Electromagnetically Induced Quantum Memory

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(Received 11 July 2003; published 29 January 2004)

We discuss the problem of creating coherence in an optically driven quantum system in conditions where decoherence is caused by the laser field itself, due to coupling of the system to a rapidly decaying state or continuum. It is shown that by applying an additional laser field between this state and a bound state the relaxation channel can be suppressed as a result of a "dark state" formation, giving rise to long living Rabi oscillations in the system. It is found that the same mechanism of preserving coherence exists in systems with level splitting or degeneracy, where the driving field interacts with multiple resonant sublevels simultaneously. We also show that specific coherent propagation phenomena assisted by the interference suppression of decoherence can be observed under these conditions.

DOI: 10.1103/PhysRevLett.92.043002 PACS numbers: 32.80.Qk, 32.80.Bx, 42.65.Re

Coherent dynamics of quantum systems interacting with complex shaped optical fields has recently become the focus of many theoretical and experimental studies. An important goal is to reveal physical factors leading to a loss of coherence (e.g., the information about the phase of the wave function of the system) and determine conditions under which the system would retain phase "memory" for a possibly longer time [1]. This is very important for the concept of coherent control, where a laser field is used to bring a system into a specific quantum state in order to facilitate chemical reactions [2,3]. Another fundamental aspect is the proposed possibility to exploit quantum memory for information storage and data processing. Recently, an optically driven two-level system was suggested as a simple model of quantum bits, and simple operations (e.g., storing and readout of quantum information using atomic [4], ionic [5], and excitonic [6] systems controlled by π and 2π pulses) were demonstrated. In the ideal case, a two-level system should allow one to do as many operations as possible during the time it is quantum mechanically coherent. This suggests that the parameter $(\Omega_{\rm Rabi}T_2)$ (where $\Omega_{\rm Rabi}$ is the Rabi frequency and T_2 is the decoherence time) should be as large as possible. One way to achieve a higher value of this parameter is to control the coupling to environment to provide a longer T_2 [5,7]. Alternatively, the efficiency can be increased by operating in the high intensity regime. However, a serious challenge arising here is that the strong field regime can bring into existence additional, intensity-dependent relaxation channels. This may occur, for example, as a result of one- or multiphoton coupling of the "working" levels to other states or continuum of states decaying via dissociation, autoionization, internal conversion, etc. Whereas much attention was focused recently on the study of decoherence processes which are independent of the driving field [5,7], dynamics of quantum systems in conditions where a loss of coherence is caused by the driving field itself has not been discussed so far. The problem is rather general and arises in many other fields, e.g., in laser chemistry and laser spectroscopy [8], resonant frequency conversion [9–11], where to create population inversion or prepare a system with maximum polarization on a short time scale one needs higher field intensities. Strong driving fields in turn lead to a decoherence in the system. This brings up the questions: to what extent can a strong driving field destroy coherence in the system, and, how to maintain coherence in such a system for as long a time as possible?

In this Letter we address these questions and show that under certain conditions the decoherence caused by a strong driving field can be significantly reduced and even completely suppressed, and as a result, the system, initially incoherent, exhibits long living Rabi oscillations. To get insight into the physics of creating coherence, we consider a model quantum system in Fig. 1(a). In this system, the working transition $|g\rangle \rightarrow |1\rangle$ interacts with the laser field via a multiphoton process, and the coupling of the upper working level $|1\rangle$ to the rapidly decaying state $|\gamma\rangle$ models the field-induced relaxation. The model describes various systems in which strong excitation of the upper level leads to a population transfer to autoionizing states [9–14], vibrational quasicontinuum [8], and upper lying electron subbands in a quantum well [15]. If this process is sufficiently fast, the system decays before it executes one Rabi oscillation. The basic idea is that the coupling of the upper level to the state $|\gamma\rangle$ can be controlled by applying an additional field on the transition $|2\rangle \rightarrow |\gamma\rangle$. In fact, this leads to a creation of a dark state in the Λ-type system formed by states $|1\rangle$, $|2\rangle$, and $|\gamma\rangle$, which switches off the relaxation channel. As a result, the system exhibits coherent Rabi oscillations between the ground state and the bright state formed by a coherent superposition of states $|1\rangle$ and $|2\rangle$. Our analysis shows that such an immunity to decoherence is inherent in many quantum systems with level splitting or degeneracy, where the laser field interacts with more than one upper resonant sublevel simultaneously [Fig. 1(c)], and leads to a specific propagation dynamics in the medium. We point out that although the effects discussed here are mediated by quantum interference phenomena ["dark states,"

