Spontaneous radiative decay of a continuum

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We obtain and discuss a closed-form expression for the time-dependent density matrix associated with spontaneous radiative decay from a continuum or a set of discrete states to a lower discrete state.

I. INTRODUCTION

An interesting aspect of atoms undergoing laser-enhanced autoionization is the spontaneous decay of the continuum back to a discrete level. This process is the analog of the spontaneous decay of a two-level atom initially prepared in the excited level, where the excited level is replaced by a continuum of levels all of which decay simultaneously to the lowest discrete state. As is shown below, there is a closed expression for the time-dependent density matrix of this system, given any dipole-strength distribution of the continuum and its density-matrix elements at $t=0$. This result amounts to the calculation of the Einstein A coefficient corresponding to a system of one discrete level and one continuum. In the proper limit the expression applies to an N-level atom where all excited, initially populated $N-1$ levels decay spontaneously to the ground state. The Einstein A coefficient is recovered for the particular $N=2$ case.

The spontaneous decay of a single level which can either ionize or photo-deexcite has been considered previously[1] in the framework of the K-matrix formalism. In the terminology of the present work, this configuration corresponds to the special case of a factorizable initial density matrix. By comparison, the expression described below [Eqs. (8) and (9)] applies for an arbitrary initial density matrix. Note also that the spontaneous decay of a continuum back to a bound state enters in the more general problem of laser-enhanced autoionization *with* spontaneous decay (recycling), which received lately much attention.[2,3] The simplicity of the present general solution can be appreciated when compared to the rather complex expressions pertaining to the autoionization problem.[2,3] Another aspect of our result is its applicability to a system of N discrete levels, all decaying back to a common lowest state. This configuration underlies the principle of beam-foil spectroscopy,[4] where the initial preparation of a group of excited states is achieved by sending an atomic beam through a thin foil, followed by monitoring the beats of the time-dependent decaying populations.

We discuss the content of the general expression for two examples: for a nonflat continuum with a Fano dipole-strength distribution[5,6] and for a three-level atom, where the population initially is in the first excited state. For the first (continuum) example we find that the time-dependent ground-state population $P_0(t)$ is of the form $P_0(t)=a+be^{-\lambda t}$, where a,b,λ depend on the initial mode of preparation of the continuum and on the dipole-strength distribution. We also point out that in the special case when the initial preparation of the continuum conforms with the continuum dipole-strength distribution, the resulting $P_0(t)$ acquires a time dependence more complicated than a simple exponent.

The three-level-atom resonance fluorescence spectrum reveals a Fano zero.[5] This comes about since the second excited state draws some of the population of the first excited state at the early stages of the process, before energy conservation is operative. At later times *both* levels decay giving rise to total destructive interference (Fano zero) at the energy of the second excited state. In the limit of near degeneracy of the two excited states, the spectrum attains an interesting "hole-burning" shape, the hole being the Fano zero.

The paper is organized as follows. The central result is derived in Sec. II. Section III is devoted to the discussion of the general expression for some representative examples. In Sec. III A we examine the density matrix in the case of a continuum with a Fano profile and two limiting modes of initial preparation. In Sec. III B we examine the spontaneous decay of the simplest nontrivial example of a three-level atom. A summary of the main results is the content of Sec. IV.

II. THE TIME-DEPENDENT DENSITY MATRIX

The central result of this paper is discussed in the context of a simple model shown schematically in Fig. 1. The model space consists of a ground state $|0\rangle$ and a continuum $|\omega\rangle$, coupled by a quantized radiation field in the dipole and rotating-wave approximation:[2]

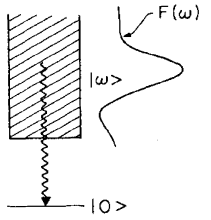

FIG. 1. Schematic display of the model space, ($|0\rangle$, $|\omega\rangle$), the continuum dipole-strength distribution $F(\omega)$ and spontaneous radiative decay of the continuum.

$$\hat{H}=E_0|0\rangle\langle 0|+\int d\omega\,\hbar\omega\hat{C}(\omega,\omega)+\sum_\lambda\hbar\omega_\lambda\hat{a}_\lambda^\dagger\hat{a}_\lambda + \sum_\lambda\int d\omega[\hbar\Omega_\lambda^*(\omega)\hat{B}^\dagger(\omega)\hat{a}_\lambda+\text{H.c.}]\,, \tag{1a}$$

where

$$\hat{B}(\omega)=|0\rangle\langle\omega|,\quad \hat{C}(\omega,\omega')=|\omega\rangle\langle\omega'|\,. \tag{1b}$$

