Quantum interference effects induced by interacting dark resonances

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(Received 15 January 1999)

We predict the possibility of sharp, high-contrast resonances in the optical response of a broad class of systems, wherein interference effects are generated by coherent perturbation or interaction of dark states. The properties of these resonances can be manipulated to design a desired atomic response.
[S1050-2947(99)02507-X]

PACS number(s): 42.50.Gy, 42.50.Lc, 42.60.Fc

The phenomenon of ''dark resonances'' or coherent population trapping [1] is by now a well-known concept in optics and laser spectroscopy. It is a basis for such effects as electromagnetically induced transparency (EIT) [2] and its applications to nonlinear optics [3], lasing without inversion [4], the resonant enhancement of the refractive index [5,6], adiabatic population transfer [7], subrecoil laser cooling [8], and atom interferometry [9].

The essential feature of dark resonances is the existence of quantum superposition states, which are decoupled from the coherent and dissipative interactions. As a general rule, interactions involving such a ''dark state'' lead to decoherence and are undesirable. In the present paper we demonstrate a qualitatively different approach to these problems. The present approach involves multiple quantum superposition states that are coupled and interact *coherently*. We find that such interacting superpositions can be used, in many instances, to mitigate various decoherence effects and to enlarge the domain of dark-state-based physics.

In particular, we show that coherent interaction leads to a splitting of dark states and the emergence of sharp spectral features. While separate parts of the resulting optical response can be explained in terms of different, appropriately chosen superposition basis, their simultaneous presence and hence the ''double-dark'' resonance structure as a whole is a definite signature of a type of quantum interference effect. The phenomenon of interfering double-dark states is very general and occurs in a broad class of multistate systems. The effect can be induced, for example, by a microwave field driving a magnetic dipole transition, by optical fields inducing multiple two-photon transitions, by a static field, or by a nonadiabatic coupling mechanism in time-dependent laser fields [10].

We show that the resonances associated with the double-dark states can be made absorptive or transparent and their optical properties such as width and position can be manipulated by adjusting the coherent interaction. Furthermore, a very weak incoherent excitation of the atoms can be sufficient to turn the absorption into optical gain. We anticipate that such ''designed'' atomic response can be of interest in areas as diverse as enhancement of optical activity in dense media, high-resolution spectroscopy, quantum well lasers, and Raman adiabatic passage.

The basic physical mechanism leading to the response can be understood by considering the generic four-state system of Fig. 1(a). Here, a resonant driving field and a weak probe field with Rabi-frequencies Ω and $\mathcal{E}$ couple two lower metastable states c and b with upper level a and, therefore, form a simple Λ configuration. The resulting dark state is coherently coupled by a real or effective coherent field with Rabi-frequency Ω_c to another metastable state d. As noted above, a variety of different mechanisms can cause this coupling. This model is quite general since it is unitary equivalent to a broad class of other four-state systems, some of which are shown in Fig. 1. All of these schemes are described by the identical semiclassical dressed-state picture [Fig. 1(d)], which provides useful insight into the origin of the interference between double-dark resonances.

Let us begin with the system of Fig. 1(a), in which all coherent processes are described—within the rotating wave approximation—by the following Hamiltonian matrix:

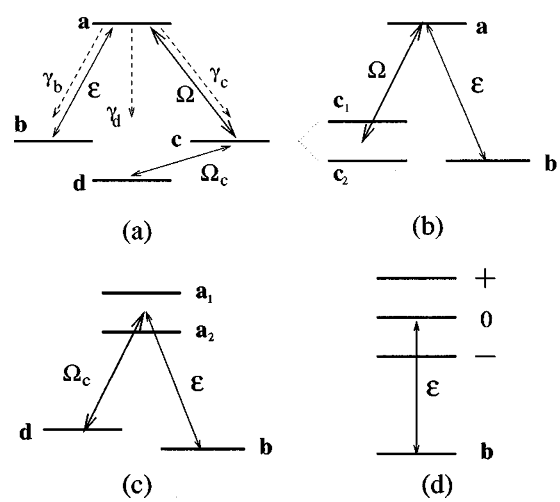

FIG. 1. (a) Four-state system displaying double-dark resonances. Unitary equivalent systems: (b) corresponds to generic model (a) after diagonalizing the interaction with the coherent perturbation Ω_c [states $|c_1\rangle$ and $|c_2\rangle$ correspond to $(|c\rangle \pm |d\rangle)/\sqrt{2}$, respectively], (c) after diagonalizing the interaction with the drive field Ω [states $|a_1\rangle$ and $|a_2\rangle$ correspond to a pair $(|a\rangle \pm |c\rangle)/\sqrt{2}$ displaying Fano interference due to spontaneous emission], and (d) dressed-state picture corresponding to the (a), (b), and (c).

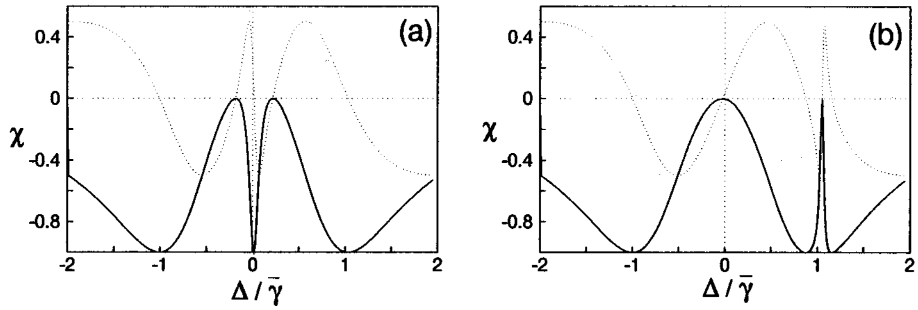

FIG. 2. Imaginary (solid lines) and real (dashed lines) parts of the probe susceptibility in the units of $\eta/\overline{\gamma}$ for a system of Fig. 1(a). Parameters are $\gamma_b=\gamma_c=\gamma_d=\overline{\gamma}$, $\Omega=\overline{\gamma}$, $\Omega_c=0.2\overline{\gamma}$, $r=\gamma_0=0$, and $\Delta_0=0$. (a) $\Delta_c=0$. (b) $\Delta_c=\overline{\gamma}$. Case (b) is the special case when the dressed energies intersect $[E(|-\rangle)\sim E(|0\rangle)]$.

$$\mathcal{H}_0=-\hbar\begin{pmatrix}\Delta_0 & \Omega & 0 & \mathcal{E}\\ \Omega & 0 & \Omega_c & 0\\ 0 & \Omega_c & -\Delta_c & 0\\ \mathcal{E} & 0 & 0 & \Delta_0-\Delta\end{pmatrix}, \tag{1}$$

corresponding to the vector of state amplitudes (c_a,c_c,c_d,c_b), $(c_\mu:=\langle\mu|\psi\rangle)$. Here $\Delta=\nu_p-\omega_{ab}$, $\Delta_0=\nu-\omega_{ac}$, and $\Delta_c=\nu_c-\omega_{cd}$ are the detunings of the probe field, the drive field, and the coherent perturbation. The three dressed sublevels, generated by the driving fields Ω and Ω_c, and the corresponding frequencies read to first order in Ω_c,

$$|+\rangle=\frac{1}{\Omega_0}\left[-\omega_+|a\rangle+\Omega\left(|c\rangle+\frac{\Omega_c}{\omega_0-\omega_+}|d\rangle\right)\right], \tag{2}$$

$$|-\rangle=\frac{1}{\Omega_0}\left[\Omega|a\rangle+\omega_+\left(|c\rangle+\frac{\Omega_c}{\omega_0-\omega_-}|d\rangle\right)\right], \tag{3}$$

