Atomic coherence effects in four-level systems: Doppler-free absorption within an electromagnetically-induced-transparency window

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(Received 9 August 1996)

We report here an effect in a four-level ladderlike system, which is in contrast to the usual quantum interference effects such as electromagnetically induced transperency (EIT) or coherent population trapping: we predict the occurrence of a narrow absorption peak within the EIT window when an EIT atomic system interacts with an additional driving rf field. The Doppler-free-central absorption appears when the three-photon resonance condition is satisfied. In the limit of the rf field strength $\Omega_{rf}\rightarrow 0$, the usual EIT profile is recovered.

PACS number(s): 42.50.Hz, 32.80.Wr, 33.80.Wz

Most quantum interference and atomic coherence effects have been observed either as dark bands in resonance flourescence [1,2] or as reduction in absorption [3,4], commonly referred to as electromagnetically induced transperancy (EIT). Modification of the spontaneous emission line shapes due to such interference effects has also been predicted [5]. Effects such as coherent population trapping (CPT) [6], EIT [3,4], lasing without inversion (LWI) [7,8], and large refractive index at zero absorption [9] have been studied extensively both at steady state [2,6] and under pulsed conditions [10]. Almost all the studies on EIT and CPT have been confined to three-level systems with a few theoretical generalizations, in the context of CPT, to n-level systems interacting with $n-1$ driving fields [11–13]. In such situations also, the CPT effects persist. A number of theories have been proposed [14] to explain the physical mechanisms behind these phenomena; the occurrence of the dark band or the dip in the absorption spectrum is generally associated with the destructive interference between the two absorption amplitudes [2,14]. One may also understand these phenomena in terms of the coherence between the atomic levels induced by the combined interaction of the two driving fields with the atomic system [4].

In this paper we report theoretical results in a four-level system that are contrary to the usual CPT and EIT effects. A weak optical probe interacting with such a system in the presence of a strong (optical) pump and an rf field shows a narrow absorption in an otherwise transparent (EIT) background. This absorption occurs when the three-photon resonance condition is satisfied. In the limit of the strength of the rf field (Ω_{rf}) approaching zero, we recover the standard EIT profile. In analogy with the three level system, the appearance of the central absorption line can be attributed to the coherence induced among the atomic levels by the interaction of the probe (Ω_p), the pump (Ω_c), and the rf field (Ω_{rf}) with the atomic system. The central absorption peak reported here shows a linewidth which is of the order of the natural linewidth of the 1-2 transition. This scheme, therefore, provides another convenient technique for high-resolution Doppler-free spectroscopy. Such high-resolution effects have been reported earlier in the context of three-level systems, where the EIT windows showed subnatural linewidths [15]. In the present case, however, the EIT window provides a flat background against which the sharp absorption peak, predicted here, can be recorded. This can be achieved by using a strong pump field, with low atomic density, to get a broad EIT window.

The system we consider is a four-level, ladderlike system schematically shown in Fig. 1. We assume that the only allowed dipole transitions are 1-2, 2-3, and 3-4. It is further assumed that the levels 3 and 4 are closely spaced. The probe beam of angular frequency ω_p connects the levels 1 and 2, the strong pumping field ω_c couples the levels 2 and 3, and the third field ω_{rf} connects the levels 3 and 4. In the absence

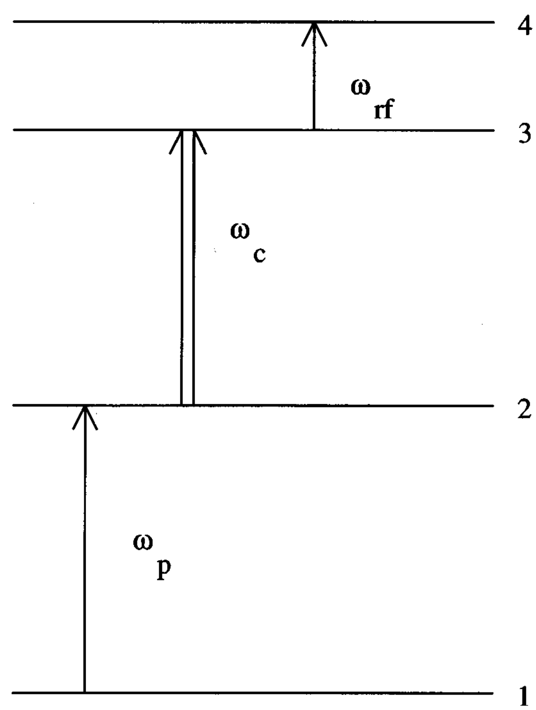

FIG. 1. Schematic energy-level diagram of the four-level atomic system. ω_c, ω_p, and ω_{rf} are, respectively, the frequencies of the pump, the tunable probe, and the rf field in the laboratory frame.

