Atomic Coherence via Modified Spontaneous Relaxation of Driven Three-Level Atoms

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A strong coherent field drastically modifies the spontaneous decay of three-level atoms when one dynamic Stark sublevel crosses a neighboring atomic state. This leads to an anomalous atomic response yielding maximal coherence, vanishing absorption, and ultralarge index of refraction.

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Recent research into the effects of atomic coherence in quantum optics has led to the concepts of, for example, lasting without inversion [1], electromagnetically induced transparency [2], enhanced index of refraction [3], and other effects [4]. Typically, these effects are produced by a coherent driving field or quantum interference associated with, e.g., a Fano resonance or spontaneous emission from an upper-level doublet to a common lower state. In this Letter, we report on a new method for achieving maximal atomic coherence utilizing level crossing between a Stark-shifted lower level and a nearby unperturbed atomic state as shown in Fig. 1.

In particular, we find that when the Stark-shifted state $|2\rangle$ drops below the bare ground state $|1\rangle$, spontaneous emission can take the atoms from 1 to 2. This has surprising consequences for the atomic coherence induced between $|2\rangle$ and $|3\rangle$ by the field which is Stark-shifting level $|2\rangle$. In particular, we find that the bare state populations and off-diagonal elements of the atomic density matrix now show *maximum atomic coherence,* that is,

$$\rho_{22} \simeq \rho_{33} \simeq \tfrac{1}{2}, \quad \rho_{32} \simeq \tfrac{1}{2} e^{-i(\phi+\omega t)},$$

where ϕ is the phase of the Rabi oscillation and ω is the frequency of the driving field. This is the main result of the present Letter. It is in contrast with the result obtained from a strongly driven two-level system consisting of the states $|2\rangle$ and $|3\rangle$ in which the atoms are initially prepared in their ground state and state $|3\rangle$ can decay into $|2\rangle$. In such a case we find $\rho_{22} \simeq \rho_{33} \simeq \frac{1}{2}$, as above, but $\rho_{32} \simeq 0$, that is, we have a "bleached" resonant transition but there is *no atomic coherence.*

We note, first of all, that this means that resonant driving of the 3-2 transition provides a way of depleting the ground-state population which is different from the traditional optical pumping and/or bleaching. Second, a field resonant with the 3-2 transition induces population inversion both between levels 2 and 1 and between levels 3 and 1. This result is unexpected. Indeed, according to the usual master equation with field-independent spontaneous emission rates, coherent pumping cannot lead to a population inversion at a transition whose frequency is larger than the frequency of the driven transition when there is no initial population inversion. It is multiphoton absorption of the strong driving field that makes the population inversion at the 3-1 transition possible.

At the same time, the magnitude of the optical polarization σ_{32} given by Eq. (6) below reaches its maximum ($|\sigma_{32}| \cong \frac{1}{2}$) instead of zero as predicted by the usual Maxwell-Bloch equations without the radiative coupling which yields the damping of the population from 1 to 2. We thus have a remarkable example wherein spontaneous emission helps to produce atoms in an almost pure quantum state ($|\sigma_{32}| \cong \sqrt{\rho_{22}\rho_{33}}$) with the largest possible coherence via depletion of the ground level. Let us emphasize also that the magnitude of $\mathrm{Im}(\sigma_{32}e^{-i\phi})$, which is responsible for resonant absorption, decreases to zero while the magnitude of $\mathrm{Re}(\sigma_{32}e^{-i\phi})$ characterizing the refraction index is maximal. Such a relation between the

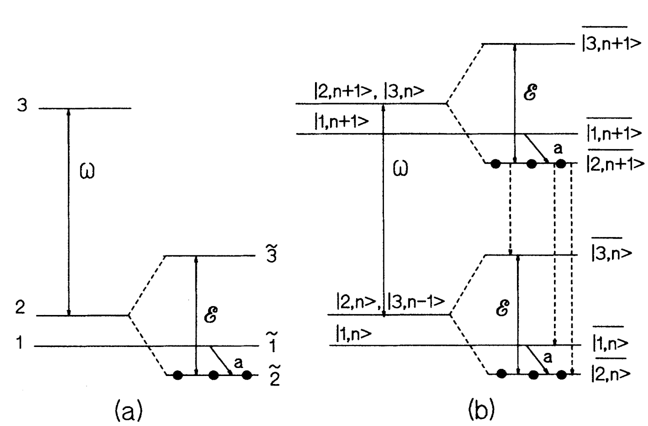

FIG. 1. Energy level in the bare and dressed state representations corresponding to the semiclassical (a) and the fully quantum (b) descriptions. ω is the frequency of the strong coherent field that drives the 2-3 transition, and (a) is the spontaneous relaxation rate that is responsible for populating the lower dressed state. Dashed lines indicate fluorescence transitions.

real and imaginary parts of the resonant susceptibility is just the opposite of the usual relation.