For convenience the ground-state energy E_0 is set to zero. The label $\lambda=(\vec{k},\vec{\epsilon}_\lambda)$ defines the radiation-field modes by specifying the momentum and polarization, respectively, $\hat{a}_\lambda$ is the destruction operator for a photon in mode λ, and the coupling induced by the radiation field in the dipole approximation[2] is

$$\Omega_\lambda(\omega)=-ig_\lambda F(\omega)\,, \tag{2}$$

where g_λ is the radiation-field form factor[7] $g_\lambda=(2\pi\hbar c^2/\omega_\lambda V)^{1/2}(\vec{\epsilon}_\lambda\cdot\vec{n}_d)$, $\omega=ck$, V is the quantization volume, and $F(\omega)=|e|\omega d_{0,\omega}/\hbar c$ entails the dipole distribution $d_{0,\omega}=\langle 0|r|\omega\rangle$.

The Heisenberg equations of motion pertaining to (1) are obtained in a straightforward manner.[7,8] After eliminating the radiation-field degrees of freedom in the Markov-Born approximation, the resulting equations are *linear* in the atomic operator $\hat{B}(\omega)$, $C(\omega,\omega')$, and $\hat{P}_0=|0\rangle\langle 0|$. In particular, the Laplace-transformed equation of motion for the dipole operator $\hat{B}(\omega)$ is

$$z\hat{B}(\omega_0,z)-\hat{B}(\omega_0,t=0)=-i\omega_0\hat{B}(\omega_0,z)-i\sum_\lambda\hat{z}_\lambda(\omega_0,z)\hat{a}_\lambda^{(v)} -F^*(\omega_0)\int d\omega' R(\omega')\hat{B}(\omega',z)\,. \tag{3a}$$

In (3) $\hat{a}_\lambda^{(v)}$ is the unperturbed photon destruction operator and

$$R(\omega)=F(\omega)Q(\omega) =F(\omega)\left[\sum_\lambda ig_\lambda^2\left[\frac{P}{\omega-\omega_\lambda}-i\pi\delta(\omega-\omega_\lambda)\right]\right] \cong\frac{2}{3}\frac{\hbar\omega}{c}F(\omega) \tag{3b}$$

is the coupling embodying the presence of spontaneous decay. The total $t=0$ density matrix $\hat{\rho}(t=0)$ is given by the usual product form

$$\hat{\rho}(t=0)=(|\phi\rangle\langle\phi|)_R \otimes\left[\int d\omega\,d\omega'\,|\omega\rangle C(\omega',\omega;t=0)\langle\omega'|\right]_A\,, \tag{4}$$

where $|\phi\rangle$ denotes the radiation vacuum state and $C(\omega,\omega';t=0)$ describes the initial preparation of the continuum.

Multiply Eq. (3) from the right by $|\phi\rangle$ and noting that $\hat{a}_\lambda^{(v)}|\phi\rangle=0$ gives a separable equation which is amenable to a solution in closed form:

$$\hat{B}(\omega_0,z)|\phi\rangle=\frac{1}{z+i\omega_0}\left[\hat{B}(\omega_0,t=0)\phi\rangle-\frac{F^*(\omega_0)}{D(z)}\int d\omega'\frac{R(\omega')\hat{B}(\omega',t=0)|\phi\rangle}{z+i\omega'}\right], \tag{5a}$$

where

$$D(z)=1+\int d\omega\frac{F^*(\omega)R(\omega)}{z+i\omega} \tag{5b}$$

and the inverse Laplace transform is

$$\hat{B}(\omega_0,t)|\phi\rangle=\frac{1}{2\pi i}\int_C dz\,e^{zt}\hat{B}(\omega_0,z)|\phi\rangle\,. \tag{5c}$$

