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Multiple-time-scale perturbation theory: Radiative decay of coupled atomic states

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The spontaneous radiative decay to the ground state of an atomic system initially in the higher of two excited states, coupled in a radiationless fashion by an external field, is investigated by the method of multiple-time-scale perturbation theory. The coupled differential equations of motion for the probability amplitudes are solved to second order, while the usual secular behavior of conventional time-dependent perturbation theory is eliminated in terms of two natural time scales: a fast time scale corresponding to the reciprocal of the level transition frequency and a slower time scale associated with the inverse of the radiation linewidth. Expressions are obtained to this order for the energy shifts and decay characteristics of the excited states and the energy distribution of the final-state photons, quantities previously determined by Fourier analysis and contour integration and by the phenomenological approach of Weisskopf and Wigner. A detailed comparison of these various methods of solution includes conditions under which the present results agree to second order with the previous calculations.

I. INTRODUCTION

It often happens that the use of conventional time-dependent perturbation theory in both classical and quantum-mechanical calculations results in the occurrence of terms that diverge at large times. This secular behavior arises, for example, in planetary orbit theory,[1] and in the determination of transition rates in quantum mechanics.[2] Multiple-time-scale perturbation theory is an especially convenient method that can provide uniform expansions in these difficult situations. This form of time-dependent perturbation theory recognizes the existence of different time or frequency scales related by a small coupling parameter λ. The physical time t is extended into a set of independent time variables $(\tau_0=t,\ \tau_1=\lambda t,\ \tau_2=\lambda^2 t,\ldots)$, which represent the distinct time scales in the system. The structure of the equations of motion of many perturbed systems permits the application of a relatively simple version of this technique.[3] In essence, the unspecified functional dependence of these time variables is completely at our disposal and is used to formulate a solution to the equations of motion in terms of an expansion in λ free of secularity.

Certain processes involving the interaction of electromagnetic radiation with atoms have provided specific contexts for the application of the multiple-time-scales expansion. This approach has been employed by Brooks and Scarfone to obtain the transition rate for induced emission by an n-level atom exposed to a classical radiation field; by Lee, Lee, and Chang,[4] and Lee and Lee,[5] to analyze spontaneous radiation processes in arrays of one, two, and N two-level atoms; by Meyer,[6] to investigate the nonlinear scattering of a laser beam from a relativistic two-level atom; and by Wong, Garrison, and Einwohner,[7–9] to study laser excitation of n-level atoms.

In the present paper, we continue the development of multiple-time-scale perturbation theory in atomic physics by treating the spontaneous radiative decay to the ground state of an atom initially in the higher of two excited states radiationlessly coupled by a constant external field. Wangsness[10] has considered this decay in terms of the phenomenological damping theory of Weisskopf and Wigner,[11] including the effects due to spontaneous transition probabilities and the Lamb shift, in order to explore the connection between Stark mixing of levels and the periodic intensity variations found in hydrogen lines.[12] Alternatively, Fontana and Lynch[13] consider a Hamiltonian formulation of this problem and solve the coupled equations for the probability amplitudes nonperturbatively by Fourier analysis and contour integration. The perturbed energy levels of the excited states are determined by the real parts of the poles of the Fourier transforms in the complex

energy plane, while the imaginary parts provide the decay characteristics. This method of solution and that based on multiple time scales automatically lead to exponential decay laws for large times. Weisskopf and Wigner showed that spontaneous atomic radiation can be accounted for by modifying the Schrödinger equation through introduction of damping terms that lead to exponential decay behavior at large times. The use of such damping terms has been found to be in conformity with the results of more sophisticated treatments.[14,15] In the multiple-time-scales analysis the energy shifts and the amplitude changes occur when functional dependences on various time scales are specified so as to eliminate secular terms in each order.[16]

Section II describes the physical system of interest and the corresponding equations of motion for the probability amplitudes, to be solved here to second order by multiple-time-scale perturbation theory. Section III indicates the use of this method in various other areas and carries out the solution process to obtain the probability amplitudes of the system states, the perturbed energies and decay characteristics of the excited states, and the energy distribution of the final-state photons. Finally, in Sec. IV the results will be discussed.