Cavity QED effects leading to a modification of the spontaneous relaxation rate of a "two-level" atom due to the change of the mode density in the cavity could provide a dramatic confirmation of the present predictions. For example, a high-Q sapphire microwave cavity of the type developed by Braginsky *et al.* [5] could be used with an atomic beam passing through the edge regions where the whispering gallery modes are strongest. When the Rabi frequency is adjusted to match a cavity resonance, the effective density of states of the radiation field will increase dramatically and the well known cavity QED enhancements of the decay rate would cause the atoms to make the desired transition to the "new" lower level.

We now proceed to develop the theory for the present effect along the lines of our recent treatment of a strongly driven atom which is undergoing spontaneous emission due to interaction with the vacuum. When the Rabi frequency exceeds one of the atomic frequencies, the traditional approach based on a master equation with the relaxation constants independent of the driving field is on longer valid. Hence we base our analysis on the generalized master equation we derived recently in the Born, Markov, and rotating-wave approximations [6]. The structure of this equation in the bare atomic state basis differs from the traditional one by the presence of cross-relaxation terms and the explicit dependence of the relaxation rates on the amplitude of the driving field. In the limit of a strong driving field, it is most appropriate to use the dressed states representation. The transformation matrix to the dressed states basis is

$$|\tilde{1}\rangle = |1\rangle, \quad |\tilde{2}\rangle = c|2\rangle - se^{-i(\phi+\omega t)}|3\rangle, \qquad |\tilde{3}\rangle = se^{-i\phi}|2\rangle + ce^{-i\omega t}|3\rangle, \tag{1}$$

$$c = \frac{\theta}{\sqrt{\theta^2 + \mathcal{E}^2}}, \quad s = \frac{\mathcal{E}S}{\sqrt{\theta^2 + \mathcal{E}^2}},$$

$$\theta = \left(|\delta| + \sqrt{\delta^2 + 4\mathcal{E}^2}\right)\Big/2, \quad \delta = \omega_{32} - \omega,$$

$$S = \delta/|\delta|.$$

Here $\mathcal{E} = \mu_{32}E/2\hbar$ is the Rabi frequency of the driving field $E = |E|e^{-i(\omega t+\phi)}$, μ_{32} is the dipole moment of the driven transition, and δ is the detuning of the field frequency ω away from the frequency of the resonant atomic transition. The transformation to the dressed states allows us to avoid the presence of the usual Hamiltonian coupling terms and, using the secular approximation in the dressed basis, we also avoid the usual cross-relaxation terms. As a result, the equations for the diagonal and the off-diagonal elements of the density matrix are decoupled and off-diagonal elements vanish in steady state. The set of equations for the populations of dressed states take the simple form

$$\dot{\tilde{\rho}}_{ii} = \sum_{j=1}^{3} R_{iijj}\tilde{\rho}_{jj}, \quad i = 1,2,3, \tag{2}$$

$$R_{1122} = c^2 w_{21}(\omega - \tilde{\omega}_{21}) + s^2 w_{31},$$

$$R_{2211} = c^2 w_{12}(\tilde{\omega}_{21} - \omega) + s^2 w_{13},$$

$$R_{1133} = s^2 w_{21}(\omega - \tilde{\omega}_{31}) + c^2 w_{31},$$

$$R_{3311} = s^2 w_{12}(\tilde{\omega}_{31} - \omega) + c^2 w_{13},$$

$$R_{2233} = s^4 w_{23} + c^4 w_{32},$$

$$R_{3322} = s^4 w_{32} + c^4 w_{23},$$

$$R_{iiii} = -\sum_{j\neq i} R_{jjii},$$

$$w_{mn}(\Omega) = \begin{cases} A_{mn}\left(-\dfrac{\Omega}{\omega_{mn}}\right)^3 [n(-\Omega) + 1], & \Omega < 0, \\ A_{mn}\left(\dfrac{\Omega}{\omega_{mn}}\right)^3 n(\Omega), & \Omega > 0, \end{cases} \tag{3}$$

$$A_{mn} \equiv A_{mn}(\omega_{mn}) = \frac{4\omega_{mn}^3 |\mu_{mn}|^2 \eta(\omega_{mn})}{3\hbar c^3},$$

$$w_{mn} = w_{mn}(\omega_{nm}),$$

$$\tilde{\omega}_{21} - \omega = \omega_{21} - \left(\sqrt{\delta^2 + 4\mathcal{E}^2} - \delta\right)\Big/2,$$

$$\tilde{\omega}_{31} - \omega = \omega_{21} + \left(\sqrt{\delta^2 + 4\mathcal{E}^2} + \delta\right)\Big/2.$$