$$|0\rangle=|d\rangle-\frac{\Omega_c\Omega}{\tilde{\Omega}^2}|a\rangle+\frac{\Omega_c(\Delta_c+\Delta_0)}{\tilde{\Omega}^2}|c\rangle, \tag{4}$$

$$\omega_\pm=-\frac{\Delta_0}{2}\mp\sqrt{\Omega^2+\frac{\Delta_0^2}{4}}, \tag{5}$$

$$\omega_0=\Delta_c, \tag{6}$$

where $\Omega_0^2:=\Omega^2+(\Delta_0/2+\sqrt{\Omega^2+\Delta_0^2/4})^2$, and we have assumed a sufficiently large splitting between the dressed-state energies such that $|\tilde{\Omega}^2|:=|\Omega^2-\Delta_c(\Delta_c+\Delta_0)|\gg\Omega_c^2$. Two of the dressed states $|\pm\rangle$ correspond, in the limit of vanishing perturbation $\Omega_c\to 0$, to the usual Autler-Townes dressed components split by $2\sqrt{\Omega^2+\Delta_0^2/4}$. Since both have a finite overlap with the excited state $|a\rangle$, there is quantum interference in the absorption or spontaneous emission on the probe transition leading to a single dark resonance. The third dressed state $|0\rangle$ coincides in this limit with the bare state $|d\rangle$ and hence is decoupled from the system. This is no longer so in the presence of a second drive field Ω_c. In this case the dressed state $|0\rangle$ contains an admixture of $|a\rangle$ and thus has a nonzero dipole matrix element with ground state $|b\rangle$. From this coupling result transitions between $|b\rangle$ and $|0\rangle$ corresponding to three-photon hyper-Raman resonances as well as additional interference effects. The latter lead to a pair of transparency points, as shown in Fig. 2(a).

Alternatively, the system can be analyzed by diagonalizing the interaction with the coherent coupling (Ω_c), which leads to the system shown in Fig. 1(b). It corresponds to a four-level system with two drive fields of Rabi-frequency $\Omega/\sqrt{2}$ forming two different Λ subsystems with lower states $|c_{1,2}\rangle$ split by $\pm\Omega_c$. In such a system, the existence of two distinct dark resonances is clear at hand, each corresponding to a two-photon resonance between $|b\rangle$ and states $|c_1\rangle$ and $|c_2\rangle$, respectively. In this basis the central narrow structure is due to interference induced by the coherent interaction between the two dark states.

Hence, in either basis quantum interference is an essential feature of interacting dark resonances. This quantum interference combined with the possibility of tuning the position of the state $|0\rangle$ and adjusting its coupling strength allows one to ''engineer'' the atomic response. We now discuss this in more detail.

To quantify the properties of double-dark resonances we examine the response of the system [Fig. 1(a)] using the full set of density-matrix equations. We assume a weak probe field, and begin with the case when all atoms are in ground-state b. We also take into account transit-time broadening with a corresponding rate γ_0. The linear susceptibility is then given by

$$\chi=\frac{i\eta\,\Gamma_{cb}}{\Gamma_{ab}\Gamma_{cb}+\Omega^2}\left(1+\frac{\Omega^2}{\Gamma_{cb}}\,\frac{\Omega_c^2}{\Gamma_{ab}(\Gamma_{cb}\Gamma_{db}+\Omega_c^2)+\Omega^2\Gamma_{db}}\right), \tag{7}$$

where $\eta=\gamma_b 3\mathcal{N}\lambda^3/(8\pi^2)$, $\mathcal{N}$ is the atomic density, and $\wp$ is the dipole matrix element of the probe transition. γ_{ij} are the relaxation rates of the respective coherences, and $\Gamma_{ab}=\gamma_{ab}+i\Delta$, $\Gamma_{cb}=\gamma_{cb}+i(\Delta-\Delta_0)$, and $\Gamma_{db}=\gamma_{db}+i(\Delta-\Delta_0-\Delta_c)$.