II. THE RADIATIVE DECAY PROBLEM

This paper is concerned with the single-photon radiative decay process that results when the higher of two excited atomic states, radiationlessly coupled by a constant external field V, decays to the ground state. The Hamiltonian of this system consists of three parts, H_0, H_{ar}, and V, where H_0 is the Hamiltonian for the unperturbed atom and the free electromagnetic radiation field, while H_{ar} describes their interaction. In the present case

$$H_{ar}=\sum_{\vec{k},\sigma}\left[\frac{2\pi}{L^3k}\right]^{3/2}[a(\vec{k},\sigma)e^{i\vec{k}\cdot\vec{r}}+a^{\dagger}(\vec{k},\sigma)e^{-i\vec{k}\cdot\vec{r}}]\hat{e}_{\sigma}(\vec{k})\cdot\vec{p}\ , \tag{1}$$

where the sum is over final-state photon momenta $\vec{k}$ and polarizations σ. The operators $a(\vec{k},\sigma)$ and $a^{\dagger}(\vec{k},\sigma)$ are, respectively, the photon annihilation and creation operators for the photon mode $(\vec{k},\sigma)$; $\hat{e}_{\sigma}(\vec{k})$ is a unit polarization vector; and $\vec{p}$ is the momentum of the electron relative to the center of mass of the atom. We take $\hbar=m=c=1$, where m is the mass of the electron.

The unperturbed atom has two excited states $|a\rangle$ and $|b\rangle$ and a ground state $|c\rangle$ with energies $E_a>E_b>E_c$, respectively. The system states of interest are $|0\rangle$, atom in state $|a\rangle$ with no photons present; $|j\rangle$, atom in state $|b\rangle$ and no photons present; and $|f\rangle$, atom in state $|c\rangle$ and one photon present with momentum $\vec{k}$ and polarization σ. Initially, i.e., at $t=0$, the atom is assumed to be in an excited state. At time t later, the system state is expressed in the Schrödinger picture as

$$|t\rangle=c_0(t)|0\rangle+c_j(t)|j\rangle+\sum_f c_f(t)|f\rangle\ , \tag{2}$$

where the summation index f is an abbreviation for the photon mode $(\vec{k},\sigma)$, and the energies E_0, E_j, and E_f are given by E_a, E_b, and E_c+k, respectively. The usual Schrödinger picture equations of motion yield the following set of three coupled differential equations for the state probability amplitudes $c_0(t)$, $c_j(t)$, and $c_f(t)$:

$$\left[\frac{d}{dt}+iE_0\right]c_0(t)+iV_{0j}c_j(t)+i\lambda\sum_f H'_{0f}c_f(t)=0\ , \tag{3}$$

$$\left[\frac{d}{dt}+iE_j\right]c_j(t)+iV_{j0}c_0(t)+i\lambda\sum_f H'_{jf}c_f(t)=0\ , \tag{4}$$

$$\left[\frac{d}{dt}+iE_f\right]c_f(t)+i\lambda H'_{f0}c_0(t)+i\lambda H'_{fj}c_j(t)=0\ , \tag{5}$$

where λ is a smallness parameter explained below and

$$V_{0j}=\langle 0|(H_{ar}+V)|j\rangle=\langle 0|V|j\rangle\ , \tag{6}$$

$$H'_{0f}=\langle 0|(H_{ar}+V)|f\rangle=\langle 0|H_{ar}|f\rangle\ , \tag{7}$$

$$H'_{jf}=\langle j|(H_{ar}+V)|f\rangle=\langle j|H_{ar}|f\rangle\ . \tag{8}$$

The matrix elements V_{j0}, H'_{f0}, and H'_{fj} are defined similarly. Equations (3)–(5) are the equations to be solved by the method of multiple time scales subject to the general initial conditions

$$c_0(0)\neq 0,\quad c_j(0)\neq 0,\quad c_f(0)=0\ . \tag{9}$$

For problems like the one at hand, the smallness

parameter λ is the strength of the atom-radiation field interaction. In Eqs. (3)—(5) the matrix elements H'_{mn} have been written as $H'_{mn}\rightarrow\lambda H'_{mn}$, where λ is set equal to unity at the end of the calculation. Its purpose is to keep track of the different orders in the perturbation expansion. The natural time scales for radiative processes in atomic systems are the inverse of the level transition frequency and the inverse of the radiation linewidth.[4] These times are of order λ^0 and λ^2, respectively; this assures us that introducing time scales ordered by powers of λ will correspond to the physical behavior of the system. On the other hand, the *a priori* identification and introduction of these two time scales is not obligatory. The mixing frequency between the states $|0\rangle$ and $|j\rangle$ is determined by the strength of the external perturbation V rather than being scaled according to the parameter λ. The time behavior corresponding to this frequency is incorporated in the solution in a straightforward manner.