Equation (5) is the key result since it allows the complete solution of the problem. To see that note from (1b) that

$$\langle\phi|\hat{C}(\omega,\omega';t)|\phi\rangle=\langle\phi|\hat{B}^\dagger(\omega,t)\hat{B}(\omega',t)|\phi\rangle \tag{6}$$

and the unitarity relation and equation of motion for $\hat{P}_0(t)$ give (after taking the $\langle\phi|\cdots|\phi\rangle$ average)

$$\frac{d}{dt}\hat{P}_0(t)=-\frac{d}{dt}\int d\omega\,\hat{C}(\omega,\omega;t)=-\int d\omega\,d\omega'[F^*(\omega)R(\omega')+F(\omega)R^*(\omega')]\hat{C}(\omega,\omega';t)\,. \tag{7}$$

Combining Eqs. (5) and (6) and introducing the notation

$$C(\omega,\omega';t)=\langle\langle\phi|\hat{C}(\omega,\omega';t)|\phi\rangle\rangle\,,$$

where the second bracket indicates trace with regard to the initial atomic density matrix, we obtain

$$C(\omega_0,\omega_0';t)=\left[\frac{1}{2\pi i}\right]^2\int\int dz_1dz_2e^{(z_1+z_2)t}K_1(\omega_0,\omega_0';z_1z_2)\,, \tag{8a}$$

where

$$K_1(\omega_0,\omega_0';z_1,z_2)=\frac{1}{(z_1-i\omega_0)(z_2+i\omega_0')}\left[C(\omega_0,\omega_0';t=0)-\frac{F(\omega_0)}{D^*(z_1)}\int d\omega'\frac{R^*(\omega')C(\omega',\omega_0';t=0)}{z_1-i\omega'}\right.$$
$$-\frac{F^*(\omega_0')}{D(z_2)}\int d\omega'\frac{R(\omega')C(\omega_0,\omega';t=0)}{z_2+i\omega'}$$
$$\left.+\frac{F(\omega_0)F^*(\omega_0')}{D^*(z_1)D(z_2)}\int d\omega\, d\omega'\frac{R^*(\omega)C(\omega,\omega';t=0')R(\omega')}{(z_1-i\omega)(z_2+i\omega')}\right]. \tag{8b}$$

Furthermore, by inserting (8) into (7) we obtain

$$\frac{d}{dt}P_0(t)=-\left[\frac{1}{2\pi i}\right]^2\int\int dz_1dz_2(z_1+z_2)e^{(z_1+z_2)t}K_2(z_1,z_2)\,, \tag{9a}$$

where

$$K_2(z_1,z_2)=\int d\omega\, K_1(\omega,\omega;z_1,z_2)\,. \tag{9b}$$

Expressions (8) and (9) are the central results of this work. They relate, *quite generally,* the time-dependent density-matrix elements to a given initial condition $C(\omega,\omega';t=0)$ and dipole distribution function $F(\omega)$. The case of N discrete levels is obtained by replacing the ω integrations by summations, or alternatively, by using δ-sharp distributions for $F(\omega)$, $C(\omega',\omega;t=0)$, see Eq. (22) below.

Finally, the spectrum of the fluorescent light $S(\omega)$ is given by[9]

$$S(\omega_\lambda)=\langle\hat{a}_\lambda^\dagger(t=\infty)\hat{a}_\lambda(t=\infty)\rangle$$
$$=g_\lambda^2\int d\omega'd\omega''F^*(\omega')F(\omega'')\int_0^\infty d\tau_1d\tau_2e^{-i\omega_\lambda(\tau_1-\tau_2)}\langle\hat{B}^\dagger(\omega',\tau_1)\hat{B}(\omega'',\tau_2)\rangle$$
$$=\left[\frac{1}{2\pi i}\right]^2\int_0^\infty d\tau_1d\tau_2e^{-i\omega_\lambda(\tau_1-\tau_2)}\int_C dz_1dz_2e^{z_1\tau_1+z_2\tau_2}K_3(z_1,z_2)\,, \tag{10a}$$

where

$$K_3(z_1,z_2)=\int d\omega'd\omega''\Omega_\lambda^*(\omega')\Omega_\lambda(\omega'')K_1(\omega',\omega'';z_1,z_2)\,. \tag{10b}$$

The structure of expression (9) implies that the $P_0(t)$ time dependence is determined by the singularities of $K_2(z_1,z_2)$ (poles, cuts). These in turn can come either from the zeros of $D(z)$, which depend *only* on the dipole-strength distribution $F(\omega)$ [Eq. (5b)], or from the other factors in (8) and (9), which explicitly depend on the initial preparation of the continuum, $C(\omega,\omega';t=0)$. The latter possibility, i.e., obtaining decay rates, which depend on the mode of preparation of the continuum, is novel. As an example in Sec. III indicates, this occurs only for a judicious choice of $C(\omega,\omega';t=0)$ conforming with the given $F(\omega)$. The structure of expression (10) indicates as well that the widths and positions of the peaks in the light spectrum are determined by the singularities of $K_3(z_1,z_2)$.

