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Spontaneous Emission of a Two-Level System and the Influence of the Rotating-Wave Approximation on the Final State. I.

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By using a modified Robertson projection technique an exact equation of motion for the expectation value of the population inversion operator S^z of a single two-level atom in the case of spontaneous emission is derived. Afterwards, by making the Markov approximation, it is shown that the ground state expectation value $\langle S^z \rangle_t = -1/2$ for $t \to \infty$ will be reached only if the rotating-wave approximation or the Born approximation is made additionally.

KEY WORDS: Quantum statistical mechanics of open systems; spontaneous emission; two-level atom; modified Robertson projection technique; Markov approximation; rotating-wave approximation.

1. INTRODUCTION

In this work we will examine the interesting question of whether a single two-level atom being in its excited state really decays to its unperturbed ground state by spontaneous emission. It seems to us that the answer to this question is not quite clear in the literature. Most of the textbooks, review works, and articles (see, e.g., Refs. 1–4), beginning with the famous Weisskopf–Wigner[(5)] article, treat this question only in the rotating-wave approximation (RWA), i.e., neglecting the antiresonant terms in the Hamiltonian, and/or in the second-order perturbation theory, the so-called Born approximation (BA). They all came to the conclusion that the final state of the atom is its unperturbed ground state. However, we will show that this is correct only as long as the RWA and/or the BA is made.

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In Section 2, by using a modified Robertson[6] projection technique developed in Refs. 7 and 8 recently, we will derive an exact equation of motion (EM) for the expectation value (EV) of the atomic population inversion operator S^z. Afterwards, by making the Markov approximation and the long-time limit, but without making any kind of approximation as to the strength of the interaction with the radiation field, we shall obtain a differential equation, whose solution shows that the ground state EV $\langle S^z \rangle_{t\to\infty} = -1/2$ will be reached only if the RWA, or the BA is made additionally.

In Section 3, by using the nondecay probability[9,10] we will examine the reasonableness of the results obtained in Section 2.

In the Appendix we will show that the dipole moment $\langle S^{\pm} \rangle_t$ remains zero if it was zero initially.

2. CLOSED EQUATION OF MOTION FOR SPONTANEOUS EMISSION OF A SINGLE TWO-LEVEL ATOM

The Hamiltonian for a single two-level atom (system S) interacting with the radiation field (system R) is given by[2,3]

$$H = H_0 + H_{SR} = \omega S^z \otimes I_R + I_S \otimes \sum_{\mathbf{k}s} \omega_k a^+_{\mathbf{k}s} a^-_{\mathbf{k}s} + (S^+ - S^-) \otimes \sum_{\mathbf{k}s} (g_{\mathbf{k}s} a^-_{\mathbf{k}s} - g^*_{\mathbf{k}s} a^+_{\mathbf{k}s}), \qquad (\hbar = 1) \quad (1)$$

where $S^z = \frac{1}{2}(|1\rangle\langle 1| - |2\rangle\langle 2|)$ is the population inversion operator, $S^+ = |1\rangle\langle 2|$, $S^- = |2\rangle\langle 1|$ are the dipole moment operators with $|1\rangle, |2\rangle$ being the atomic excited and ground state, $a^{\pm}_{\mathbf{k}s}$ are the photon creation and annihilation operators for the mode $\mathbf{k}s$, and $g_{\mathbf{k}s} = -i\omega(2\pi/\omega_k L^3)^{1/2}(\mathbf{D}_{12} \cdot \mathbf{e}_{\mathbf{k}s})$ is the coupling constant with $\mathbf{D}_{12}$ as the dipole matrix element, $\mathbf{e}_{\mathbf{k}s}$ as the polarization vector (s is the polarization index), and L^3 as the volume of the field. Further, ω is the energy separation of the two atomic levels, $\omega_k = kc$ and I_S, I_R are the unit operators in the Hilbert spaces $\mathcal{H}_S$ and $\mathcal{H}_R$ of systems S and R.

At $t = 0$, we assume that the atom is in the excited state and the radiation field in the vacuum state:

$$\rho(0) = \rho_S(0) \otimes \rho_R(0) = |1\rangle\langle 1| \otimes |\{0\}\rangle\langle\{0\}| \quad (2)$$

where $\rho(t)$ is the statistical density operator, which satisfies the Liouville (von Neumann) equation, and $\rho_S(t) = \mathrm{Tr}_R \rho(t)$, $\rho_R(t) = \mathrm{Tr}_S \rho(t)$ are the reduced density operators.