III. MULTIPLE-TIME-SCALES EXPANSION

Multiple-time-scale perturbation theory is an adaptation of techniques developed by Krylov and Bogoliubov,[17] and Bogoliubov and Mitropolsky[18] for the avoidance of secular behavior in the treatment of quasiperiodic classical systems. The version of this formalism to be used here was devised by Frieman[19] and Sandri[20] in the treatment of problems in the kinetic theory of gases and plasmas. The similar two-variable method used by Meyer[6] was introduced by Cole and Kevorkian.[21]

Multiple-time-scale methods are generally well known and extensively employed in engineering and applied mathematics.[3] Applications in the physics literature are less common, but the range of topics extends beyond the atomic and statistical physics examples cited above. In quantum theory, Montgomery and Ruijgrok[22] analyzed transitions of a spin-$\frac{1}{2}$ particle in crossed, time-dependent magnetic fields, and Balasubramanian[23] extended the treatment to the spin-1 particle case; Boldt and Sandri[24] obtained an exponential decay law in the Weisskopf-Wigner model; Varga and Aks[25] generated a first-order renormalized Hamiltonian in the process of solving the weakly nonlinear ϕ^4 model of quantum field theory; and Khoo and Wang[26] use multiple-time-scales analysis as a nondiagrammatic procedure for obtaining the frequency shift and decay constants for phonons in an anharmonic crystal. In the area of statistical mechanics, Cukier and Deutsch[27] extracted from the Liouville equation for a Brownian system the Fokker-Planck equation for the distribution function of the Brownian particle.

In these examples, the physical systems inherently contain a smallness parameter and a number of natural time scales which can be distinguished or labeled in some way as a function of the parameter. For applications in quantum and statistical physics, it has been possible to order the times simply according to powers of the small parameter, although the general multiple-time-scales method does not require this.[3,25] Analysis by conventional time-dependent perturbation theory yields expansions whose uniformity is guaranteed only if the time is restricted to sufficiently small values. For greater values of the time, higher-order terms can become nonuniformly large by relative growth in proportion to time or due to the effect of nearly resonant denominators (quasisecular behavior). This expansion breakdown is caused by the cumulative effect of contributions by physical processes occurring at the higher-order "slow" time scales.[25] In the multiple-time-scales approach, the different time scales are identified and separately considered in the process of assuring uniformity order by order.[4]

Proceeding with the solution of the present case, we begin by solving Eq. (4) for $c_0(t)$ and find

$$c_0(t)=\frac{i}{V_{j0}}\left[\frac{d}{dt}+iE_j\right]c_j(t)-\frac{\lambda}{V_{j0}}\sum_f H'_j\,c_f(t)\ . \tag{10}$$

Equations (3) and (5) then become, after substitution of Eq. (10) and rearrangement,

$$\left[\frac{d^2}{dt^2}+i(E_0+E_j)\frac{d}{dt}+(|V|^2-E_0E_j)\right]c_j(t)+i\lambda\sum_f\left[H'_{jf}\left[\frac{d}{dt}+iE_0\right]-iV_{j0}H'_{0f}\right]c_f(t)=0\ , \tag{11}$$

and

$$\left[\frac{d}{dt}+iE_f\right]c_f(t)+i\lambda\left[\frac{iH'_{f0}}{V_{j0}}\left[\frac{d}{dt}+iE_j\right]+H'_{fj}\right]c_j(t)-i\lambda^2\frac{H'_{f0}}{V_{j0}}\sum_g H'_{jg}C_g(t)=0\ , \tag{12}$$

respectively, where $|V|^2=V_{0j}V_{j0}$ and the summation index g in Eq. (12) is over final states.

To introduce the multiple-time-scales formalism, we let $c_n(t)\rightarrow c_n(\tau)$ where τ represents collectively the set of time variables $\tau_0,\tau_1,\tau_2,\ldots$, which can be put into correspondence with the physical time t as indicated previously. Associated with this extension to a multicomponent variable is the derivative expansion

$$\frac{d}{dt}\rightarrow\frac{\partial}{\partial\tau_0}+\lambda\frac{\partial}{\partial\tau_1}+\lambda^2\frac{\partial}{\partial\tau_2}+\cdots$$
$$=D_0+\lambda D_1+\lambda^2 D_2+\cdots\ , \tag{13}$$

where a change of notation has been made for compactness. Briefly, the method as applied to Eqs. (11) and (12) involves replacing t, $c_n(t)$, and the time derivative by τ, $c_n(\tau)$, and the right-hand side of Eq. (13), respectively. We then seek a solution of the resulting equations to various orders by expanding $c_n(\tau)$ in the form

$$c_n(\tau)=c_n^{(0)}(\tau)+\lambda c_n^{(1)}(\tau)+\lambda^2 c_n^{(2)}(\tau)+\cdots\ , \tag{14}$$

according to the initial conditions. In the course of carrying out the solution, the $\tau_0,\tau_1,\tau_2,\ldots$ are to be treated as independent variables, and, when otherwise unspecified, the functional dependence on these variables can be chosen arbitrarily for convenience or, more importantly, can be chosen so as to forestall behavior leading to nonuniformity. When results to the desired order are secured in terms of $\tau_0,\tau_1,\tau_2,\ldots$, they are converted to the physical time t by setting $\tau_0=t$, $\tau_1=\lambda t$, $\tau_2=\lambda^2 t,\ldots$, which defines the so-called "physical line."

