Complete Suppression of Spontaneous Decay of a Manifold of States by Infrequent Interruptions

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Complete suppression of spontaneous decay of a manifold of states is shown to be achievable in a model system by a combination of coherent excitation of overlapping resonances and a judicious infrequent application of microwave pulses.

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The possibility of suppressing spontaneous emission and other decay processes has been a source of great interest in recent years. Broadly speaking, one can identify three main approaches: (1) the modification of the spectrum of the (spontaneous photons) "bath," (2) use of the "quantum Zeno" effect, and (3) the coherent cancellation of the dipole matrix elements to a common ground state.

The modification of the spontaneously emitted photon bath can be performed in (micro) cavities [1–5] and in photonic band gap materials [6–10]. Whereas this is a highly important field, our interest here is in the suppression of the decay of an atom or a molecule in *vacuum* where the photon bath is assumed intact. Such is the case in the quantum Zeno effect [11–16] which is a method of suppressing spontaneous decay in vacuum: One can suppress the decay by continuously "resetting the clock" to zero time where the decay is in its "Gaussian phase." If one performs this "clock resetting" often enough, one can suppress the decay altogether. However, it turns out that the Gaussian phase of the decay is extremely short, occurring at intervals which are roughly inversely proportional to the frequency of the emitted photon. For visible-UV photons, this means that interruptions must be executed at a rate of $\sim 10^{15}$ Hz [16]. In addition to the practical difficulties of such extremely frequent interruptions, the system is really never freely evolving. Moreover, as shown recently [17], such extremely frequent interruptions can often *accelerate,* rather than suppress, the decay. Several level configurations which enable the prolonging of the short-time quadratic decay are discussed [18], but such schemes will not work for an arbitrary set of final states.

The third class of methods is based on the coherent cancellation of a transition due to the interference between two (or three) matrix elements leading to a common final state. This cancellation can occur in a radiatively or nonradiatively broadened line [19–21], or due to the interference between two transitions [22–24], which can also be Autler-Townes [25] split by radiative interaction with a third state. While these schemes work (on the conceptual level) [26–30], they cannot work for an arbitrary number of final states to which the system might decay. It is simply impossible to satisfy the destructive interference conditions simultaneously for all the transitions. Thus, whereas the "coherent cancellation" schemes may work for some atoms, they can never work for molecules where a host of vibrational levels in the ground electronic state can serve as the final states to which the system decays. In other works, inhibition of autoionization in two electron atoms by modification of a radial wave packet with an electric field has been demonstrated theoretically [31] and experimentally [32].

In this Letter we develop another method of suppressing spontaneous decay. It is based on the interference within a manifold of (two or more) decaying states. The great advantage of the method is that it works in the vacuum and for an *arbitrary* number of final states. It does so because it exploits interferences within the decaying manifold which are independent of the nature and number of the states to which the system decays. The scheme is not confined to spontaneous emission and can be applied to nonradiative decay of a system coupled to an arbitrary bath. Moreover, we achieve this goal by *infrequent* interruptions which operate *within* the decaying manifold, determined by the decay rate and not by the photon frequency. Thus, we can completely suppress the spontaneous emission of a visible photon of a set of excited electronic states of an atom or a molecule by the application of a *microwave* pulse every few nsecs.

Confining, for clarity, our attention to the suppression of spontaneous emission, we consider a system composed of a set of N_α material levels $|\alpha\rangle$ with zero photons coupled to a set of N_γ material levels $|\gamma\rangle$ and a set of one-photon modes in all directions $\hat{k}$ and polarizations $\hat{\epsilon}$, $|1_{\hat{k},\hat{\epsilon}}\rangle$, denoted jointly as $|E,\beta\rangle$. E is the total (matter + radiation) energy, given as $E = E_\gamma + \hbar\omega_k$, where $\omega_k = ck$, and $\beta(= \gamma, \hat{k}, \hat{\epsilon})$ is a joint index of the material state γ and the $\hat{k}$ directed, $\hat{\epsilon}$ polarized one-photon state.