By introducing the generalized canonical density operator for S,[8]

$$\sigma_S(t) = \tfrac{1}{2} I_S + 2S^z \langle S^z \rangle_t \quad (3)$$

where we made use of the fact that the dipole moment $\langle S^{\pm}\rangle_t$ remains zero, if it was zero initially (see the exact proof in the Appendix), we can write

$$\sigma_S(t) \otimes \rho_R(0) = P\rho(t) + \tfrac{1}{2} I_S \otimes \rho_R(0) \tag{4}$$

$$PA = \rho_R(0) \otimes 2S^z \mathrm{Tr}_{SR}(S^z A) \tag{5}$$

with any operator A in the product space $\mathscr{H}_S \otimes \mathscr{H}_R$. By using the modified Robertson projection-operator technique,[7,6] i.e., by differentiating and transforming Eq. (4), and afterwards integrating it by applying an integrating operator $T(t,t')$, we obtain a connecting equation between $\rho(t)$ and $\sigma_S(t) \otimes \rho_R(0)$:

$$\rho(t) - \sigma_S(t) \otimes \rho_R(0) = -i\int_0^t dt'\, T(t,t')(I-P)(L_0 + L_{SR})\sigma_S(t') \otimes \rho_R(0) \tag{6}$$

with

$$T(t,t') = \exp\left[-i(t-t')(I-P)(L_0 + L_{SR})\right] \tag{7}$$

and $I = I_S \otimes I_R$, $L_0 = [H_0, \ldots]$, $L_{SR} = [H_{SR}, \ldots]$.

We now let the operator $iS^z(L_0 + L_{SR})$ act upon Eq. (6) and afterwards take the trace over it, which gives us an *exact closed* EM for the EV $\langle S^z\rangle_t$:

$$\frac{\partial \langle S^z\rangle_t}{\partial t} = -\int_0^t d\tau \left[k_1(\tau) + 2k_2(\tau)\langle S^z\rangle_{t-\tau}\right] \tag{8}$$

with

$$k_1(\tau) = \mathrm{Tr}_{SR}\left[\tilde{U}(\tau,0) L_{SR} \tfrac{1}{2} I_S \otimes \rho_R(0)\right] \tag{9}$$

$$k_2(\tau) = \mathrm{Tr}_{SR}\left[\tilde{U}(\tau,0) L_{SR} S^z \otimes \rho_R(0)\right] \tag{10}$$

$$\tilde{U}(\tau,0) = S^z L_{SR} U(\tau,0) e^{-i\tau L_0} \tag{11}$$

$$U(\tau,0) = \mathscr{T} \exp\left[-i\int_0^\tau dt'\,(I-P) e^{-it'L_0} L_{SR} e^{it'L_0}\right] \tag{12}$$

where we made an expansion of $T(t,t')$ in powers of the interaction H_{SR} by using a time-ordering operator $\mathscr{T}$ and the fact that P commutes with H_0.

The RWA on the Hamiltonian H_{SR} of Eq. (1), i.e., the neglecting of the antiresonant terms $S^- \otimes a_{\mathbf{k}s}^-$ and $S^+ \otimes a_{\mathbf{k}s}^+$ gives

$$L_{SR}^{\mathrm{RWA}} \tfrac{1}{2} I_S \otimes \rho_R(0) = L_{SR}^{\mathrm{RWA}} S^z \otimes \rho_R(0) \tag{13}$$

and

$$k_1^{\mathrm{RWA}}(\tau) = k_2^{\mathrm{RWA}}(\tau) \tag{14}$$

By taking the limit $V \to \infty$ (i.e., replacing the summation over $\mathbf{k}$ by an integral), and making the Markov approximation with long-time limit [i.e., in integrand of Eq. (8) $\langle S^z \rangle_{t-\tau}$ is replaced by $\langle S^z \rangle_t$ and for $t \gg 1/\omega$ the upper limit of the time integration is replaced by ∞ by introducing a damping factor $e^{-\epsilon\tau}$, where $\epsilon \to {}^+0$ after performing the integration], and retaining all order of interaction we obtain

$$\frac{\partial \langle S^z \rangle_t}{\partial t} = -\bar{k}_1^{\mathrm{RWA}}(1 + 2\langle S^z \rangle_t) \tag{15}$$

$$\langle S^z \rangle_t = -\tfrac{1}{2} + c e^{-2\bar{k}_1^{\mathrm{RWA}} t}, \qquad c = \mathrm{const} \tag{16}$$

$$\bar{k}_i \equiv \lim_{\epsilon \to {}^+0} \int_0^\infty d\tau\, e^{-\epsilon\tau} k_i(\tau), \qquad i = 1, 2 \tag{17}$$

In the literature c is usually calculated from the initial condition.