It is instructive to first examine the case of infinitely long-lived lower-level coherence, $\gamma_0\to 0$. Figure 2 shows typical susceptibility spectra in the case of radiative broadening for the system with weak coherent perturbation of the dark state. We note that the original dark resonance is split into a pair of dark lines. Indeed from Eq. (7) one finds that the probe susceptibility vanishes at the two frequencies:

$$\Delta=\Delta_0+\frac{\Delta_c}{2}\pm\sqrt{\left(\frac{\Delta_c}{2}\right)^2+\Omega_c^2}, \tag{8}$$

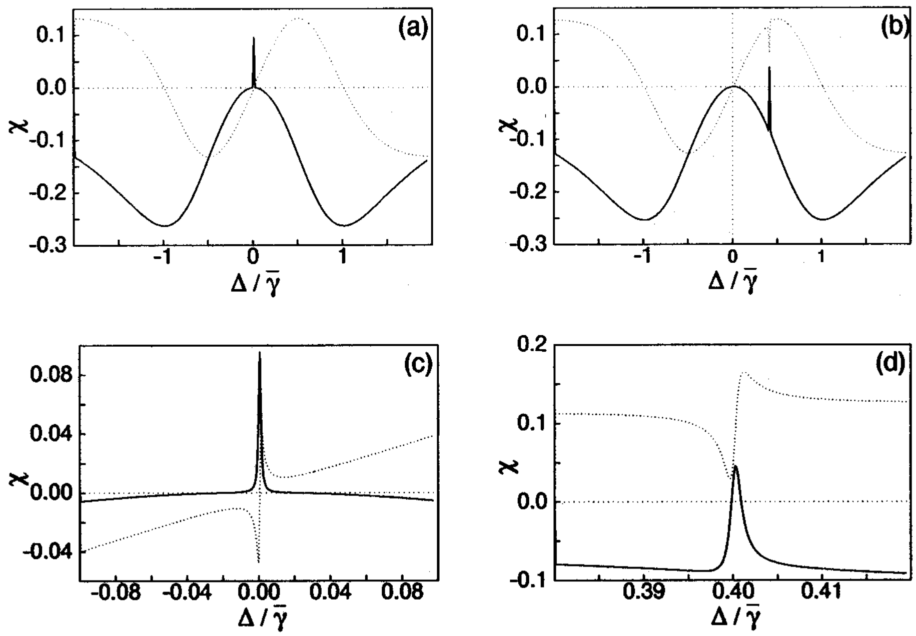

FIG. 3. Imaginary (solid lines) and real (dashed lines) part of the susceptibility in the units of $\eta/\overline{\gamma}$ for (a) and (c) optical gain and (b) and (d) enhanced index of refraction without absorption. The parameters are $\gamma_b=\gamma_c=\gamma_d=\overline{\gamma}$, $\Omega=\overline{\gamma}$, $\Omega_c=0.01\overline{\gamma}$, $\gamma_0=3r_b=3r_c=3r_d=0.0001\overline{\gamma}$, and $r=0.01\overline{\gamma}$; $\Delta_c=0$ for (a) and (c) and $r=0.005\overline{\gamma}$, $\Delta_c=0.45\overline{\gamma}$ for (b) and (d).