III. SPONTANEOUS DECAY OF A FANO CONTINUUM AND A THREE-LEVEL ATOM

To demonstrate the content of expressions (9) and (10) we consider two examples of current interest: a continuum with a Fano dipole distribution and a three-level atom, initially prepared in the first excited states. We are interested in the ensuing $P_0(t)$ and the resonance fluorescence spectrum, respectively.

A. The spontaneous decay of a Fano continuum

The Fano dipole-strength distribution[5] has been employed in recent models for laser-enhanced autoionization in atoms.[2,3,6,8] The assumed dipole distribution results from admixing a bound state at energy ω_1 with the continuum of scattering states and can be written in the form[6]

$$F(\omega)=\frac{\Omega_0}{\sqrt{4\pi}\gamma_1}\left[\frac{\gamma_1}{\omega-\omega_1+i\gamma_1}-\frac{1}{1+iq}\frac{\sigma}{\omega-\omega_1+i\sigma}\right]. \tag{11}$$

In (11) γ_1 is the autoionization width, q is the parameter controlling the asymmetry of Fano's profile ($q=\infty$ corresponds to a Lorentzian), σ is a cutoff parameter taken to infinity after the calculation, and the Ω_0 parameter plays the role of a Rabi frequency when $q\gg1$.

The second input into expression (9) is the initial density matrix $C(\omega,\omega';t=0)$. In general $C(\omega,\omega';t=0)$ will have both diagonal and off-diagonal elements depending on the particular physical process used to prepare the continuum. For definiteness consider the following two limits. When the continuum is prepared "coherently," e.g., by populating an autoionizing state $|1\rangle$, the corresponding density matrix has the form

$$C(\omega,\omega';t=0)=A(\omega)A^*(\omega') \tag{12a}$$

with the normalization

$$\int d\omega\,|A(\omega)|^2=1\,. \tag{12b}$$

The normalization (12b) is consistent with the dimensionality $[C(\omega,\omega';t)]=[\omega]^{-1}$ as evident from the unitarity relation (7). For the particular mode of preparation by a radiative transition to level $|1\rangle$ it is suggestive to write for

$$F(\omega)=\sqrt{N(q)}\frac{\Omega_0}{2}A(\omega)\ , \tag{13}$$

where $N(q)$ is dimensionless, inserted to satisfy (12b).

The "maximum coherence" limit (12a) corresponds to the previously studied case[1] of one initially populated level that can ionize or photo-deexcite. The difference in approaches is that in Ref. 1 the electron and photon decays are solved simultaneously. In the present formulation we first solve for the electron decay, which produces the perturbed dipole distribution of the continuum (Fano profile) and then superimpose the photoemission. This sequence simplifies the final expression considerably. Another difference is that below we evaluate $P_0(t)$. In Ref. 1 the complementary populations are considered.

The other extreme is that of maximum incoherence, when the atomic density matrix has only diagonal elements. This limit is modeled for an atom with a dense, yet discrete spectrum (quasicontinuum) by writing $\langle i|\hat{\rho}|j\rangle=\rho_{ii}\delta_{ij}$. In the limit of a true continuum

$$C(\omega,\omega';t=0)=p(\tfrac{1}{2}(\omega+\omega'))\delta(\omega-\omega') \tag{14}$$

and $p(\omega)$ is dimensionless. A possible method of preparing an atom with a density matrix of the type (14) is by pumping the continuum (from the ground state or any other state) with a weak white-noise laser, i.e.,

$$\langle V(\omega,t)V^*(\omega',t)\rangle=2V_0(\omega,\omega')\delta(t-t')$$

and $V(\omega,t)$ is the dipole-transition amplitude. A special case in this class is when

$$|F(\omega)|^2=\frac{\Omega_0^2}{4}\frac{1}{N(q)\pi\gamma_1}p(\omega)\ , \tag{15a}$$

where the prefactors are extracted to make $p(\omega)$ dimensionless. In particular, the $q=\infty$ limit (which for simplicity is the only one to be considered below) is a Lorentzian:

$$p(\omega)=N\frac{\gamma_p^2}{(\omega-\omega_1)^2+\gamma_p^2}\ . \tag{15b}$$

Having specified $F(\omega)$, $C(\omega,\omega';t=0)$ it is a matter of straightforward algebra to evaluate (9). Consider first the "coherent" case, Eqs. (12) and (13), for which all integrals become separable. Neglecting the divergent principal part in $R(\omega)$, Eq. (3b), the spontaneous decay width γ_s is defined by

$$\gamma_s=\frac{\Omega_0^2}{4}Q(\omega_1)\ . \tag{16}$$

This definition gives for the two-level atom $A=2\gamma_s$, where A is the Einstein coefficient. The result for $P_0(t)$ is

$$P_0(t)=\frac{D}{2\gamma}(1-e^{-2\gamma t})\ , \tag{17a}$$

where

$$\gamma=\gamma_1+\frac{\gamma_s}{\mu_3}\mathrm{Re}(\mu_1)\ , \tag{17b}$$

$$\mu_1=-(1+iq)/(1-iq),\quad \mu_2=\frac{\gamma_s}{\gamma_1(1+q^2)}=\mu_3-1\ ,$$

and

$$D=2(-\gamma_1\mathrm{Re}(\mu_1)+\mathrm{Re}\{(\mu_1+i\mu_2)[-i(\gamma_s/\mu_3)\mathrm{Im}(\mu_1)]+\gamma\})/\mu_3^2\ . \tag{17c}$$

Note that the q dependence is quite involved. However, in the limit $q\to\infty$ the expression simplifies considerably:

$$P_0(t)=\frac{\gamma_s}{\gamma_1+\gamma_s}(1-e^{-2(\gamma_1+\gamma_s)t})\ . \tag{18}$$

The decay constant $2(\gamma_1+\gamma_s)$ is plausible and reflects the existence of the scattering channel "γ_1," besides the decay channel "γ_s." For $\gamma_1\to 0$ we recover the two-level result. The prefactor is a plausible branching ratio: For $\gamma_s=0$ no decay channel exists and $P_0(t)=0$ as it should, while when $\gamma_1=0$ (or $\gamma_s\gg\gamma_1$), the two-level result is recovered.

The other limit of completely incoherent preparation of the continuum is discussed for two models of $p(\omega)$, Eq. (14), namely when $p(\omega)$ follows the dipole distribution, Eq. (15a), and when it is much wider than the dipole distribution, i.e., Eq. (15b) with $\gamma_p\gg\gamma_1$.

In this context the double integrals in (9) collapse to single integrals; however, the expressions are quite involved and only the $q\to\infty$ case is presented. For the model (15a) the kernel to be inserted in (9) is

$$K_2(z_1,z_2)=\frac{N}{2}\frac{1}{(z_1+z_2)^2}[y_1(z_1,z_2)+y_2(z_1,z_2)]\ ,$$

$$y_1(z_1,z_2)=-\frac{2\gamma_s(z_1+z_2)}{d(z_1)d^*(z_2)}\ , \tag{19}$$

$$y_2(z_1,z_2)=-\frac{\gamma_1\gamma_s^2}{d(z_1)d^*(z_2)}\times\frac{(2\gamma_1+z_1+z_2)^2}{(z_1+\gamma_1-i\omega_1)^2(z_2+\gamma_1+i\omega_1)^2}\ ,$$

where

$$d(z)=z+\gamma_1+\gamma_s-i\omega_1\ . \tag{20}$$

The interesting feature of (19) is the appearance of double poles, which gives rise to terms of form $t^pe^{-\lambda t}$, $p\geq 1$, implying deviation from the purely exponentials encountered so far. This is an example for the situation mentioned in the Introduction, when some of the singularities of $K_2(z_1,z_2)$, or equivalently the decay rate of $P_0(t)$, depend on the mode of preparing the continuum. The origin of the double-pole terms in (19) can be traced back to integrals of the type $\int d\omega\,|F(\omega)|^4/[(z_1-i\omega)(z_2+i\omega)]$ occurring in (9) when (15a) is inserted. This kind of coalescence is the result of an exact matching of $C(\omega,\omega';t=0)$ with the underlying dipole-strength distribution. Note also that in the $\gamma_1\to 0$ limit we recover the all-familiar two-level atom result $P_0(t)=1-e^{-2\gamma_s t}$ as it

should. For $\gamma_1 \neq 0$ the double poles in (19) give rise to nonstandard line shapes when inserted in Eq. (10).