When no RWA is made then

$$L_{SR} S^z \otimes \rho_R(0) = L_{SR} \tfrac{1}{2} I_S \otimes \rho_R(0) - \sum_{\mathbf{k}s} \left(g_{\mathbf{k}s} S^- \otimes |\{0\}\rangle\langle\{0\}| a_{\mathbf{k}s}^- - g_{\mathbf{k}s}^* S^+ \otimes a_{\mathbf{k}s}^+ |\{0\}\rangle\langle\{0\}| \right) \tag{18}$$

and

$$\Delta\bar{k} = \bar{k}_1 - \bar{k}_2 = \lim_{\epsilon \to {}^+0} \int_0^\infty d\tau\, e^{-\epsilon\tau} \times \mathrm{Tr}_{SR} \Big\{ (L_{SR} S^z) U(\tau, 0) \sum_{\mathbf{k}s} \Big[g_{\mathbf{k}s}^* e^{-i(\omega_k + \omega)\tau} S^+ \otimes a_{\mathbf{k}s}^+ |\{0\}\rangle\langle\{0\}| - g_{\mathbf{k}s} e^{i(\omega_k + \omega)\tau} S^- \otimes |\{0\}\rangle\langle\{0\}| a_{\mathbf{k}s}^- \Big] \Big\} \tag{19}$$

In the BA $U(\tau, 0) = I$, and only the antiresonant terms with $e^{\pm i(\omega_k + \omega)\tau}$ play a role:

$$\Delta\bar{k}^{\mathrm{BA}} = \pi \sum_{\mathbf{k}s} |g_{\mathbf{k}s}|^2 \delta(\omega_k + \omega) = 0 \tag{20}$$

because $\omega > 0$ and $\omega_k \geqslant 0$. But in an approximation of higher order than the BA it can easily be shown that the terms in $U(\tau, 0)$ give rise to a nonzero value of $\Delta\bar{k}$, i.e., $\bar{k}_1 \neq \bar{k}_2$. Then instead of Eq. (16) we have

$$\langle S^z \rangle_t = -\frac{\bar{k}_1}{2\bar{k}_2} + c_1 e^{-2\bar{k}_2 t}, \qquad c_1 = \mathrm{const} \tag{21}$$

From this it can be followed that if no RWA or BA is made, the atom initially in its excited state does not decay to its unperturbed ground state, i.e., $\langle S^z \rangle_{t\to\infty} \neq -1/2$, which means $\langle S^z \rangle_{t\to\infty} > -1/2$.

An interesting fact, which is being often overlooked, is that the state $|\psi(0)\rangle = |2\rangle \otimes |\{0\}\rangle$, in which the atom is in its ground state $|2\rangle$, is not a stationary state because $[H_0, H_{SR}] \neq 0$. Knowing this it can be immediately seen from Eq. (21) that for the above initial condition ($\langle S^z \rangle_0 = -1/2$), $\bar{k}_1 \neq \bar{k}_2$, because for $\bar{k}_1 = \bar{k}_2$ the solution of Eq. (21) would be a steady state value: $\langle S^z \rangle_t = -1/2$. This means that only in the RWA it will be $\langle S^z \rangle_{t\to\infty} = -1/2$, because $[H_0, H_{SR}^{\mathrm{RWA}}] = 0$, but otherwise $\langle S^z \rangle_{t\to\infty} > -1/2$.

3. THE NONDECAY PROBABILITY OF THE INITIAL STATE

That the results obtained in Section 2 are reasonable can easily be seen by examining the nondecay probability[9,10] of the initial state $|\psi(0)\rangle$ of system $S + R$. The condition for the decay of the initial state is[9,10]

$$\lim_{t\to\infty} |\langle \psi(0) \mid \psi(t) \rangle|^2 = 0 \tag{22}$$

(As pointed out by Krylov and Fock[9] the vanishing of the nondecay probability of an unstable state at the infinity follows from the Riemann–Lebesgue lemma.)

The decay condition can be fulfilled by the general state vector

$$|\psi(t\to\infty)\rangle = \sum_{\alpha=1,2} \sum_{n_1, n_2, \ldots, n_i, \ldots = 0}^{\infty} c_{\alpha n_1 n_2 \ldots n_i \ldots}(t\to\infty) \times |\alpha\rangle \otimes |n_1 n_2 \ldots n_i \ldots\rangle \tag{23}$$

$$1 = \langle \psi(t) \mid \psi(t) \rangle = \sum_{\alpha=1,2} \sum_{n_1, n_2, \ldots, n_i, \ldots = 0}^{\infty} |c_{\alpha n_1 n_2 \ldots n_i \ldots}(t)|^2 \tag{24}$$

with $c_{1\{0\}}(t\to\infty) = 0$ and $\alpha + \sum_{i=0}^{\infty} n_i = 2l + 1$, $l = 0, 1, 2, \ldots$ for the atom being initially in its excited state $|\psi(0)\rangle = |1\rangle \otimes |\{0\}\rangle$. $c_{\alpha n_1 n_2 \ldots n_i \ldots}(t)$ are the probability amplitudes, n_i is the number of photons in i mode, and each $i = 1, 2, \ldots$ stands for a $\mathbf{k}s$ mode.