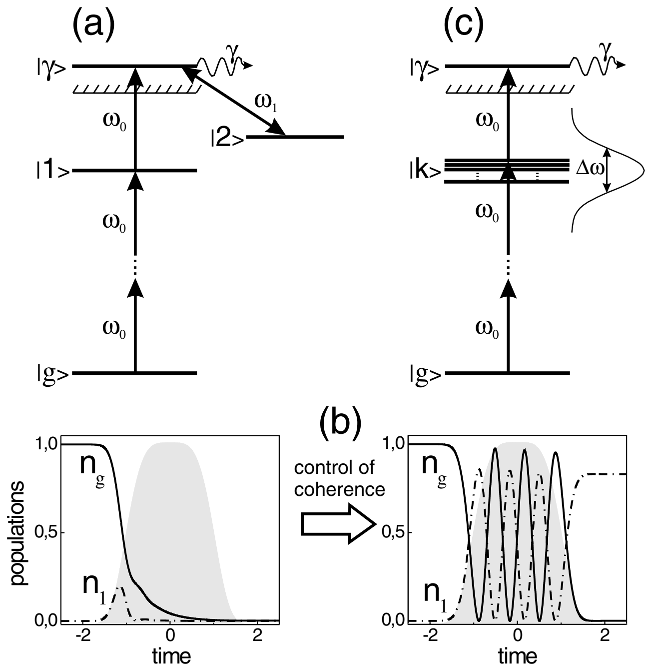

FIG. 1. (a) Schematic of a two-level system which decoheres due to a one-photon coupling to a decaying state $|\gamma\rangle$. (b) Suppression of the relaxation channel is achieved by applying an additional field on the transition $|2\rangle \leftrightarrow |\gamma\rangle$, leading to a dark-state formation within the system $|1\rangle - |\gamma\rangle - |2\rangle$. (c) Schematic of a system with quasidegenerate resonant upper sublevels.

population trapping, electromagnetically induced transparency (EIT) [12–16]], they represent a different type of coherent behavior. Unlike the steady-state (adiabatic) interactions in EIT, the response of the quantum system to the field contains explicit information about its past states. Note that more complex systems (e.g., three level Λ systems) have also been in the focus of many studies, especially due to the ability of the systems to store not the energy and the momentum of photons, but their quantum states (memory for photons [16]). We also note some analogy between the idea of coupling the rapidly decaying state to an additional state to suppress decoherence in the two-level systems and the method of stimulated Raman adiabatic passage in Λ systems [17].

We begin our analysis with the equations of motion for the systems shown in Fig. 1. The working transition $|g\rangle \rightarrow |1\rangle$ in Fig. 1(a) is characterized by the N-photon Rabi frequency $\Omega^{(N)}(t) = (\mu_{1g}^{(N)}/\hbar)E_0(t)^N$, where $\mu_{1g}^{(N)}$ is the N-photon matrix element [10,18]. The relaxation is modeled by a one-photon process $|1\rangle \rightarrow |\gamma\rangle$, since this is the case of most rapid decay of coherence [14], and control of coherence is performed by the field E_1 at ω_1 applied on the transition $|2\rangle \rightarrow |\gamma\rangle$. The corresponding Rabi frequencies are $\Omega_{1\gamma} = (d_{1\gamma}/\hbar)E_0(t)$ and $\Omega_{2\gamma} = (d_{2\gamma}/\hbar)E_1(t)$, where $d_{1\gamma}$ and $d_{2\gamma}$ are the dipole moments. In our notation the laser fields (E_0 and E_1) are taken in the form $\varepsilon_j(t) = E_j(t)\exp[i(\omega_j t + \varphi_j(t))] + \text{c.c.}$ $(j = 0, 1)$, where $E_j(t)$ is the real amplitude, ω_j is the carrier frequency, and $\varphi_j(t)$ is the phase. Also, by assuming that the state $|\gamma\rangle$ decays with a rate γ, we avoid the need for the density matrix formalism [13,14]. Before proceeding further, we notice that in the resonant approximation, the equations of motion for the system in Fig. 1(a) may formally be rewritten in terms of a system with degenerate upper working levels as in Fig. 1(c). This is possible if the amplitudes of the two fields are proportional. Setting $E_1(t) = s \cdot E_0(t)$, where $s = \text{const}$, one can formally replace the matrix element $d_{2\gamma}$ of the transition $|2\rangle \rightarrow |\gamma\rangle$ by $(s \cdot d_{2\gamma})$. After that the equations for the amplitudes may be rewritten in terms of a degenerate system driven by the field E_0, where the matrix element of the transition $|g\rangle \rightarrow |2\rangle$ formally equals zero. By drawing this analogy, we consider the equations for the probability amplitudes of the generalized system [Fig. 1(c)]:

$$\dot{a}_g = i\sum_k \Omega_{gk}^{(N)} a_k, \tag{1a}$$

$$\dot{a}_k = i\Delta_{kg} a_k + i\Omega_{kg}^{(N)} a_g + i\Omega_{k\gamma} a_\gamma, \tag{1b}$$

$$\dot{a}_\gamma = i\sum_k \Omega_{\gamma k} a_k - \gamma a_\gamma. \tag{1c}$$

Here $\Omega_{kg}^{(N)}(t) = (\mu_{kg}^{(N)}/\hbar)E_0^N(t)$ and $\Omega_{k\gamma}(t) = (d_{k\gamma}/\hbar)E_0(t)$ are the Rabi frequencies of the N-photon ($|g\rangle \rightarrow |k\rangle$) and one-photon ($|k\rangle \rightarrow |\gamma\rangle$) transitions with the matrix elements $\mu_{kg}^{(N)}$ and $d_{k\gamma}$ (which are assumed real), and $k = 1, 2, \ldots, K_0$, where K_0 is the number of sublevels. In general, the detuning Δ_{kg} of the kth sublevel from the resonance contains the frequency difference $\Delta\omega_{kg} = (N\omega_0 - \omega_{kg})$, the dynamical Stark shift of the levels in the field $\Delta\omega_{\text{St}}^{(k)} = (\alpha_k - \alpha_g)E_0^2/\hbar$ (where $\alpha_{k,g}$ are level polarizabilities) and the term $(N\partial\varphi(t)/\partial t)$ arising due to time modulation of the phase [18]. Below we assume that the sublevels are quasidegenerate ($\Delta\omega_{kg} \approx 0$), which holds for sufficiently short driving pulses ($\tau_p \ll \Delta\omega_{kg}$), and that the Stark shift is canceled by the phase-modulation term. In these conditions, the frequency detunings in Eq. (1b) can be neglected.

Some important properties of the system described by Eqs. (1a)–(1c) can be established analytically. To do this, we suppose the linewidth of state $|\gamma\rangle$ to be sufficiently large compared to the derivative on the left-hand side of Eq. (1c). This allows the probability amplitude a_γ to be eliminated, and then Eqs. (1b) for the amplitudes a_k take the form:

$$\dot{a}_k = i\Omega_{kg}^{(N)} a_g - \frac{\Gamma_k}{d_{k\gamma}}\left(\sum_{k'} d_{\gamma k'} a_{k'}\right), \tag{2}$$

where we introduced the golden rule transition probability $\Gamma_k = (\Omega_{k\gamma}^2/\gamma)$ from state $|k\rangle$ to state $|\gamma\rangle$. From Eq. (2) it follows that concurrent with transitions to the ground state $|g\rangle$, each $|k\rangle$ state decays with a relaxation rate determined by a coherent superposition of all the $|k\rangle$ states coupled by the field. To clarify this behavior, we built up two combinations of the $|k\rangle$ states. The first one is defined as $\tilde{a}_D := (\sum_k d_{k\gamma} a_k)/(\sum_k d_{k\gamma}^2)^{1/2}$ and (as will be shown below) evolves towards formation of a "dark" state on the transition from the states $\{|k\rangle\}$ to the state $|\gamma\rangle$, while the second combination $\tilde{a}_B := (\sum_k \mu_{gk}^{(N)} a_k)/(\sum_k [\mu_{gk}^{(N)}]^2)^{1/2}$ is a composite state associated with transitions between the ground state $|g\rangle$ and the levels $\{|k\rangle\}$. By combining Eq. (2) with Eqs. (1a) and (1b), we come to a closed system of equations for $\tilde{a}_D$, $\tilde{a}_B$, and a_g:

$$\dot{\tilde{a}}_D = i\Omega_0(\vec{\mu}, \vec{d})a_g - \Gamma_\Sigma \tilde{a}_D, \tag{3a}$$

$$\dot{\tilde{a}}_B = i\Omega_0 a_g - \Gamma_\Sigma(\vec{\mu}, \vec{d})\tilde{a}_D, \tag{3b}$$

$$\dot{a}_g = i\Omega_0 \tilde{a}_B. \tag{3c}$$

In Eqs. (3) $(\vec{\mu}, \vec{d})$ is a scalar product of two normalized K_0-component coupling vectors defined as $\vec{\mu} = (\mu_{1g}^{(N)}, \mu_{2g}^{(N)}, \ldots,)/(\sum_k [\mu_{kg}^{(N)}]^2)^{1/2}$ and $\vec{d} = (d_{1\gamma}, d_{2\gamma}, \ldots,)/(\sum_k d_{k\gamma}^2)^{1/2}$, $\Omega_0 = (\sum_k [\mu_{kg}^{(N)}]^2)^{1/2} E_0^N/\hbar$ is the Rabi frequency of the system in the absence of the relaxation channel, and $\Gamma_\Sigma = \sum_k \Gamma_k$ is the net relaxation rate. Dynamics of the system (3) can be fully characterized by its characteristic frequencies. On the assumption that the driving field amplitude is constant, the system can be reduced to one third-order differential equation, whose characteristic frequencies are found from the algebraic equation:

$$i\Omega^3 + \Gamma_\Sigma \Omega^2 - i\Omega_0^2 \Omega - \Gamma_\Sigma \Omega_0^2[1 - (\vec{\mu}, \vec{d})^2] = 0. \tag{4}$$

A key role of collective effects in the evolution of the system becomes apparent if we compare the case of a single upper working level with that of multiple upper sublevels. In the case of $K_0 = 1$, the value of $(\vec{\mu}, \vec{d})^2$ in Eq. (4) is unity, and Eq. (4) reduces to the common equation of damped oscillations: $\Omega^2 - i\Gamma_1\Omega - \Omega_0^2 = 0$. With the increase of Γ_1, coherence in the system progressively decreases, and for $\Gamma_1/2 > \Omega_0$ the system shows an aperiodic behavior, suggesting a complete loss of coherence. However, when $K_0 > 1$, dynamics of the system is qualitatively different. Surprisingly, the increase of relaxation does not lead to a faster decay, but, in contrast, enhances coherence. This collective property of the system can be established by examining Eq. (4) in the limit of large Γ_Σ, in which the characteristic frequency of the system can be sought in the form of expansion $\Omega = \Omega^{(0)} + \Omega^{(1)} + \cdots$ with a small parameter $(\Omega_0/\Gamma_\Sigma) \ll 1$. We find that in this limit the system exhibits damped Rabi oscillations with the frequency and the relaxation rate determined by

$$\Omega \approx \Omega_0\sqrt{1 - (\vec{\mu}, \vec{d})^2}, \qquad \Gamma \approx \frac{1}{2}\frac{\Omega_0^2}{\Gamma_\Sigma}(\vec{\mu}, \vec{d})^2. \tag{5}$$