Here $n(\Omega)$ is the number of photons in the mode of the field reservoir with frequency Ω, μ_{mn} and ω_{mn} are the dipole moments and frequency of the m-n atomic transition; $\eta(\omega_{mn})$ is a dimensionless factor characterizing the mode density of the reservoir as distinct from that of the vacuum [in vacuum $\eta(\omega_{mn}) = 1$]. The same set of equations can be obtained from the fully quantum description using the Wigner-Weisskopf formula for the relaxation rates between the fully quantized dressed states defined by the relations [see Fig. 1(b)]

$$|\overline{2,n}\rangle = c|2, n+1\rangle - se^{-i\phi}|3,n\rangle,$$

$$|\overline{3,n}\rangle = se^{-i\phi}|2, n+1\rangle + c|3,n\rangle,$$

and applying the semiclassical approximate which amounts to assuming that the field is in a coherent state with $n \gg 1$. Hence

$$\langle\overline{2,n}|\rho|\overline{2,n}\rangle \cong \langle\overline{2,n+1}|\rho|\overline{2,n+1}\rangle,$$

$$\langle\overline{3,n}|\rho|\overline{3,n}\rangle \cong \langle\overline{3,n+1}|\rho|\overline{3,n+1}\rangle.$$

If $\mu_{12} = 0$, these equations reduce to those derived by Cohen-Tannoudji and Reynaud [7]. In this Letter, we find novel effects which are due to field-dependent relaxation of the low-frequency 2-1 transition. These effects emerge only for $\mu_{12} \neq 0$ and become especially important when spontaneous decay at this transition is sufficiently strong.

In accordance with the definitions (2) and (3) the relaxation rates R_{2211} and R_{1122} are drastically modified when one of the ac Stark sublevels of the bare level 2 crosses level 1. In this case there is a change in the direction of spontaneous transition between these two levels. On resonance, this occurs when the Rabi frequency $\mathcal{E}$ exceeds the frequency difference $\omega_2 - \omega_1$. This effect is especially apparent in the steady-state response of atoms when the spontaneous decay from 2 to 1 is the dominant relaxation process. It corresponds to a relatively large frequency interval between these two levels, $\hbar\omega_{21} \gg kT$, or to a "cold" reservoir, $n(\Omega) \cong 0$ for $\Omega \geq \omega_{21}$. Here k is the Bolzmann constant and T is the temperature of the reservoir. Even at the liquid helium temperature ($T \sim 3$ K) this condition implies a fairly large frequency splitting $\omega_{21} \geq 4 \times 10^{11}$ s^{-1}. In this case, atoms do not respond on the field according to the usual Maxwell-Bloch equations. Indeed, both resonant levels 2 and 3 are empty for $\hbar\omega_{21} \gg kT$ and hence atomic polarization cannot be induced. However, the solution of Eq. (2) on resonance ($\delta = 0$) and for $\mathcal{E} \gg \omega_{21}$ is

$$\tilde{\rho}_{33} = [1 + 2(a + A_{31})(a + A_{31} + A_{32})/aA_{32}]^{-1}, \quad (4)$$

$$\tilde{\rho}_{11} = (A_{31} + a\tilde{\rho}_{33})/(A_{31} + a), \quad \tilde{\rho}_{22} = 1 - (\tilde{\rho}_{11} + \tilde{\rho}_{33}), \quad (5)$$

with $a = A_{21}(\mathcal{E})$, which yields a different conclusion. The action of the strong field ($\mathcal{E} > \omega_{21}$) leads to an exchange in the relative position of levels $\tilde{2}$ and 1 [Fig. 1(a)]. This implies the emergence of a spontaneous relaxation rate from level 1 to level $\tilde{2}$ which is responsible for populating level 2 and hence for "switching on" the interaction with the field. In the limit $\mathcal{E} \gg \omega_{21}$, when the frequency separation between levels 1 and $\tilde{2}$ is equal to the Rabi frequency, this spontaneous relaxation rate grows in vacuum [where $\eta(\mathcal{E}) = 1$] like the cube of the Rabi frequency $a \sim \mathcal{E}^3$. The condition $\mathcal{E} > \omega_{21}$, where $\omega_{21} \sim 10^{12}$ s^{-1}, $\mu_{32} \sim 1$ D requires a strong coherent field $E \geq 10^3$ cgs that corresponds to the intensity $W = cE^2/8\pi \geq 100$ MW/cm^2. If the relaxation rate a remains sufficiently small, nothing unexpected happens. All the atoms remain in level 1. However if $a \gg A_{31}$, the situation is completely changed. According to Eq. (4) we have $\tilde{\rho}_{33} \cong [1 + 2(a + A_{32})/A_{32}]^{-1}$. For $A_{31} \ll a \ll A_{32}$, all levels are equally populated both in the dressed and in the bare bases. For $a \gg A_{31}$ and $a \gg A_{32}$ all atoms are trapped in level $\tilde{2}$. This requires a relatively large dipole moment and mode density at the low-frequency 1-2 transition:

$$\mu_{21}^2\eta(\omega_{21})/\mu_{31}^2\eta(\omega_{31}) \gg (\omega_{31}/\mathcal{E})^3 \gg 1,$$

$$\mu_{21}^2\eta(\omega_{21})/\mu_{32}^2\eta(\omega_{32}) \gg (\omega_{32}/\mathcal{E})^3 \gg 1.$$

Let us note that one of two optical transitions, for instance 3-1, should normally be forbidden because of the selection rules in a three-level atom. Hence we are dealing basically with the second inequality. For $\mathcal{E} \sim 4 \times 10^{12}$ s^{-1} and $\omega_{32} \sim 2 \times 10^{14}$ s^{-1}, this leads to the requirement $\mu_{21}^2\eta(\omega_{21})/\mu_{32}^2\eta(\omega_{32}) > 10^5$. In this case, we find that the bare state populations and the complex amplitude of the off-diagonal density matrix element are

$$\rho_{22} \cong \rho_{33} \cong \tfrac{1}{2}, \quad \sigma_{32} \cong e^{-i\phi}/2, \quad (6)$$

and, since $\sigma_{32} \cong \rho_{32}e^{i\omega t}$, we have the result quoted in the beginning.

Trapping of atoms in one of the two dressed states ($\tilde{\rho}_{22} = 1$) modifies both the resonant fluorescence and the probe field absorption spectra differently than in the case of a strongly driven two-level system. That is, it breaks the symmetry of such spectra which occurred in the two-level case at resonant driving [Fig. 2(a)] due to equal populations of dressed states. As a result, the fluorescence spectrum contains two peaks of equal amplitudes at resonance ($\omega' = \omega_{32}$) and at the left sideband ($\omega' = \omega_{32} - \mathcal{E}$) while the right sideband peak at $\omega' = \omega_{32} + \mathcal{E}$ is absent. Accordingly, a probe field absorption spectrum in the case $\mu_{31} = 0$ contains an amplification peak at the left sideband and an absorption peak at the right sideband [Fig. 2(c)]. For $\mu_{31} \neq 0$, the additional amplification and absorption peaks should appear at $\omega' = \omega_{32} - \mathcal{E}/2$ and at $\omega' = \omega_{32} + (\mathcal{E}/2 - \omega_{21})$.

In conclusion, the crossing of a dynamic Stark sublevel and a neighboring atomic energy level results in a drastic modification of the spontaneous relaxation, vanishing absorption, ultralarge refraction index, and strong symmetry breaking of the traditional Mollow spectrum. In order to observe these effects one needs to simultaneously satisfy three main conditions:

$$\hbar\omega_{21} \gg kT, \quad \mathcal{E} > \omega_{21},$$

$$\mu_{21}^2\eta(\omega_{21})/\mu_{32}^2\eta(\omega_{32}) \gg (\omega_{32}/\mathcal{E})^3 \gg 1.$$

This requires some special effort, as follows from the estimates given above. Hence a careful selection of the appropriate transitions in atoms or molecules needs to be made for a successful experiment. As mentioned in the introduction, one technique for observing the effect would be to use the high-Q sapphire microwave cavity developed by Braginsky *et al.* [5] in order to enhance

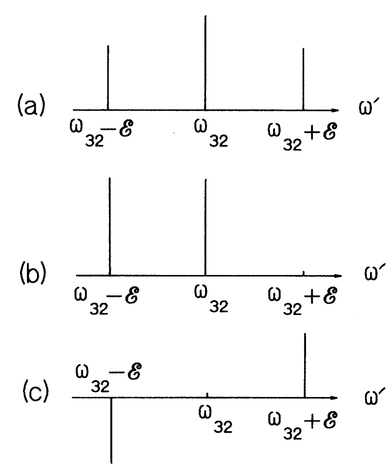

FIG. 2. (a) Fluorescence spectrum of a strongly driven two-level system consisting of only states $|2\rangle$ and $|3\rangle$ compared with (b) the fluorescence and (c) the probe field absorption spectra of a three-level system trapped in the lower dressed state with resonant driving ($\omega = \omega_{32}$) in the case where the 3-1 transition is forbidden ($\mu_{31} = 0$).

the spontaneous emission rate and thus induce atomic coherence.

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