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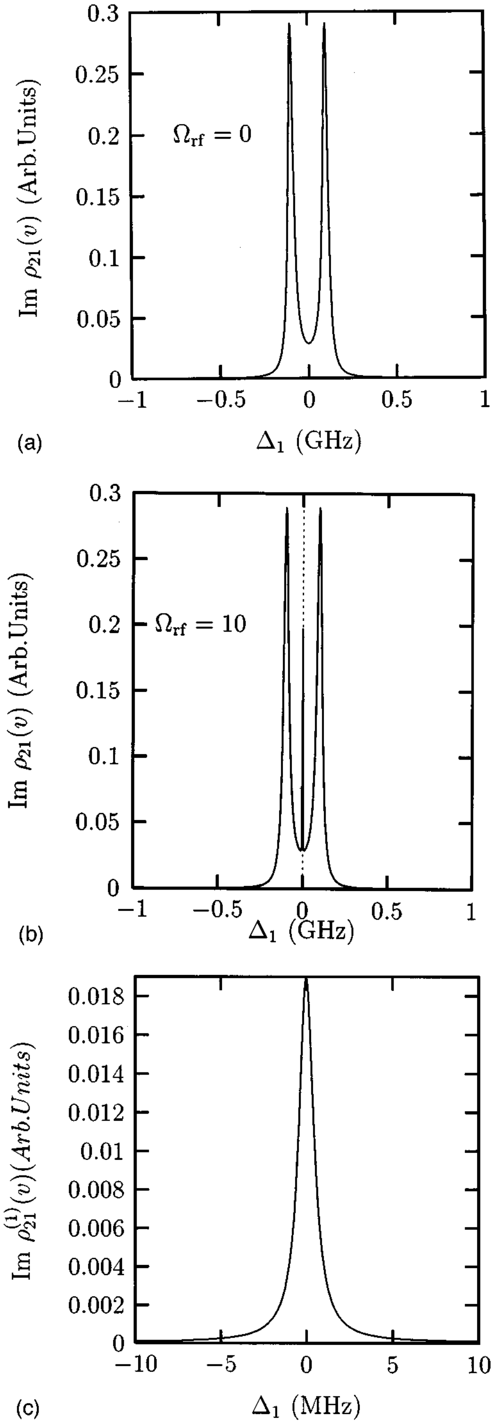

FIG. 2. (a) $\mathrm{Im}\rho_{21}(v)$ versus the detuning Δ_1 of the probe field for the four-level ladder system. The parameters used are $\Delta_2=0$, $\Delta_3=0$, $\gamma_{21}=3$ MHz, $\gamma_{31}=\Omega_{41}=0.5$ MHz, $\gamma_{32}=\gamma_{42}=3.5$ MHz, $\Omega_p=10$ MHz, and $\Omega_c=100$ MHz. Note the usual EIT feature for $\Omega_{rf}=0$. (b) Doppler-free absorption peak within the EIT window for $\Omega_{rf}=10$ MHz at $\Delta_1=0$. All the parameters are the same as in (a). (c) Details of the absorption peak in (b). All the parameters are the same as in (a).

of level 4 the system is exactly like the Rb three-level system of Ref. [4]. In fact, all the relaxation parameters of levels 1, 2, and 3 and their energy separations used in the present calculations correspond to those of the rubidium system. The 3-4 transition could, in principle, be between any of the hyperfine components of level 3. The pump and the probe beams are taken to be counterpropagating and the transitions 1-2 and 2-3 are assumed to be of almost the same energy. The 3-4 transition being in the rf region does not suffer from any significant Doppler shift. We extend the set of Eq. (1) in Ref. [4] to our four level system by including the additional dipole transition between 3 and 4 and get