But in the case of the RWA it holds that

$$\sum_{i=0}^{\infty} n_i = \alpha - 1, \qquad \alpha = 1, 2 \tag{25}$$

According to this, for the decay of $|\psi(0)\rangle = |1\rangle \otimes |\{0\}\rangle$, it must hold that $c_{1 n_1 n_2 \ldots n_i \ldots}(t\to\infty) = 0$ for all n_i. This means that the atom must decay to its unperturbed ground state. In the RWA the ground state $|2\rangle$ is a steady state.

4. CONCLUSION

In Section 2 it was derived a closed EM for the energy of a single two-level atom, interacting with the vacuum radiation field, in the exact and Markov approximated form. Naturally, Eq. (21) is only exact in the Markov approximation and the exponential behavior of $\langle S^z \rangle_t$ is an approximation, as was pointed out by Khalfin.[10] But just this Markov approximation makes it possible to show that there is a radiative correction to the ground state of the atom, when no kind of RWA is made. This radiative correction has the consequence that the final state of the atom is not its unperturbed ground state, as is usually expected in the literature, but the radiative corrected ground state. In the subsequent paper we will show that in the BA there is a radiative correction to the final state of the atom.

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APPENDIX

In the case of spontaneous emission we will exactly show that if the dipole moment of the atom was zero initially (i.e., the atom is in the excited or in the ground state), it remains zero for all future times.

For the initial condition

$$\rho_S(0) = |1\rangle\langle 1|, \quad \text{or} \quad \rho_S(0) = |2\rangle\langle 2| \tag{A1}$$

it can easily be shown that

$$\langle S^{\pm} \rangle_0 = \mathrm{Tr}_{SR}\big[S^{\pm} \rho_S(0) \big] = 0 \tag{A2}$$

$$\mathrm{Tr}_S\big[S^{\pm} L_{SR}^{2l} \rho_S(0) \big] = 0, \qquad l = 0, 1, 2, \ldots \tag{A3}$$

$$\begin{aligned} \mathrm{Tr}_R\big[L_{SR}^{2l+1} \rho_R(0) \big] &= \mathrm{Tr}_R\big[L_{SR}^{2l+1} |\{0\}\rangle\langle\{0\}| \big], \qquad l = 0, 1, 2, \ldots \\ &= 0 \end{aligned} \tag{A4}$$

From this and the formal solution of the Liouville (von Neumann) equation:

$$\rho(t) = e^{-itL_0} \mathscr{T} \exp\left(-i \int_0^t dt'\, e^{it'L_0} L_{SR} e^{-it'L_0} \right) \rho_S(0) \otimes \rho_R(0) \tag{A5}$$

it follows that

$$\langle S^{\pm} \rangle_t = 0 \tag{A6}$$

REFERENCES

1. M. Sargent III, M. O. Scully, and W. E. Lamb, Jr., *Laser Physics* (Addison-Wesley, Reading, Massachusetts, 1974).
2. L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975).
3. G. S. Agarwal, *Springer Tracts in Modern Physics* No. 70 (Springer, Berlin, 1974).
4. P. L. Knight and P. W. Milonni, *Phys. Lett.* **56A**:275 (1976); P. Carrazana and G. Vetri, *Nuovo Cimento* **55B**:191 (1980).
5. V. F. Weisskopf and E. P. Wigner, *Z. Phys.* **63**:54 (1930), **65**:18 (1930).
6. B. Robertson, *Phys. Rev.* **144**:151 (1966).
7. J. Seke, *Phys. Rev. A* **21**:2156 (1980).
8. G. Adam and J. Seke, *Phys. Rev. A* **23**:3118 (1981).
9. N. S. Krylov and V. A. Fock, *Zh. Eksp. Teor. Fiz.* **17**:93 (1947).
10. L. A. Khalfin, *Zh. Eksp. Teor. Fiz.* **33**:1371 (1957) [*Sov. Phys. JETP* **6**:1053 (1958)]; L. Fonda, G. C. Ghirardi, and A. Rimini, *Rep. Prog. Phys.* **41**:587 (1978).