For a model of "flat" $p(\omega)$, namely, Eq. (15b) with $\gamma_p \gg \gamma_1$, the coalescence of poles in expression (9) particular to (15a) does not occur. The resulting expression is

$$P_0(t)=p_1\left[\frac{\gamma_s}{\gamma_1+\gamma_s}\right]^2(1-e^{-2(\gamma_1+\gamma_s)t}), \tag{21}$$

where $p_1=p(\omega_1)$ is the average constant population of the continuum.

It can be easily checked that in the limit $\gamma_1 \to 0$, i.e., when the Fano profile becomes infinitely narrow, the correct two-level-atom expression is recovered. In the other limit, $\gamma_s \to 0$, no decay to the ground state is possible, hence $P_0(t)=0$. By comparing (18) and (21) we note that while the decay rates in this example do not depend on the mode of preparation, the prefactors determining the *absolute values* of $P_0(t)$ do. This reflects the effects of interferences due to the initial coherences in both modes of preparation.

B. Spontaneous decay of a three-level (or N-level) atom

The central expressions (8)—(10) include the case of a N-level atom as a special limit when the dipole-strength distribution is comprised of a series of infinitely narrow spikes at the energies of the levels

$$\begin{aligned}
F(\omega)&=\lim_{\gamma_1\to 0}\sum_{j=1}^{N-1}\left[\frac{\Omega_0}{2}\right]_j\left[\frac{\gamma_0}{\pi}\right]^{1/2}\frac{1}{\omega-\omega_j+i\gamma_1}\,,\\
|F(\omega)|^2&=\sum_{j=1}^{N-1}\left[\frac{\Omega_0}{2}\right]_j^2\delta(\omega-\omega_j)\,,\\
A(\omega)&=\lim_{\gamma_1\to 0}\sum_{j=1}^{N-1}\sqrt{C_j}\left[\frac{\gamma_1}{\pi}\right]^{1/2}\frac{1}{\omega-\omega_j+i\gamma_1}\,,\\
p(\omega)&=\lim_{\gamma_p\to 0}\sum_{j=1}^{N-1}C_j\frac{\gamma_p^2}{(\omega-\omega_j)^2+\gamma_p^2}\,,
\end{aligned} \tag{22}$$

with the normalization $\sum_{0=1}^{N-1}C_j=1$. To demonstrate the type of results in this limit, consider the simplest nontrivial case of a three-level atom, prepared initially in the first excited states [Fig. 2(a)]. The only couplings allowed are between ($|1\rangle, |0\rangle$) and ($|2\rangle, |0\rangle$) pairs of states, and we examine the spectrum of the resonance fluorescence.

The underlying "physics" of this simple problem can be understood from two angles. Note that the photon emitted as state $|1\rangle$ starts to decay can be reabsorbed and, thereby, promotes the atom to state $|2\rangle$, and vice versa. These virtual transitions occur at the very initial stages of the decay, i.e., for $t\Delta\omega \ll 1$, when energy conservation is not possible. Therefore, at later times, the spectrum of the emitted light is composed of contributions from the decay of both $|1\rangle$ and $|2\rangle$. This, in turn, must lead to an interference pattern of the Fano type[5] characterized by a Fano zero (where the decays of $|1\rangle$ and $|2\rangle$ exactly cancel each other) and skewed line shapes. The same conclusion is reached from yet another point of view.

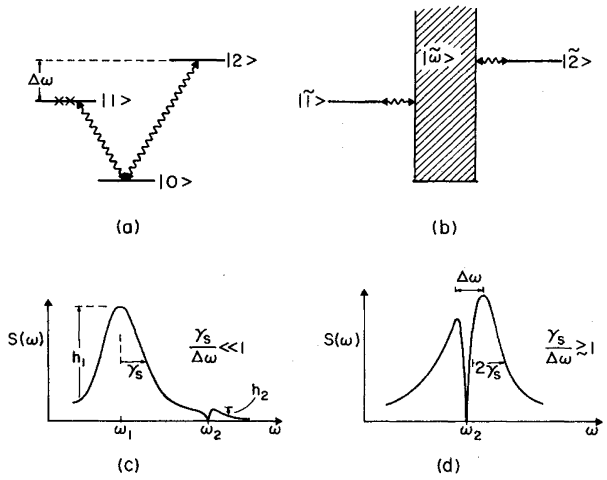

FIG. 2. Schematic display of the three-level atom and the resulting resonance fluorescence spectrum. (a) shows the energy levels and the one-photon transitions indicated by wavy lines. (b) depicts the equivalent Fano problem, see text. (c) and (d) show the spectrum in the limits of well-separated and almost degenerate lines, respectively. Note the Fano zero at $\omega=\omega_2$.