The above expressions are the main result of our analysis. As follows from Eqs. (5), the collective decay rate Γ is inversely proportional to the net relaxation rate of the upper sublevels, and in the limit $(\Omega_0/\Gamma_\Sigma) \ll 1$ is much smaller than the collective Rabi frequency Ω. The coherent behavior results from the transformation of the combined state $\tilde{a}_D$ into a dark state. Indeed, in the limit $\Gamma_\Sigma \gg \Omega_0$ the amplitude of the combined state in Eq. (3a) $\tilde{a}_D \approx 0$. According to Eq. (1c), this suggests a suppression of population transfer to the state $|\gamma\rangle$. In this regime the upper levels execute the same quantum mechanical motion: for a rectangular driving field applied at a moment $t = 0$ we find that the amplitudes are given by $a_g = \cos(\Omega t)$ and

$$a_k = i\frac{[(\vec{\mu})_k - (\vec{d})_k(\vec{\mu}, \vec{d})]}{\sqrt{1 - (\vec{\mu}, \vec{d})^2}}\sin(\Omega t),$$

where $(\vec{\mu})_k$ and $(\vec{d})_k$ are the kth components of the coupling vectors $(\vec{\mu})$ and $(\vec{d})$.

Using Eqs. (5), we can readily determine the corresponding characteristics of the system shown in Fig. 1(a). In particular, in the case of a one-photon transition $|g\rangle \to |1\rangle$, we have

$$\Omega = \Omega_{1g}\sqrt{1 - \frac{d_{1\gamma}^2}{d_{1\gamma}^2 + s^2 d_{2\gamma}^2}}, \qquad \Gamma = \frac{\gamma}{2}\frac{\mu_{1g}^2 d_{1\gamma}^2}{(d_{1\gamma}^2 + s^2 d_{2\gamma}^2)^2}, \tag{6}$$

where s is the above introduced ratio of the field amplitudes $s = E_1/E_0$. As follows from (6), with an increase of the control field E_1 the frequency trends to Ω_{1g}, while the relaxation rate rapidly drops, and in the limit $s \to \infty$ formally approaches zero. The above analysis does not answer the question what happens to the system during turning on the fields. To answer it we solved the exact

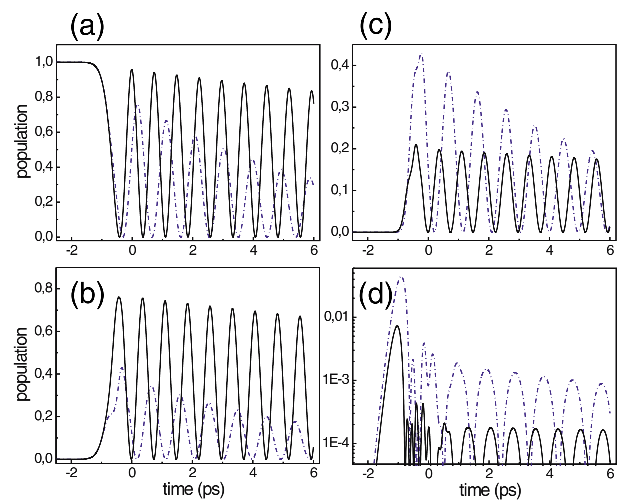

FIG. 2 (color online). Creating of quantum memory in the system of Fig. 1(a). Parameters of the transitions are such that in the absence of the control field (E_1) the system decays before executing one Rabi oscillation ($\Omega_{1g} = 0.95$ and $\Omega_{1\gamma} = 2.85$ in units of $\gamma = 5.0\ \mathrm{ps}^{-1}$). (a)–(c) Dynamics of the populations of the ground state $|g\rangle$, state $|1\rangle$, and state $|2\rangle$, correspondingly, and (d) the dynamics of the "dark" composite state, $|\tilde{a}_D|^2$, for different relative values of the control field ($s = E_1/E_0$). Dashed lines: $s = 1$ ($\Omega_{2\gamma} = 2.85$); solid lines: $s = 2$ ($\Omega_{2\gamma} = 5.7$).