The zeroth-order equations are found to be

$$[D_0+i(\beta+\alpha)][D_0+i(\beta-\alpha)]c_j^{(0)}(\tau)=0\ , \tag{15}$$

$$(D_0+iE_f)c_f^{(0)}(\tau)=0\ , \tag{16}$$

where α and β are defined by

$$\alpha\equiv\{|V|^2+[\tfrac{1}{2}(E_0-E_j)]^2\}^{1/2}\ , \tag{17}$$

$$\beta\equiv\tfrac{1}{2}(E_0+E_j)\ . \tag{18}$$

The solutions in this case are

$$c_j^{(0)}(\tau)=A_+(\tau_1,\tau_2,\ldots)\exp[-i(\beta+\alpha)\tau_0]+A_-(\tau_1,\tau_2,\ldots)\exp[-i(\beta-\alpha)\tau_0]\ , \tag{19}$$

$$c_f^{(0)}(\tau)=c_f^{(0)}(\tau_0=0,\tau_1,\tau_2,\ldots)e^{-iE_f\tau_0}=0\ . \tag{20}$$

In Eq. (20) we have chosen the zero solution because we are assuming the system to be in an excited atomic state initially. The coefficients $A_\pm$ in Eq. (19) are independent of the time scale τ_0 but do possess as-yet-unspecified dependence on the higher-order time scales.

With the above results, the first-order equation for c_j is given by

$$\{[D_0+i(\beta+\alpha)][D_0+i(\beta-\alpha)]\}c_j^{(1)}(\tau)+[2D_0D_1+i(E_0+E_j)D_1]c_j^{(0)}(\tau)=0\ . \tag{21}$$

When Eq. (21) is integrated with respect to τ_0, the second term will cause the appearance of secular behavior in $c_j^{(1)}(\tau)$. To avoid this breakdown in the perturbation expansion, we take $D_1A_\pm(\tau_1,\tau_2,\ldots)=0$ so that $A_\pm$ is not only independent of τ_0, but additionally independent of τ_1. The ability to specify functional dependence in this way is the key feature of the multiple-time-scales solution procedure. Under these conditions, Eq. (21) becomes

$$[D_0+i(\beta+\alpha)][D_0+i(\beta-\alpha)]c_j^{(1)}(\tau)=0\ . \tag{22}$$

Here we take the zero solution $c_j^{(1)}(\tau)=0$ because we elect to express the initial conditions in the zeroth-order approximation.

Turning to the first-order equation for c_f, we find, again using the above results, the expression

$$(D_0+iE_f)c_f^{(1)}(\tau)+i\left[\frac{iH'_{f0}}{V_{j0}}(D_0+iE_j)+H'_{fj}\right]c_j^{(0)}(\tau)=0\ , \tag{23}$$

from which it follows that

$$c_f^{(1)}(\tau)=B_{f+}(\tau_2,\cdots)e^{-iE_f\tau_0}\int_0^{\tau_0}\exp\{i[E_f-(\beta+\alpha)]x\}dx$$
$$+B_{f-}(\tau_2,\cdots)e^{-iE_f\tau_0}\int_0^{\tau_0}\exp\{i[E_f-(\beta-\alpha)]x\}dx\ , \tag{24}$$

where the quantities $B_{f\pm}$ are defined by

$$B_{f\pm}(\tau_2,\ldots)\equiv i\left[\frac{H'_{f0}}{V_{j0}}[E_j-(\beta\pm\alpha)]-H'_{fj}\right]A_{\pm}(\tau_2,\ldots) \tag{25}$$

and we have taken $c_f^{(1)}(\tau_0=0,\tau_1,\ldots)=0$ in agreement with the initial conditions. For simplicity, we consider the limit of large τ_0 and make the following approximation[28] in the expression for $c_f^{(1)}$:

$$\lim_{\tau_0\to\infty}\int_0^{\tau_0}\exp\{i[E_f-(\beta\pm\alpha)]x\}dx=\int_0^{\infty}\exp\{i[E_f-(\beta\pm\alpha)]x\}dx\equiv I_{f\pm}\ , \tag{26}$$

where

$$I_{f\pm}\equiv i\left[\mathscr{P}\frac{1}{(\beta\pm\alpha)-E_f}-i\pi\delta((\beta\pm\alpha)-E_f)\right]\ , \tag{27}$$

and $\mathscr{P}$ implies the Cauchy principal value at $E_f=(\beta\pm\alpha)$. Therefore, for large τ_0,

$$c_f^{(1)}(\tau)=B_{f+}(\tau_2,\ldots)I_{f+}\exp[-i(\beta+\alpha)\tau_0]+B_{f-}(\tau_2,\ldots)I_{f-}\exp[-i(\beta-\alpha)\tau_0]\ . \tag{28}$$

In making this approximation, we average out behavior occurring at small values of τ_0. On the physical line, however, τ_0 turns out to measure times on the order of the reciprocal of the energy difference between the excited states and the ground state. We can thus allow τ_0 to be "large" without interfering with investigation of decay processes, which occur on a much slower time scale.