The relevant states for the present configuration are $|\tilde{1}\rangle=|1\rangle_A|0\rangle_R$, $|\tilde{2}\rangle=|2\rangle_A|0\rangle_R$, and $|\tilde{\omega}\rangle=|0\rangle_A|\omega\rangle_R$, where the subscripts A and R refer to the atom and radiation field, respectively. *Only* one-photon states are involved in the rotating-wave approximation. Note also that the sole couplings are between $|\tilde{1}\rangle$ and $|\tilde{\omega}\rangle$, $|\tilde{2}\rangle$ and $|\tilde{\omega}\rangle$ [Fig. 2(b)]. This is precisely the Fano problem of embedding two bound states into one continuum[5,8] with $q=0$, where q is the Fano asymmetry parameter. Hence the spectrum is expected to have a (Fano) zero at $\omega=\omega_2$ (the energy of the upper state) and two skewed peaks. The same configuration has been previously considered also in the context of multiphoton ionization and laser-enhanced autoionization.[10]

According to the discussion above, the calculation of the spectrum can be worked out in two ways: either by applying the general expressions (8) and (10), or by solving the equivalent Fano problem. Consider first the general expression (8), which yields $P_2(t)$, the time-dependent population of level $|2\rangle$. Straightforward manipulations using (8) and (22) with $C_1=1$, $C_2=0$, and

$$p(\omega)=\lim_{\gamma_p\to 0}\frac{\gamma_p^2}{(\omega-\omega_1)^2+\gamma_p^2} \tag{23}$$

give for well-separated lines

$$P_2(t)=\frac{\gamma_s(1)\gamma_s(2)}{(\Delta\omega)^2}\left|e^{-\gamma_s(1)t}-e^{-i(\Delta\omega)t}e^{-\gamma_s(2)t}\right|^2, \tag{24}$$

where $\gamma_s(j)=(\Omega_0/2)_j^2 Q(\omega_j)$ is the natural linewidth of level $|j\rangle$ and $\Delta\omega=\omega_2-\omega_1$. To simplify the analysis assume hereafter that $\gamma_s(1)=\gamma_s(2)=\gamma_s$. Then (24) implies that initially the population $P_2(t)$ rises as t^2, reflecting the two-photon process [Fig. 2(a)], which depletes the population in state $|1\rangle$. This rise continues over a duration

$\Delta t\Delta\omega\sim\pi$, followed by a decaying-oscillating behavior at all later times. The oscillations, of frequency $\Delta\omega$, reflect the beats between the decay of $|2\rangle$ and $|1\rangle$. The frequency measurement of such beats is at the basis of beam-foil spectroscopy.[4] To compute the spectrum of the resonance fluorescence we use (14) and (23) in conjunction with (10). The result is

$$K_3(z_1,z_2)=\frac{\left[\frac{\Omega_0}{2}\right]_1^2}{P^*(z_1)P(z_2)}[(z_1-i\omega_2)(z_2+i\omega_2)+(z_1+z_2)\gamma_s(2)\eta+\gamma_s^2(2)\eta^2] \tag{25a}$$

with the secular equation

$$P(z)=[z+i\omega_1+\gamma_s(1)][z+i\omega_2+\gamma_s(2)]-\gamma_s(1)\gamma_s(2)\ , \tag{25b}$$

and

$$S(\omega_\lambda)=\frac{C[(\omega_\lambda-\omega_2)^2+\gamma_s^2\eta^2]}{[(\omega_\lambda-\tilde{\omega}_1)^2+\tilde{\gamma}_s(1)^2][(\omega_\lambda-\tilde{\omega}_2)^2+\tilde{\gamma}_s(2)^2]}\ . \tag{26}$$

In (26) $\eta=1-Q(\omega_1)/Q(\omega_2)=1-\omega_1/\omega_2\cong\Delta\omega/\omega_1$, $\tilde{\Gamma}_j=-i\tilde{\omega}_j-\tilde{\gamma}_s(j)$ are the two roots of the secular equation (25b), and C is an uninteresting normalization constant.