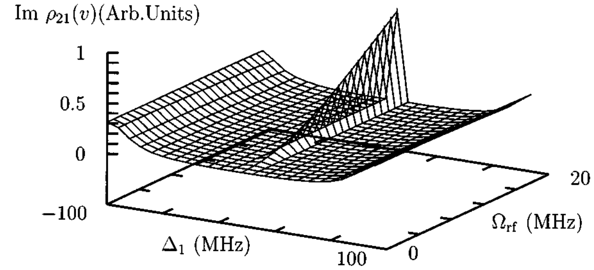

FIG. 3. Growth of $\mathrm{Im}\rho_{21}(v)$ with Ω_{rf} and varying probe detuning Δ_1. All the other parameters are the same as in Fig. 2.

$$\dot{\rho}_{21}=-(\gamma_{21}-i\Delta_1)\rho_{21}+i\Omega_p(\rho_{22}-\rho_{11})-i\Omega_c\rho_{31}, \quad (1)$$

$$\dot{\rho}_{32}=-(\gamma_{32}-i\Delta_2)\rho_{32}+i\Omega_c(\rho_{33}-\rho_{22})+i\Omega_p\rho_{31}-i\Omega_{rf}\rho_{42}, \quad (2)$$

$$\dot{\rho}_{31}=-[\gamma_{31}-i(\Delta_1+\Delta_2)]\rho_{31}-i\Omega_c\rho_{21}+i\Omega_p\rho_{32}-i\Omega_{rf}\rho_{41}, \quad (3)$$

$$\dot{\rho}_{41}=-[\gamma_{41}-i(\Delta_1+\Delta_2+\Delta_3)]\rho_{41}-i\Omega_{rf}\rho_{31}+i\Omega_p\rho_{42}, \quad (4)$$

$$\dot{\rho}_{42}=-[\gamma_{42}-i(\Delta_3+\Delta_2)]\rho_{42}-i\Omega_{rf}\rho_{32}+i\Omega_c\rho_{43}+i\Omega_p\rho_{41}, \quad (5)$$

$$\dot{\rho}_{43}=-(\gamma_{43}-i\Delta_3)\rho_{43}-i\Omega_{rf}(\rho_{33}-\rho_{44})+i\Omega_c\rho_{42}, \quad (6)$$

where we have assumed the Rabi frequencies of the pump (Ω_c), the probe (Ω_p), and the rf transition (Ω_{rf}) to be real; Δ_1, Δ_2, and Δ_3 represent the detunings of the probe, the pump, and the rf fields from their respective atomic transitions. The dephasing rate γ_{ij} in the absence of collisions is given by $(\Gamma_i+\Gamma_j)/2$, Γ_i being the decay rate of the ith level. We have obtained analytical solutions for the set of equations (1)–(6) in the steady state using MATHEMATICA. In the weak field limit of the probe, its absorption coefficient is calculated up to the first order in the probe field strength Ω_p. A similar analysis has been done earlier by Hansch and Toschek [16] for three-level systems. The probe absorption, which is proportional to the imaginary part of $\rho21^{(1)}$, is given by

$$\mathrm{Im}[\rho_{21}^{(1)}(v)]=\mathrm{Re}\left[\frac{(L_{12}+\Omega_{rf}^2/L_{123})\Omega_p}{(\Omega_c^2+L_1L_{12}+\Omega_{rf}^2L_1/L_{123})}\right],$$

$$L_1=(\gamma_{21}-i\Delta_1), \quad L_{12}=[\gamma_{31}-i(\Delta_2+\Delta_1)],$$

$$L_{123}=[\gamma_{41}-i(\Delta_1+\Delta_2+\Delta_3)], \quad (7)$$

where we have further assumed $\rho_{11}=1$ and $\rho_{ii}=0$ for $i=2$, 3, and 4. For $\Omega_{rf}\rightarrow 0$ we get the usual expression for the EIT

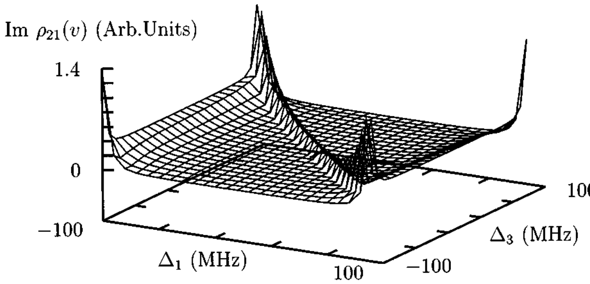

FIG. 4. Im$\rho_{21}(v)$ as a function of the detunings Δ_1 and Δ_3 with $\Delta_2=0$. $\Omega_{rf}=10$ MHz and all the rest of the parameters are the same as in Fig. 2. The Doppler-free absorption occurs when the three photon resonance condition $\Delta_1+\Delta_2+\Delta_3=0$ is satisfied.