The second-order equation for c_j is, after dropping terms known to be equal to zero,

$$\{[D_0+i(\beta+\alpha)][D_0+i(\beta-\alpha)]\}c_j^{(2)}(\tau)+D_2[2D_0+i(E_0+E_j)]c_j^{(0)}(\tau)$$
$$+i\sum_f[H'_{jf}(D_0+iE_0)-iV_{j0}H'_{0f}]c_f^{(1)}(\tau)=0\ . \tag{29}$$

The second and third terms in Eq. (29) will contribute secular behavior to $c_j^{(2)}$, so, once again exploiting the freedom to dictate higher-order τ dependence, we require

$$D_2[2D_0+2(E_0+E_j)]c_j^{(0)}(\tau)+i\sum_f[H'_{jf}(D_0+iE_0)-iV_{j0}H'_{0f}]c_f^{(1)}(\tau)=0\ . \tag{30}$$

Using Eqs. (19) and (28) in Eq. (30) and equating coefficients of like exponentials, we have, for example,

$$D_2A_+(\tau_2,\ldots)=\frac{-1}{2\alpha}\sum_f\{H'_{jf}[E_0-(\beta+\alpha)]+V_{j0}H'_{0f}\}$$
$$\times\left[\frac{H'_{f0}}{V_{j0}}[E_j-(\beta+\alpha)]-H'_{fj}\right]I_{f+}A_+(\tau_2,\ldots)\ . \tag{31}$$

To simplify the calculation we assume that the upper and lower excited atomic states, $|a\rangle$ and $|b\rangle$, respectively, have definite values of angular momentum.[29] Under these conditions, the cross terms in Eq. (31) vanish and the resulting equation has the solution

$$A_+(\tau_2,\ldots)=A_+(\tau_2=0,\tau_3,\ldots)\exp\left[-\left[\frac{1}{2\alpha}[(\beta+\alpha)-E_0]G_{j+}+\frac{1}{2\alpha}[(\beta+\alpha)-E_j]G_{0+}\right]\tau_2\right]$$
$$\equiv A_+(\tau_2=0,\tau_3,\ldots)e^{-X_+\tau_2}\ , \tag{32}$$

where

$$G_{0\pm}\equiv\sum_f |H'_{0f}|^2 I_{f\pm}\ , \tag{33}$$

$$G_{j\pm}\equiv\sum_f |H'_{jf}|^2 I_{f\pm}\ . \tag{34}$$

Similarly,

$$A_-(\tau_2,\ldots)=A_-(\tau_2=0,\tau_3,\ldots)\exp\left[-\left[\frac{1}{2\alpha}[E_j-(\beta-\alpha)]G_{0-}+\frac{1}{2\alpha}[E_0-(\beta-\alpha)]G_{j-}\right]\tau_2\right]$$

$$\equiv A_-(\tau_2=0,\tau_3,\ldots)e^{-X_-\tau_2}\ . \tag{35}$$

The exponential quantities $X_\pm$ are complex numbers whose real parts are positive and correspond to exponential decays and whose imaginary parts correspond to energy shifts for the perturbed excited states. After elimination of secular behavior, Eq. (29) becomes

$$[D_0+i(\beta+\alpha)][D_0+i(\beta-\alpha)]c_j^{(2)}(\tau)=0\ . \tag{36}$$

As in the first-order case, we choose the zero solution $c_j^{(2)}(\tau)=0$.

Consider now the second-order equation for $c_f(\tau)$. Making use of the results obtained above for the τ dependence of the lower-order terms, there remains the equation $(D_0+iE_f)c_f^{(2)}(\tau)=0$, which has the solution

$$c_f^{(2)}(\tau)=c_f^{(2)}(\tau_0=0,\tau_1,\cdots)e^{-iE_f\tau_0}$$

$$=0\ , \tag{37}$$

again subject to the initial conditions.

By carrying out the multiple-time-scale expansion we have seen how the second-order solution may be attained without encountering the usual secular behavior in conventional time-dependent perturbation theory. As far as the calculation is concerned all τ_k's, for $k\geq 3$, may be set equal to zero. Only the time scales τ_0 and τ_2 are involved in the description of the probability amplitudes. In the higher orders, the calculation becomes very unwieldy, and little additional information about the system is revealed.[3] Thus we terminate the expansion process and present our second-order results.