The decay process starts at t_0, at which time we assume the system to be in a superposition of zero-photon states, $|\Psi(t = t_0)\rangle = \sum_\alpha c_\alpha|\alpha,0\rangle$. Expanding the time-evolution operator $e^{-iH(t-t_0)/\hbar}$ in $|E,\beta^-\rangle$, the eigenstates of the full (matter + radiation) Hamiltonian H, i.e., $[E - H]|E,\beta^-\rangle = 0$, we obtain at subsequent times that

$$|\Psi(t)\rangle = \sum_\alpha c_\alpha \sum_\beta \int_{E_i}^{E_f} dE\, e^{-iE(t-t_0)/\hbar}|E,\beta^-\rangle a^*_{\alpha,\beta}(E), \tag{1}$$

where $a^*_{\alpha,\beta}(E) \equiv \langle E, \beta^- \mid \alpha, 0\rangle$. Following Fano [19] we expand $|E,\beta^-\rangle = \sum_\alpha a_{\alpha,\beta}(E)|\alpha,0\rangle + \sum_{\beta'}\int dE' \times b^{E,\beta}_{E',\beta'}|E',\beta'\rangle$. The amplitude of finding the system in the zero-photon state and material state $|\alpha'\rangle$ at the time t is given as

$$c_{\alpha'}(t) \equiv \langle \alpha', 0 \mid \Psi(t)\rangle = \sum_\alpha c_\alpha M_{\alpha'\alpha}(t - t_0), \quad (2)$$

where $m_{\alpha'\alpha}(E) \equiv \sum_\beta a_{\alpha',\beta}(E) a^*_{\alpha,\beta}(E)$ and $M_{\alpha'\alpha}(t) = \int_{E_i}^{E_f} dE\, e^{-iEt/\hbar} m_{\alpha'\alpha}(E)$. Explicit expressions for the amplitude of having emitted a photon in direction $\hat{\mathsf{k}}$ and having made a transition to material state $|\gamma\rangle$ can also be written [19,33]. To obtain $a_{\alpha,\beta}(E)$ we proceed in the standard Feshbach partitioning technique [34–36] by defining an operator $Q = \sum_\alpha |\alpha,0\rangle\langle\alpha,0|$, which projects out the zero-photon states, and its orthogonal projector $P = I - Q$. Assuming that $|\alpha\rangle$ and $|\gamma\rangle$ diagonalize the material part of the Hamiltonian, we obtain [20,33,35,36] that

$$a_{\alpha,\beta}(E) = \sum_{\alpha'} [(E - i\epsilon - Q\Gamma Q)^{-1}]_{\alpha\alpha'} V(\alpha' \mid E, \beta), \quad (3)$$

where $Q\Gamma Q = QHQ + QHP(E - i\epsilon - PHP)^{-1}PHQ$; explicitly, its elements are given by $\langle \alpha, 0|Q\Gamma Q|\alpha', 0\rangle = E_\alpha \delta_{\alpha\alpha'} + \hbar\Delta_{\alpha\alpha'}(E) + i\frac{\hbar}{2}\Gamma_{\alpha\alpha'}(E)$, where

$$\Gamma_{\alpha\alpha'}(E) = \frac{2\pi}{\hbar}\sum_\beta V(\alpha \mid E, \beta) V(E, \beta \mid \alpha'), \quad (4)$$

$$\Delta_{\alpha\alpha'}(E) \equiv \frac{1}{2\pi} P_V \int_{E_i}^{E_f} dE'\, \Gamma_{\alpha\alpha'}(E')/(E - E'), \quad (5)$$

P_V denotes the principal-value integral, and $V(\alpha \mid E, \beta) \equiv \langle \alpha, 0|QHP|E, \beta\rangle$. We note that all the $|\alpha\rangle$ levels involved must have the same total angular momentum J, so that the off-diagonal elements $\Gamma_{\alpha\alpha'}(E)$ do not vanish.

The above set of equations allows us to calculate in an exact manner the amplitudes for the decay from a set of overlapping resonances. An example for the $|a_{\alpha,\beta}(E)|$ probability amplitude for two overlapping resonances is given in Fig. 1. (Γ is the average interaction strength $\Gamma \equiv \frac{1}{N_\alpha}\sum_\alpha \Gamma_{\alpha\alpha}$ and ΔE is the level spacing in the zero-photon manifold.) The interference leads to a dark state [20,22] at a midpoint. We now exploit the interference between resonances to choose $\{c_\alpha\}$—the preparation coefficients of our initial state such as to force the decay to proceed via quiescent period followed by "photon bursts." Such steplike decay is known to occur [33] for interfering overlapping resonances.