i.e., coherent perturbation does not simply eliminate the dark resonance, but rather splits it into two. These two transparency points correspond, for resonant coherent fields, to the dark states of the two Λ subsystems of Fig. 1(b). In addition, a narrow feature emerges, which is superimposed on the original transparency line. It is represented by the second term on the right-hand side of Eq. (7). In the case of a sufficiently strong driving field such that $|\tilde{\Omega}^2|\gg\Omega_c^2$, this feature has an approximately Lorentzian line shape with line center and width given by

$$\overline{\Delta}\equiv\frac{\Delta_c+\Delta_0}{1+\Omega_c^2/\tilde{\Omega}^2},\qquad \Gamma_0=\gamma_{ab}\frac{\Omega^2}{|\tilde{\Omega}^2|}\frac{2\Omega_c^2}{|\tilde{\Omega}^2+\gamma_{ab}\Delta_c|},\qquad (9)$$

where $|\tilde{\Omega}^2+\gamma_{ab}\Delta_c|\gg\Omega_c^2$ is assumed. At $\Delta=\overline{\Delta}$ the susceptibility scales like $\chi=\eta/\gamma_{ab}$. That means that the amplitude of the resonances created by the coherent perturbation is of the same order as that of a fully resonant two-level absorber. The position and width of the absorption line can be manipulated by tuning (Δ_c) and varying the strength (Ω_c) of the coherent perturbation.

The interference nature of the effect becomes most profound for $\Omega^2\approx\Delta_c(\Delta_c+\Delta_0)$, i.e., when the energy of one of the dressed states $|+\rangle$ or $|-\rangle$ approaches the energy of dressed state $|0\rangle$. In this case the feature turns from a sharp Lorentzian absorption line into a transparency line of width $\sim 2\Omega_c^2(\Delta_0+\Delta_c)/(\gamma_{ab}\Delta_c)$ close to the point of three-photon resonance [see Fig. 2(b)]. Hence, by proper tuning of the coherent perturbation the multiphoton processes can be either resonantly enhanced or completely eliminated. We emphasize that the ultimate limit for the widths of the described high-contrast spectral features in the limit of small Rabi frequency Ω_c is determined by the finite relaxation rate of the long-lived coherences between the metastable states. It can, therefore, be extremely small compared to the width of the optical transition [11–13].

We now extend our treatment to include the case when some atoms are injected into one of the levels coupled by the coherent perturbation, specifically into the state $|d\rangle$. In a dressed-state picture such injection implies the selective population of the dressed state $|0\rangle$, which may result in optical gain or in an increase of the refractive index at the transparency point. In the following we focus on the case where the atoms are excited by a weak incoherent pump rate r. Let us consider, for example, the case when all fields are on resonance [i.e., $\Delta_0=\Delta_c=\Delta=0$, see Fig. 3(a)]. Assuming, additionally, a weak coherent perturbation and small ground-state relaxation ($\gamma_0\ll|\Omega_c|, r\ll\gamma_{\rm rad}$) we find that the absorptive feature is turning into gain when

$$r\geq\frac{\gamma_b}{\gamma_d}\left|\frac{\Omega_c}{\Omega}\right|^2\gamma_a+\left(1+\frac{\gamma_b}{\gamma_d}\right)\gamma_0.\qquad (10)$$

Since $|\Omega_c/\Omega|^2$ can be made very small, an incoherent pump strength orders of magnitude smaller than necessary to invert the optical transition is sufficient to produce gain. It is interesting to note that the upper level population in this case is very small ($\rho_{aa}^{(0)}\sim r/\gamma_b$). Furthermore, double-dark lines can be used to create a medium with an enhanced refractive index without absorption. To this end, it is favorable to produce a double-dark line at a frequency where the refractive index is large in the absence of coherent perturbation, i.e., in the vicinity of the dressed states $|\pm\rangle$. This can be readily achieved by tuning the coherent perturbation [see Fig. 3(b)].