For $c_j(\tau)$, we have

$$c_j(\tau)=c_j^{(0)}(\tau)+\lambda c_j^{(1)}(\tau)+\lambda^2 c_j^{(2)}(\tau)+\cdots$$

$$=c_j^{(0)}(\tau)+0+0+\cdots\ . \tag{38}$$

From Eqs. (19), (32), (35), and (38), the amplitude $c_j(\tau)$ becomes

$$c_j(\tau)=A_+(\tau_2=0,\tau_3,\ldots)\exp[-X_+\tau_2-i(\beta+\alpha)\tau_0]+A_-(\tau_2=0,\tau_3,\ldots)\exp[-X_-\tau_2-i(\beta-\alpha)\tau_0]\ . \tag{39}$$

Similarly,

$$c_f(\tau)\simeq\lambda c_f^{(1)}(\tau)=\lambda B_{f+}(\tau_2=0,\tau_3,\cdots)I_{f+}\exp[-X_+\tau_2-i(\beta+\alpha)\tau_0]$$

$$+\lambda B_{f-}(\tau_2=0,\tau_3,\cdots)I_{f-}\exp[-X_-\tau_2-i(\beta-\alpha)\tau_0]\ . \tag{40}$$

To express our results in terms of the fundamental time variable t, it is necessary to invoke the "physical line" condition. In this way we recover the original amplitudes $c_j(t)$ and $c_f(t)$. Thus suppressing the higher-order τ dependence and letting λ go to unity, we arrive at

$$c_j(\tau)=A_+e^{-Z_+t}+A_-e^{-Z_-t}\ , \tag{41}$$

and

$$c_f(t)=B_{f+}I_{f+}e^{-Z_+t}+B_{f-}I_{f-}e^{-Z_-t}\ , \tag{42}$$

where $Z_{\pm}$ is defined by $i(\beta\pm\alpha)+X_{\pm}$. To obtain an expression for $c_0(t)$, we substitute these forms for $c_j(t)$ and $c_f(t)$ into Eq. (10), which becomes

$$c_0(t)=\frac{i}{V_{j0}}(-Z_+ +iE_j+G_{j+})A_+e^{-Z_+t}+\frac{i}{V_{j0}}(-Z_- +iE_j+G_{j-})A_-e^{-Z_-t}\ . \tag{43}$$

Our expressions for c_f so far have been intermediate ones used in computing the up-to-second-order results for c_j and c_0. We now substitute Eqs. (41) and (42) into Eq. (5), which then readily yields

$$c_f(t)=\frac{-iA_+}{iE_f-Z_+}\left[\frac{iH'_{f0}}{V_{j0}}(-Z_+ +E_j+G_{j+})+H'_{fj}\right](e^{-Z_+t}-e^{-iE_ft})$$
$$+\frac{-iA_-}{iE_f-Z_-}\left[\frac{iH'_{f0}}{V_{j0}}(-Z_- +E_j+G_{j-})+H'_{fj}\right](e^{-Z_-t}-e^{-iF_ft})\ . \tag{44}$$

Finally, we evaluate the constants $A_{\pm}$ in terms of the initial conditions. If we take the atom at $t=0$ to be in the upper excited state $|a\rangle$, then the initial conditions are $c_0(0)=1$, $c_j(0)=c_f(0)=0$. Hence we have

$$c_0(t)=\frac{(-Z_- +iE_j+G_{j-})e^{-Z_-t}-(-Z_+ +iE_j+G_{j+})e^{-Z_+t}}{Z_+-Z_-+G_{j-}-G_{j+}}\ , \tag{45}$$

$$c_j(t)=\frac{iV_{j0}e^{-Z_+t}-iV_{j0}e^{-Z_-t}}{Z_+-Z_-+G_{j-}-G_{j+}}\ , \tag{46}$$

$$c_f(t)=\frac{V_{j0}\left[\frac{iH'_{f0}}{V_{j0}}(-Z_+ +iE_j+G_{j+})+H'_{fj}\right](e^{-Z_+t}-e^{-iE_ft})}{(iE_f-Z_+)(Z_+-Z_-+G_{j-}-G_{j+})}$$
$$+\frac{(-V_{j0})\left[\frac{iH'_{f0}}{V_{j0}}(-Z_- +iE_j+G_{j-})+H'_{fj}\right](e^{-Z_-t}-e^{-iE_ft})}{(iE_f-Z_-)(Z_+-Z_-+G_{j-}-G_{j+})}\ . \tag{47}$$

In the above formulas for the c amplitudes, we note energy shifts given by

$$\mathrm{Im}Z_{\pm}-(\beta\pm\alpha)=\frac{1}{2\alpha}\left[\pm[(\beta\pm\alpha)-E_j]\sum_f|H'_{0f}|^2\mathscr{P}\frac{1}{\beta\pm\alpha-E_f}\right.$$
$$\left.\pm[(\beta\pm\alpha)-E_0]\sum_f|H'_{jf}|^2\mathscr{P}\frac{1}{\beta\pm\alpha-E_f}\right] \tag{48}$$

and exponential decay behavior characterized by the constants

$$\mathrm{Re}Z_{\pm}=\frac{1}{2\alpha}\left[\pm[(\beta\pm\alpha)-E_j]\sum_f|H'_{0f}|^2\pi\delta((\beta\pm\alpha)-E_f)\ \pm[(\beta\pm\alpha)-E_0]\sum_f|H'_{jf}|^2\pi\delta((\beta\pm\alpha)-E_f)\right]\ . \tag{49}$$