In order to determine the preparation coefficients which induce the widest quiescent periods we solve an optimization problem in which our objective is to minimize the decay during a given time interval τ. Using Eq. (2) we write $P_0(t \mid \mathsf{c})$, the total population in the zero-photon manifold at time t as $P_0(t \mid \mathsf{c}) = \sum_{\alpha'} |\langle \alpha', 0 \mid \Psi(t)\rangle|^2 = \mathsf{c}^\dagger \mathsf{K}(t)\mathsf{c}$, where c is the vector of c_α coefficients, and

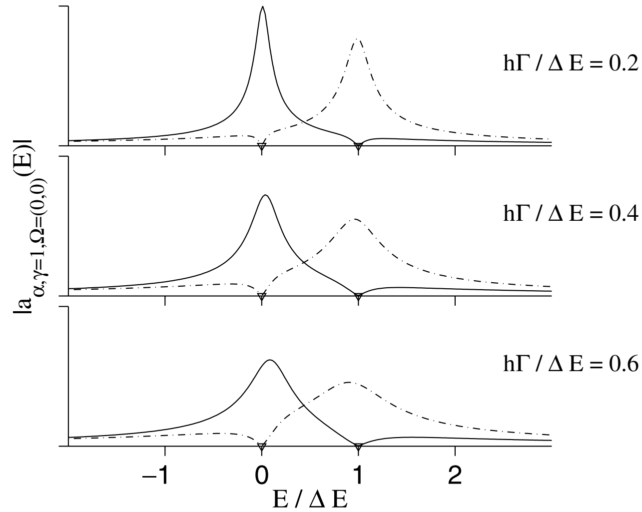

FIG. 1. Overlapping resonances $a_{\alpha\beta}(E)$ for $N_\alpha = 2$ (solid: $\alpha = 1$, dashed: $\alpha = 2$) and $\beta \equiv \{\gamma = 1, \hat{\mathsf{k}} = (\theta = 0, \phi = 0)\}$ at three different $\hbar\Gamma/\Delta E$ values. Clearly seen are the "dark energies" at which the emission probability is zero resulting from interference between neighboring resonances.

$\mathsf{K}(t) \equiv \mathsf{M}^\dagger(t)\mathsf{M}(t)$ with $\mathsf{M}(t)$ denoting the $M_{\alpha'\alpha}(t)$ matrix. Adding a Lagrange multiplier λ to assure the normalization $\sum_\alpha |c_\alpha|^2 = 1$, we now seek to maximize the quantity

$$P_{0,\lambda}(t \mid \mathsf{c}) = \mathsf{c}^\dagger \mathsf{K}(t)\mathsf{c} - \lambda \mathsf{c}^\dagger \mathsf{c}. \quad (6)$$

Differentiating with respect to $c^*_{\alpha'}$ and equating the result to zero at the target time τ, we obtain that the optimal vector $\mathsf{c}^{(\tau)}$ is a solution of the eigenvalue problem,

$$0 = \frac{\partial P_{0,\lambda}(t \mid \mathsf{c}^{(\tau)})}{\partial c^{*(\tau)}_{\alpha'}}\bigg|_{t=\tau} = \sum_\alpha K_{\alpha'\alpha}(\tau) c^{(\tau)}_\alpha - \lambda(t) c^{(\tau)}_{\alpha'}. \quad (7)$$

The results obtained from this optimization procedure are shown in Figs. 2a and 3a for $N_\alpha = 2, 3$ and for $\hbar\Gamma/\Delta E = 0.2$. For spontaneous emission the $\Gamma_{\alpha\alpha'}(E)$

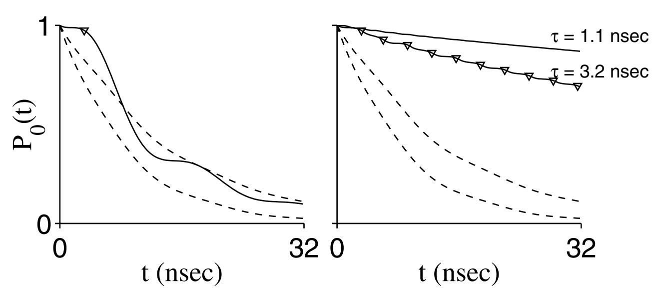

FIG. 2. Left: Naturally evolving optimized superposition state (solid lines) for $N_\alpha = 2$, $\Delta E = 0.5$ GHz (2.5×10^{-3} cm^{-1}), $1/\Gamma = 10.62$ nsec. The triangle denotes the optimization time τ; and the natural decay of the two states comprising the zero photon manifold (dashed lines). Right: Suppression of the decay of the optimized superposition state due to the application (at period τ of 1.1 and 3.2 nsec) of microwave-π pulses (solid lines). The triangles denote the times at which pulses are applied.