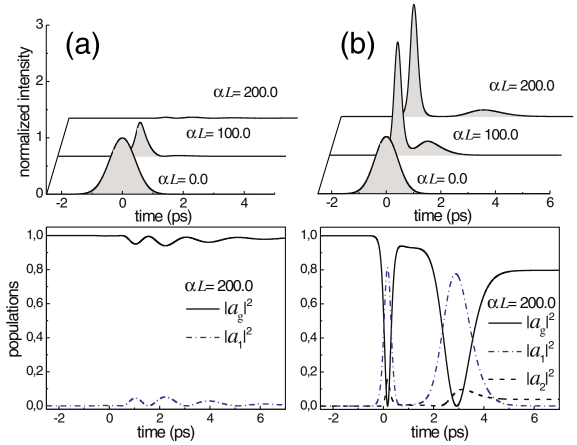

FIG. 3 (color online). Snapshots of a laser pulse in a resonant absorber in (a) of only one, and (b) two upper resonant sublevels coupled to continuum, at different propagation lengths αL. Also shown is the dynamics of the populations at $\alpha L = 200$. The length is given in units of inverse absorption coefficient ($\alpha = 2\pi\omega_0\mu_{1g}^2\mathcal{N}\tau_p/c\hbar$, where $\mathcal{N}$ is the atomic density, τ_p is the input pulse duration). The input parameters: $\tau_p = 1$ ps, $I_{\text{Peak}} = 10^{11}$ W/cm^2, (a) $\Omega_{1g} = 0.95$, $\Omega_{1\gamma} = 2.85$; (b) $\Omega_{1g} = 0.95$, $\Omega_{2g} = 0.31$, $\Omega_{1\gamma} = 2.85$, and $\Omega_{2\gamma} = 5.7$ (in units of $\gamma = 5.0$ ps^{-1}).

Eqs. (1a)–(1c) numerically. We assumed that the field amplitudes E_0 and E_1 were proportional and both have a steplike temporal profile. The results in Fig. 2 evidence that there exists a "preparation" stage during which the upper levels are populated, thus enabling creation of the dark state [Fig. 2(d)]. Once the dark state is formed, the system turns to the regime of regular slowly damped Rabi oscillations [Fig. 2(c)], where the two upper states build up a coherent superposition that is immune to the photoabsorption. An increase of the amplitude of the control field E_1 leads to a shorter preparation stage, a higher oscillation frequency, and a longer decoherence time (Fig. 2, solid lines), in accordance with the formulas (6).

The effect of induced coherence raises the interesting question of the existence of related phenomena in pulse propagation. Here we will discuss these effects for quasidegenerate systems as in Fig. 1(c). By solving Eqs. (1a)–(1c) self-consistently with the Maxwell equations for the field, we studied propagation of a short laser pulse in two situations. In the first one, a ps-laser pulse interacts with only one upper resonant level coupled to continuum, and, as one would expect, experiences a strong attenuation in the medium [Fig. 3(a)]. In the situation shown in Fig. 3(b), there are two closely lying upper resonant sublevels which are excited by the laser field simultaneously (as it takes place, for example, in excitation of the doublet $4s\ 2S_{1/2} \rightarrow \{5p\,^2P_{1/2}, 5p\,^2P_{3/2}\}$ of atomic potassium [8]). The coupling of the levels via continuum immediately leads to the collective behavior resulting in creating coherence in the system. The pulse is seen to break up into two subpulses in perfect analogy to a 4π pulse in a coherent absorber [19]. Unlike the classical self-induced transparency (SIT), the pulse "area" is now given by $\Theta \approx \hbar^{-1}\sqrt{(\mu_{1g}d_{2\gamma} - \mu_{2g}d_{1\gamma})^2/(d_{1\gamma}^2 + d_{2\gamma}^2)}\int_{-\infty}^{\infty} E_0(t)dt$. In fact, the dynamics represents a nontrivial example of SIT mediated by EIT, where a lossless coherent propagation arises as a result of interference suppression of the relaxation channel.

In conclusion, we have shown how coherence in an optically driven quantum system can be effectively controlled in conditions where phase relaxation is caused by the laser field itself. The proposed mechanism of preserving quantum memory based on the dark state formation seems to be rather general and may exist in different systems with degeneracy and level splitting. It may be interesting not only for quantum control and coherence control, but also for diverse nonlinear optical processes in the UV and VUV region, where coupling to autoionizing states and ionization continuum is typically involved in the interaction and can strongly affect resonant frequency mixing [9,11] and harmonic generation [10]. By using an appropriate superposition of upper resonant sublevels, decoherence in the system can be considerably reduced, thus allowing one to achieve a higher conversion efficiency.

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