It is evident that the final-state amplitude $c_f(t)$ peaks at $E_f=\mathrm{Im}Z_{\pm}$. This feature is expected for a system of two coupled decaying states. The exponential decay shown in Eqs. (45)–(47) is a natural consequence of the foregoing multiple-time-scales procedure of systematically removing secular terms in each order of the expansion. By contrast, the damping-term approach of Weisskopf and Wigner introduces this mathematical form of decay into the original coupled differential equation of motion.

IV. DISCUSSION

We have presented a calculation of the decay of two coupled atomic levels, using multiple-time-scale perturbation theory. The differential equations of motion for the probability amplitudes have been solved up to second order, while eliminating the secular terms characteristic of conventional time-dependent perturbation theory. The physical consequences of the mixing of levels produced by the coupling field are contained in the state probabilities, obtained by multiplying Eqs. (45)–(47) by their respective complex conjugates. We have

$$|c_0(t)|^2=\frac{1}{|M_+-M_-|^2}\{|N_+|^2e^{-2\mathrm{Re}Z_+t}+|N_-|^2e^{-2\mathrm{Re}Z_-t} -2|N_+N_-^*|\exp[-(\mathrm{Re}Z_++\mathrm{Re}Z_-)t]\cos[(\mathrm{Im}Z_+-\mathrm{Im}Z_-)t+\phi]\}\ , \tag{50}$$

$$|c_j(t)|^2=\frac{2|V|^2}{|M_+-M_-|^2}\exp[-(\mathrm{Re}Z_++\mathrm{Re}Z_-)t][\cosh(\mathrm{Re}Z_+-\mathrm{Re}Z_-)t-\cos(\mathrm{Im}Z_+-\mathrm{Im}Z_-)t]\ , \tag{51}$$

$$\begin{aligned}|c_f(t)|^2=&\frac{1}{|D|^2}\{|R_+|^2[1+e^{-2\mathrm{Re}Z_+t}-2e^{-\mathrm{Re}Z_+t}\cos(E_f-\mathrm{Im}Z_+)t] \\ &+|R_-|^2[1+e^{-2\mathrm{Re}Z_-t}-2e^{-\mathrm{Re}Z_-t}\cos(E_f-\mathrm{Im}Z_-)t]\} \\ &+\frac{2|R_+R_-^*|}{|D|^2}\{\cos\theta+\exp[-(\mathrm{Re}Z_++\mathrm{Re}Z_-)t]\cos[(\mathrm{Im}Z_--\mathrm{Im}Z_+)t+\theta] \\ &+e^{-\mathrm{Re}Z_+t}\cos[(E_f-\mathrm{Im}Z_+)t+\theta]-e^{-\mathrm{Re}Z_-t}\cos[(E_f-\mathrm{Im}Z_-)t-\theta]\}\ ,\end{aligned} \tag{52}$$

where

$$\begin{aligned}&M_\pm\equiv Z_\pm-G_{j\pm},\quad D\equiv M_+-M_-\ , \\ &N_\pm\equiv iE_j-M_\pm,\quad N_+N_-^*\equiv|N_+N_-^*|e^{i\phi}\ , \\ &R_\pm\equiv\frac{iH'_{f0}N_\pm+H'_{fj}V_{j0}}{(iE_f-Z_\pm)}\ , \\ &R_+R_-^*\equiv|R_+R_-^*|e^{i\theta}\ .\end{aligned} \tag{53}$$

The probabilities of the excited states $|0>$ and $|j>$ each exhibit two exponentially decaying terms, with decay constants $2\mathrm{Re}Z_+$ and $2\mathrm{Re}Z_-$, and a sinusoidally modulated exponential with decay constant $\mathrm{Re}Z_++\mathrm{Re}Z_-$. The expressions are formally the same as the Fourier-transform results of Fontana and Lynch, and the results of the Weisskopf-Wigner procedure as applied by Wangsness. Comparison in detail will be provided below. Here we reemphasize the fact that the exponential decays are a natural outcome of the multiple-time-scale perturbation theory solution to this problem.