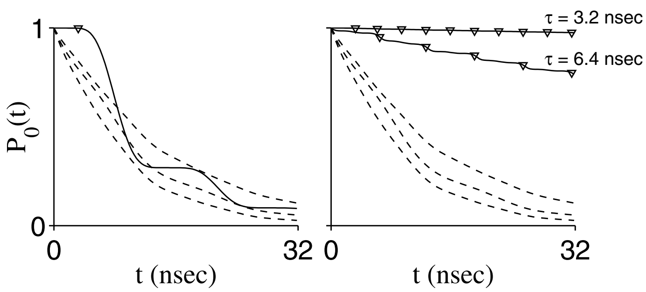

FIG. 3. The same as in Fig. 2, for $N_\alpha = 3$. At $\tau = 3.2$ nsec spontaneous emission is completely suppressed.

matrix elements are given as

$$\frac{e^2}{3\pi\varepsilon_0\hbar^4 c^3}\sum_{\{\gamma:E>E_\gamma\}}\mathbf{D}_{\alpha,\gamma}\cdot\mathbf{D}^*_{\alpha',\gamma}(E-E_\gamma)^3, \tag{8}$$

with $\mathbf{D}_{\alpha,\gamma}$ being the dipole matrix elements. Figures 2a and 3a show the time dependence of $P_0(t\,|\,\mathsf{c})$, the zero-photon manifold population, corresponding to the optimal choice of the preparation coefficients $\{c_\alpha\}$. The optimization time $t = \tau$ is marked on the figure by a triangle. The dashed lines denote the natural decay of each zero-photon state $|\alpha', 0\rangle$, i.e., when $c_\alpha = \delta_{\alpha,\alpha'}$. We note that the experimental realization of the required superposition can be achieved by photoabsorption of a shaped pulse [37,38].

The decay pattern is seen to proceed in steps: following a quiescent period, the ensemble decays rapidly by releasing a burst of photons. This phenomenon repeats itself periodically, until the decay is complete. The quiescent period, which is longer and flatter the more states are involved (i.e., the larger is N_α), is a result of a constructive interference between the various (radiatively) broadened overlapping line shapes $a_{\alpha,\beta}(E)$. It occurs *exclusively* in the excited manifold and, due to the fact that $\Gamma_{\alpha\alpha'}(E)$ and $\Delta_{\alpha\alpha'}(E)$ involve a general sum over final states, the interference holds for an arbitrary number of final (one-photon and material) channels.

The photon bursts noted above arise when, during its natural evolution, the c_α coefficients describing the superposition of overlapping resonances no longer assume the *specific* relations which bring about the constructive interference which results in the quiescent periods. It would therefore appear that one might be able to suppress the bursts by applying, just before the onset of the bursts, (microwave) pulses which periodically *reverse* the time evolution of the c_α coefficients. In this way the system would be shuttling back and forth in time in the quiescent phase and the spontaneous decay would be suppressed for all times. In the $N_\alpha = 2$ case the pulses we seek are identical to the π pulses used in the field of *photon echoes* [39].

In the following we present a detailed study and demonstration of this idea. Assuming that the π pulses are very short compared to the $1/\Gamma$ average decay time, we can safely neglect the decay during their application. As a result, when the pulses are applied on resonance, the coefficients describing the superposition state in the two-state Q manifold evolve as $\dot c_1(t) = i\Omega c_2(t)$ and $\dot c_2(t) = i\Omega^* c_1(t)$. Assuming that the duration of the pulse is $\delta\tau$, we have that $c^+_\alpha(t)$, the $c_\alpha(t)$ coefficients after the pulse, are related to the $c_\alpha(t)$ coefficients before the pulse as

$$\begin{aligned} c^+_1(t) &= c_1(t)\cos(\Omega\,\delta\tau) + ic_2(t)\sin(\Omega\,\delta\tau),\\ c^+_2(t) &= ic_1(t)\sin(\Omega\,\delta\tau) + c_2(t)\cos(\Omega\,\delta\tau). \end{aligned} \tag{9}$$