Provided $(\gamma_s/\omega_1)^2\ll1$, which is a very good approximation in all cases of interest, expression (26) indeed shows a Fano zero at $\omega=\omega_2$ as expected from the discussion above. The actual form of the spectrum is analyzed in two limits—of well-separated lines and almost degenerate lines. The secular equation [(25b)] then gives

$$\tilde{\omega}_1=\omega_1+\frac{\gamma_s^2}{\Delta\omega},\quad \tilde{\omega}_2=\omega_2-\frac{\gamma_s^2}{\Delta\omega}\ ,$$
$$\tilde{\gamma}_s(1)=\tilde{\gamma}_s(2)=\gamma_s\quad\text{for}\quad\frac{\gamma_s}{\Delta\omega}\ll1 \tag{27a}$$

and

$$\tilde{\omega}_1=\tilde{\omega}_2=\tfrac{1}{2}(\omega_1+\omega_2)\ ,$$
$$\tilde{\gamma}_s(1)=\frac{(\Delta\omega)^2}{8\gamma_s},\quad \tilde{\gamma}_s(2)=2\gamma_s-\tilde{\gamma}_s(1)\quad\text{for}\quad\frac{\gamma_s}{\Delta\omega}\gg1\ . \tag{27b}$$

The spectra in these two limits are quite different. In the well-separated lines limit, $\gamma_s/\Delta\omega\ll1$ [Fig. 2(c)], there are two peaks with a large disparity in heights:

$$\frac{h_1}{h_2}\cong\frac{S(\tilde{\omega}_1)}{S(\tilde{\omega}_2)}\cong\left[\frac{\Delta\omega}{\gamma_s}\right]^4\gg1\ . \tag{28}$$

The spectrum in the almost-degenerate-lines limit [Fig. 2(d)], on the other hand, exhibits a "hole-burning" shape provided $\gamma_s,\Delta\omega$ are about the same magnitude. In the $\Delta\omega=0$ limit, the Fano zero is canceled out in (26) and the spectrum is one Lorentzian of width $2\gamma_s$.

The second method of calculating the spectrum, i.e., by solving the equivalent Fano problem involving two bound states and one continuum, is worked out elsewhere[8] and will not be repeated. It gives the *same* result (26) and secular equation (25b) using the identification $S(\omega)=\langle\tilde{\omega}|\hat{\rho}(t=\infty)|\tilde{\omega}\rangle$.

IV. CONCLUSIONS

We derived a general expression [Eqs. (8) and (9)] for the time-dependent density matrix describing the spontaneous decay of a continuum and the ensuing resonance fluorescence spectrum [Eq. (10)]. The input to these expressions is the initial (atomic) density matrix and the dipole-strength distribution of the continuum. In the proper limit, the expressions apply to an N-level atom where all excited states can decay only to the ground state. The present expression generalizes the results of an earlier study[1] to an arbitrary initial density matrix and dipole strength. It also underscores the basic simplicity of the spontaneous-decay problem of any number of levels or continuum to a common lower level.

The expressions have been applied to two cases of interest, namely to a continuum with a Fano-type dipole distribution and to a three-level atom, initially prepared in the first excited state. In the latter case we examined both the spectrum of the fluorescent light and $P_2(t)$, the time-dependent population of the second excited state. The emerging picture is that at short times $t\Delta\omega\ll1$, where $\Delta\omega=\omega_2-\omega_1$ is the energy separation of the two levels, the second excited-state population increases. At later times the populations of both excited states decay and beat against each other giving rise to a Fano zero at ω_2 and two skewed peaks. The occurrence of a Fano zero in the context of spontaneous decay is interesting. Underlying it, as usual,[5,6,8,10] is an interference of two pathways for a transition between a given pair of initial and final states. The continuum $P_0(t)$ involves pure exponentials with time constants combining the effects of spontaneous decay and width of the dipole-strength distribution. In the limit when there are no initial coherences and the initial population is proportional to the squared Fano dipole distribution, we note more complicated forms of $P_0(t)$. This is an example for a situation when the decay rate of the system depends on the mode of preparation.

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