profile for the three-level ladder system [4]. Variation of Im $[\rho_{21}^{(1)}(v)]$ as a function of probe detuning for $\Omega_{rf}=0$ and 10 MHz is shown in Fig. 2. We note the appearance of a sharp absorption peak in the middle of the EIT window for $\Omega_{rf}=10$ MHz [Fig. 2(b)]. This narrow peak has a full width at half maximum (FWHM) ≈ 1.5 MHz and is shown in Fig. 2(c). Figure 3 is a three-dimensional plot showing the growth of this narrow absorption within the EIT window, with increasing Ω_{rf} and varying detuning Δ_1. In Fig. 4 we plot the probe absorption as a function of the detunings, Δ_1 and Δ_3, while Δ_2 is kept fixed at zero. The narrow absorption peak appears only when the condition $\Delta_1+\Delta_3=0$ is satisfied, in addition to Δ_2 being zero, which clearly indicates that it is a three-photon resonance effect. The contribution to the probe absorption from all velocity groups is obtained by integrating Eq. (7) over the Maxwellian velocity distribution given by $N(v) = (N_0/u\sqrt{\pi})\exp^{[-v^2/u^2]}$; N_0 is the atomic density and $u/\sqrt{2}$ is the rms atomic velocity, so that

$$\mathrm{Im}[\rho_{21}] = \frac{N_0}{u\sqrt{\pi}}\int \mathrm{Im}[\rho_{21}(v)]\exp(-v^2/u^2)dv. \tag{8}$$

The Doppler width for the 1-2 transition is taken to be 540 MHz. The Doppler shifts in the pump and the counterpropagating probe frequencies are assumed to cancel exactly and the Doppler shift in the rf field (in the atomic rest frame) is neglected. We have performed this integration numerically and present the results in Fig. 5. The width (FWHM) of the

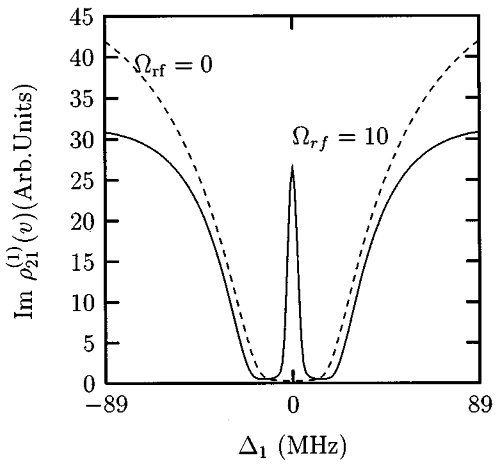

FIG. 5. Doppler averaged Imρ_{21} as a function of the probe detuning Δ_1. $\Delta_2=\Delta_3=0$. (All other parameters are the same.) The FWHM of the central peak ≈ 6 MHz.

velocity integrated central absorption is of the order of the natural linewidth of the 1-2 transition. On the other hand, when the 3-4 transition is in the optical region we find that the Doppler averaging masks the central absorption almost completely, for either direction of propagation of the additional optical beam. Thus, only on the application of an rf field between the hyperfine levels of level 3 does one get a Doppler-free absorption of the probe.

It is interesting to see if the central absorption peak shows any homogeneous broadening, such as power broadening, when the power of the probe beam is increased. In this limit, however, the set of equations (1)–(6) would become insufficient to describe the population dynamics. We would instead need to solve the full set of equations involving both the diagonal and the off-diagonal matrix elements of the density matrix. This is being carried out numerically. It would also be interesting to see if in the Λ-like four-level systems there is a release of trapped states due to the presence of an additional field in an otherwise trapped system. These problems are being studied at present.

We thank Professor S. P. Tewari for useful discussions and Dr. K. P. Rajeev for providing computational facilities. S.N.S. wishes to thank the Department of Science and Technology, India for financial assistance.

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