Based on the photon-echo experience, our first guess is to choose Ω such that $\Omega\,\delta\tau = \frac{\pi}{2}$ (a π pulse). In that case we have that the pulse transfer matrix in the Q space is given by the (unitary) transformation,

$$\mathsf{T} = \begin{pmatrix} 0 & i \\ i & 0 \end{pmatrix}.$$

Likewise, in the three-state case our first guess is to apply a 3×3 transfer matrix of

$$\mathsf{T} = \begin{pmatrix} 0 & 0 & i \\ 0 & i & 0 \\ i & 0 & 0 \end{pmatrix},$$

which can be realized by a variety of two (microwave) pulse configurations.

If one desires the decaying states to be coupled radiatively to the same ground state $|E, \beta\rangle$, a two-photon π pulse can be used to reverse the population between two $|\alpha, 0\rangle$ states. Such a two-photon π pulse can be realized by a Raman process conducted with visible light or by a two-photon microwave process.

We note that although the microwave pulses operate in the Q manifold (of vacuum visible photon states), and due to the vast energy mismatch cannot couple directly the Q manifold to the P manifold, the microwave pulse can nevertheless indirectly affect the one-photon P manifold. We have calculated the effect of the microwave pulse on this manifold by substituting the postpulse $c^+_\alpha(t)$ coefficients into the expression for $\Psi(t)$ [Eq. (2)].

In Figs. 2 and 3 we present a series of simulations of the suppression of spontaneous emission by the above scenario. We have considered spontaneous decay due to the coupling [40] of a zero-photon manifold composed of 2 or 3 material $|\alpha\rangle$ states to a one-photon manifold composed of all photon directions and two material $|\gamma\rangle$ states. We have assumed (highly allowed) dipole matrix elements $D_{\alpha,\gamma}$, corresponding to Einstein A coefficients of $\sim 5 \times 10^8$ sec^{-1}. The resulting decay time is $1/\Gamma = 10.62$ nsec and the natural period defined by the energy spacing is taken as $\hbar/\Delta E = 0.2/\Gamma$; therefore picosecond pulses can be considered as instantaneous. The required Rabi rates for 1 psec square pulses are $\Omega = \frac{\pi}{2\delta\tau} = 1.6$ THz ($3.8 * 10^{-5}$ a.u.). The optimized superposition is given by $\mathsf{c} = (0.80, -0.44 - 0.41i)$ for Fig. 2a and $\mathsf{c} = (-0.36 + 0.34i, 0.78, -0.28 - 0.26i)$ for Fig. 3a. In Figs. 2b and 3b we show the zero-photon manifold population resulting from applying a train of π pulses to the freely evolving superposition state of dimensionality $N_\alpha = 2$ and 3, respectively, whose steplike decay is shown in Figs. 2a and 3a. Quite clearly, the decay is effectively suppressed. The suppression becomes complete as the frequency of interruptions increases (compare the 1.1 nsec period to the 3.2 nsec period in Fig. 2 or the 3.2 nsec period to the 6.4 nsec period in Fig. 3), or the dimensionality of the Q manifold increases (compare Fig. 2b to Fig. 3b).

As an example to a molecular system on which this scheme can work, consider molecules with congested spectra where the widths emanating from spontaneous emission and/or predissociation are in the order of magnitude of the spacing between resonances. This is the case with the NaI molecule [41], possessing a complex spectrum with overlapping resonances, due to interferences induced by curve crossing processes.

An atomic system where our scheme can be most easily realized experimentally is an atom with a ground state and an excited electronic state possessing one resonance split by a magnetic field. The coherences on the excited states may be created by a pulse transferring population initially on the ground state. An x-polarized pulse would populate the two states $M + 1, M - 1$, and a z-polarized pulse would populate the state M. The selection rules for this system conform with the requirements imposed by our suppression scheme.

An additional example makes use of the Autler-Townes splitting effect [25]. A single resonance is split to a doublet in the presence of a resonant strong cw field, coupling it to a third state (which does not emit spontaneously itself). The two resulting resonances are of the same symmetry, and are suitable for our suppression scheme. The intensity of the cw field is controlled such that the splitting is comparable with the decay widths.

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