The energy distribution of the final-state photons is described by the expression

$$\lim_{t\to\infty}|c_f(t)|^2=\frac{1}{|D|^2}(|R_+|^2+|R_-|^2+2|R_+R_-^*|\cos\theta)\ . \tag{54}$$

Recalling

$$|R_\pm|^2\sim|iE_f-Z_\pm|^{-2}=|i(E_f-\mathrm{Im}Z_\pm)-\mathrm{Re}Z_\pm|^{-2}\ , \tag{55}$$

we see that, in formal agreement with Ref. 13, the final-state energy has two peaks, one at $E_f=\mathrm{Im}Z_+$ and the other at $E_f=\mathrm{Im}Z_-$, with widths $2\mathrm{Re}Z_+$ and $2\mathrm{Re}Z_-$, respectively. The quantities $\mathrm{Im}Z_+$ and $\mathrm{Im}Z_-$ are the perturbed energies [incorporating the energy shifts given in Eq. (48)] of the upper and

lower excited atomic levels, and $2\,\mathrm{Re}Z_+$ and $2\,\mathrm{Re}Z_-$ are decay constants associated with those levels.

We see further effects of the mixing of levels in that $|c_0(t)|^2$ and $|c_j(t)|^2$ both contain exponential behavior involving $\mathrm{Re}Z_+$ and $\mathrm{Re}Z_-$ and that these quantities each have contributions from the uncoupled-case transition matrix elements H'_{0f} and H'_{jf}. In particular, $\mathrm{Re}Z_+$ has terms proportional to $\mathrm{Re}G_{0+}$ and $\mathrm{Re}G_{j+}$, while the corresponding terms in $\mathrm{Re}Z_-$ are proportional to $\mathrm{Re}G_{0-}$ and $\mathrm{Re}G_{j-}$, where

$$\mathrm{Re}G_{0\pm}\equiv\sum_f |H'_{0f}|^2\pi\delta(\beta\pm\alpha-E_f)\,,$$
$$\mathrm{Re}G_{j\pm}\equiv\sum_f |H'_{jf}|^2\pi\delta(\beta\pm\alpha-E_f)\,. \tag{56}$$

The quantities $\mathrm{Re}G_{0\pm}$ and $\mathrm{Re}G_{j\pm}$ each represent the splitting into two distinct contributions of the uncoupled-level decay constants $\frac{1}{2}\gamma_0$ and $\frac{1}{2}\gamma_j$, respectively, where

$$\tfrac{1}{2}\gamma_n=\sum_f |H'_{nf}|^2\pi\delta(E_n-E_f),\quad n=0,j\,. \tag{57}$$

Under the conditions that

$$\mathrm{Re}G_{0\pm}\to\tfrac{1}{2}\gamma_0,\quad \mathrm{Re}G_{j\pm}\to\tfrac{1}{2}\gamma_j\,, \tag{58}$$

we find that our results for the amplitudes and probabilities agree to second order with those of Fontana and Lynch, and with the excited-state amplitudes and probabilities of Wangsness. In the Weisskopf-Wigner method employed by the latter author the final states are eliminated by the inclusion of damping terms at the outset. The quantities $G_{0\pm}$ and $G_{j\pm}$ do not appear because, in contrast with the multiple-time-scale approach, two exponential decay constants γ_0 and γ_j are specified at the beginning in the damping terms. Fontana and Lynch introduce the two constants γ_0 and γ_j in the process of fixing the values of a function $\gamma_{nm}(E)$, where E is an energy parameter, appearing in the denominator of the arguments of the inverse transform integrals. They choose to specify two values of E whose pole contributions provide exponential behavior involving γ_0 and γ_j.

It should be pointed out that the conditions given in Eq. (58) for the up-to-second-order agreement described above are plausible in transitions among atomic states where the excited states are closely spaced in energy compared with energy difference between the excited states and the final state, such as in the case for the $2S_{1/2}$, $2P_{1/2}$, and $1S_{1/2}$ levels of hydrogen. Then, given modest values of the perturbation V, the value of

$$\tfrac{1}{2}\gamma_n(E)\equiv\sum_f |H'_{nf}|\pi\delta(E-E_f),\quad n=0,j \tag{59}$$

is relatively insensitive to E, so that Eq. (58) is acceptable. On the other hand, in the case of transitions among high Rydberg states, the distinctions among the four quantities $G_{0\pm}$ and $G_{j\pm}$ generated by multiple-time-scale perturbation theory may have to be taken into account.

Finally, in taking the limit $|V_{0j}|\to 0$ of Eqs. (50)–(52) we find that states $|0>$ and $|j>$ independently decay exponentially to the ground state with decay constants γ_0 and γ_j, respectively. The quantities γ_0 and γ_j appear as a natural consequence of the limiting process, and not through the application of Eq. (58). The limiting results agree with multiple-time-scale perturbation theory calculations of the decay of uncoupled atomic levels by Brooks and Scarfone, who treated stimulated emission, and by Lee, Lee, and Chang, who subsequently considered spontaneous decay. For the excited states, these results are in turn the same as those generated by the damping terms of Weisskopf and Wigner.

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