(z - i\Omega_1)^{n+1}(z - i\Omega_2)^{n+1}} , \tag{6}$$

with $\Omega_1 = \beta_1 + i(\omega_1 - \omega_2)/2$, $\Omega_2 = \beta_2 - i(\omega_1 - \omega_2)/2$. The integrals $I_n^{(1)}(\tau)$ and $I_n^{(2)}(\tau)$ vanish for $\tau < 0$. For $\Omega_1 = \Omega_2$, the case of identical atoms, we recover our previous results of Ref. 1.

The older work on this problem was aimed at showing that $b_2(t) = 0$ for $t < r/c$. This aspect of the system can be seen by retaining only the first term on the right-hand side of Eq. (2):

$$b_2(t) \approx F(\bar{k}_0 r) I_0^{(2)}(t - r/c) ,$$

$$= F(\bar{k}_0 r) U(t - r/c) \left(\frac{e^{-\Omega_2(t - r/c)} - e^{-\Omega_1(t - r/c)}}{(\Omega_2 - \Omega_1)} \right) , \tag{7}$$

where U is the unit step function. The retention of only this term can be justified by assuming that the atoms are very far apart $(\bar{k}_0 r \gg 1)$. Then $F(\bar{k}_0 r)$ can be approximated by its far-field part:

$$F(\bar{k}_0 r) \cong -(3i/2\bar{k}_0 r)(\beta_1\beta_2)^{1/2} e^{i\bar{k}_0 r} . \tag{8}$$

With these approximations, the probability that atom 2 is excited is

$$|b_2(t)|^2 \cong \frac{9\beta_1\beta_2}{4\bar{k}_0^2 r^2} \frac{U(t - r/c)}{(\beta_2 - \beta_1)^2 + (\omega_2 - \omega_1)^2}$$

$$\times [e^{-2\beta_1(t - r/c)} + e^{-2\beta_2(t - r/c)}$$

$$-2 e^{-(\beta_1 + \beta_2)(t - r/c)} \cos(\omega_2 - \omega_1)(t - r/c)] . \tag{9}$$

This is just the result given by Breit.[8] Fermi's discussion[7] is restricted to the case $\beta_1 \gg \beta_2$ and for times such that $\beta_1(t - r/c) \gg 1$, but $\beta_2(t - r/c) \ll 1$. If we impose these restrictions on (9) we obtain

$$|b_2(t)|^2 \cong \frac{9\beta_1\beta_2}{4\bar{k}_0^2 r^2} \frac{U(t - r/c)}{(\omega_2 - \omega_1)^2 + \beta_1^2} , \tag{10}$$

which, under the previously used assumption[9] that $(\omega_1 + \omega_2)^2 \approx 4\omega_1\omega_2$, is Fermi's result. The apparent discontinuity in Eq. (10) is seen to be spurious, since in approximating the result (9) by (10) it is assumed that $t \gg (r/c) + 1/\beta_1$. For this reason, Eq. (10) exhibits the retardation-time causal effects in a less convincing form than (9).

For the opposite condition $\beta_1 \rightarrow \beta_2$, $\omega_1 \rightarrow \omega_2$, we

obtain from (7), for $k_0 r \gg 1$, the amplitude that the second atom be excited:

$$b_2(t) = (3i/2k_0 r) e^{ik_0 r} \beta(t - r/c) U(t - r/c) e^{-\beta(t - r/c)} + \cdots , \tag{11}$$

and the probability

$$|b_2(t)|^2 = (3/2k_0 r)^2 U(t - r/c)[\beta(t - r/c)]^2 e^{-2\beta(t - r/c)} + \cdots . \tag{12}$$

This is exactly our previous result for identical atoms.

Equation (9) only applies for times $t < 3r/c$, but may be regarded as an accurate approximation if $\bar{k}_0 r \gg 1$, since successive terms in the series (2) are multiplied by further powers of $(\bar{k}_0 r)^{-1}$. Arguments of this kind were invoked[7,8] to justify the approximation

$$b_1(t) \cong I_0^{(1)}(t) = e^{-\Omega_1 t} \tag{13}$$

used right from the start of the calculation in the earlier work. Note, however, that the second term on the right-hand side of Eq. (1), although of order $(\bar{k}_0 r)^{-2}$, still contributes to $|b_1(t)|^2$ a term of order $(\bar{k}_0 r)^{-2}$ due to an interference cross term with the first term of Eq. (1). If terms of order $(\bar{k}_0 r)^{-2}$ are kept in $|b_2(t)|^2$, they should be retained in $|b_1(t)|^2$.

Of course, the two atoms need to have quite closely matching transition frequencies before they can exchange excitation energy, and this is reflected in the sharply peaked resonance behavior exhibited in Eq. (9).[10]

Finally, we should mention that neither the appearance of retardation times nor the phenomenon of cooperation are inherently quantum mechanical in origin. It can be shown quite straightforwardly that both effects are exhibited by a system of two classical dipoles a distance r apart, coupled by their radiation fields.

To summarize, we have presented a formal solution to the problem of spontaneous radiative decay from a pair of *nonidentical* atoms which exhibits all of the retardation times nr/c. As was the case with our earlier work on identical atoms, the truncated solutions involving only the first retardation time r/c reduce to previously reported results.

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[2] See Ref. 1 for references to earlier work.

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[8] G. Breit, Rev. Mod. Phys. **5**, 91 (1933).

[9] This assumption is also made by Breit (Ref. 8); this is a reasonable approximation: if ω_1 is very different from ω_2, the atoms cannot of course exchange excitation. This is reflected in the resonance denominator of Eq. (9).

[10] The cooperative decay of two nonidentical atoms has been considered by A. A. Varfolomeev, Zh. Eksp. Teor. Fiz. **59**, 1702 (1970) [Sov. Phys.—JETP **32**, 926 (1971)].