Hence, the above considerations allow us to conclude that using coherently coupled double-dark resonances, it is possible to efficiently ''engineer'' the atomic response. We now focus on some of the applications of such designed atomic response. Consider, first of all, the problem of optical activity enhancement in a dense medium, and in particular, the enhancement of the refractive index without absorption. One of the major obstacles in the realization of a large refractive index (i.e., of susceptibility χ' comparable to unity) in usual schemes [5,6] is the requirement of a large excited-state population density. Excited-state population and the corresponding energy dissipation due to spontaneous emission represent an important limitation to the optical activity enhancement in a dense, partially excited ensemble, as they lead to the requirement of large continuous energy input, absorption of coherence generating fields, superradiant decays, frequency shifts, and other decoherence effects. The potential advantage of using double-dark lines is that large susceptibility values can be achieved in dense media avoiding the above-mentioned problems. We note, for example, that in the case depicted in Figs. 3(b) and 3(d), the refractive index at the point of vanishing absorption is comparable to the maximum value obtained in a two-level system in the vicinity of an atomic resonance. It is obtained with a very small excitation of atoms and correspondingly small energy dissipation. The parameters used for the present simulations correspond to a possible realization of double-dark resonances within the Rb D_1 absorption line using hyperfine and Zeeman sublevels of the ground state. Here a pair of optical fields can be used together with an rf or a microwave field to generate a double-dark line. The relaxation rate between metastable lower levels in such a system can easily be within a few kHz, being limited, in a dense medium, only by the very slow dephasing due to spin-exchange collisions [15]. In this case, atomic densities up to $10^{15}-10^{16}$ cm^{-3} can be used and a few centimeters of transparent Rb vapor with refractivity $\chi' \sim 1$ can be created [15,16]. This can be put in prospective by noting that resonant $\chi' \sim 10^{-4}$ was observed in the experiment of Ref. [6] utilizing a Λ-type EIT scheme in Rb.

Furthermore, the narrow features associated with double-dark resonances can be of interest in high-resolution laser spectroscopy. They can provide a sensitive tool for direct measurements of, e.g., strength of the coherent perturbation such as magnetic fields. High sensitivity can be expected similar to EIT-based techniques [14] but without the need for involved dispersive measurements, since narrow double-dark resonances can be observed with a large signal-to-noise ratio. Indeed, high-contrast narrow features persist even in the presence of strong optical fields and are not limited by power broadening.

Another interesting application of double-dark resonances are unipolar and bipolar quantum well lasers. Here the properties of double-dark resonances can provide a way to mitigate the problems associated with large inhomogeneous broadening [19].

Finally, the possibility of using double-dark states in adiabatic passage techniques [7] is intriguing, in that it offers a way of a robust preparation and phase-sensitive probing of arbitrary quantum superpositions of lower states [17], which is of particular interest for quantum computation [18]. This, as well as other applications of double-dark lines will be addressed in detail elsewhere.

The authors gratefully acknowledge useful discussions with L. Hollberg, K. Bergmann, E. Fry, P. Hemmer, Yu. Rostovtsev, R. Pfund, V. Sautenkov, B. W. Shore, R. Unanyan, V. Velichansky, and A. Zibrov, and the support from the U.S. Office of Naval Research, the Welch Foundation, the Texas Advanced Research and Technology Program, the National Science Foundation, and the U.S. Air Force Laboratories. M.F. thanks the Alexander-von-Humboldt Foundation and S.F.Y. thanks the ''Studienstiftung des Deutschen Volkes'' for financial support.

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[15] See, e.g., J. Vanier and C. Audoin, *The Quantum Physics of Atomic Frequency Standards* (A. Hilger, Bristol, 1989); We note that a careful choice of hyperfine and Zeeman sublevels should be made in order to minimize the effect of spin-exchange interaction, which preserves the total spin, e.g., Happer and Tang [Phys. Rev. Lett. **31**, 273 (1973)] observed narrow ($\sim$200 Hz) resonances at densities $>10^{14}$ cm^{-3}. These interactions as well as Doppler and collisional broadening of the optical polarizations have here been taken into account for the estimates of refractivity.
[16] We further note that although the atomic response of the different systems in Fig. 1 on the probe field is similar, the propagation dynamics of the other fields can differ. In particular, a dense atomic ensemble can give rise to a nonlinear generation of fields components, which can significantly modify double-dark lines.
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