Absorption resonance and large negative delay in rubidium vapor with a buffer gas

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We observe a narrow, isolated, two-photon absorption resonance in ^{87}Rb for large one-photon detuning in the presence of a buffer gas. In the absence of a buffer gas, a standard Λ configuration of two laser frequencies gives rise to electromagnetically induced transparency (EIT) for all values of one-photon detuning throughout the inhomogeneously (Doppler) broadened line. However, when a buffer gas is added and the one-photon detuning is comparable to or greater than the Doppler width, an absorption resonance appears instead of the usual EIT resonance. We also observe a large negative group delay (≈ -300 μs for a Gaussian pulse that propagates through the media with respect to a reference pulse not affected by the media), corresponding to a superluminal group velocity $v_g = -c/(3.6 \times 10^6) = -84$ m/s.

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Quantum coherence effects have attracted great attention in the past few years. Phenomena such as ultralow optical group velocity,[1–4] superluminal group velocity,[4–6] enhanced nonlinear optical effects,[7] and quantum information storage[8–10] have all been studied under the condition of electromagnetically induced transparency (EIT).[11] More recently, complementary coherence effects such as electromagnetically induced absorption (EIA) have been predicted and studied.[6,12–14] Here we present the experimental observation of a new narrow absorption resonance that appears at large optical density and large detuning in Doppler-broadened media with a buffer gas.

In typical experiments, the narrow transmission linewidth in EIT is limited by the relaxation rate of ground-state coherence, which is usually determined by the interaction time of an atom with applied laser radiation. Two common methods for increasing the interaction time are by use of wall coatings[15] that allow an atom to maintain its coherence while it travels into and out of the interaction region many times, and by use of a buffer gas that allows the atom to diffuse out of the interaction region slowly by velocity changing collisions that still preserve the ground-state coherence.[16,17]

For a Λ-type EIT system, the dependence of the EIT resonance on one-photon laser detuning has been studied experimentally[18] and theoretically.[19] They show that within the limit of high buffer gas pressure, when the decay rate of the upper level is comparable with the Doppler broadening, the EIT resonance shape can be described by a Lorentzian plus a dispersionlike curve:

$$f(\delta) = \gamma \frac{A\gamma + B(\delta - \delta_0)}{\gamma^2 + (\delta - \delta_0)^2} + C, \qquad (1)$$

where γ is the width of the resonance, δ is the two-photon detuning, δ_0 is the one-photon-dependent resonance shift with respect to resonance position for zero one-photon detuning, A is the amplitude of the Lorentzian part, B is the amplitude of the dispersionlike part, and C is an offset. However, previous experimental studies[18] have only seen the resonance shape become somewhat asymmetric while still maintaining an EIT resonancelike shape, in other words $A > 0$ and $|B/A| < 2$.

In our experiment we achieved an absorptionlike resonance ($A < 0$) by detuning the drive laser into the wings of the Doppler distribution in a cell with buffer gas. For our conditions, no absorption was found without a buffer gas. The absorption resonance is accompanied by large anomalous dispersion so that negative group delay is observed for a pulse that propagates through the medium. It is important to stress that the previous observation of EIA in Refs. 6 and 12–14 and the enhanced absorption seen in the Hanle effect[20] have a different nature than what we observe here, since those previous observations require that the degeneracy of the ground state be lower than that of the excited state,[13] i.e., $F < F'$, for a drive field transition, which is not necessary in our experiments.

We used a weak probe and a strong drive (or coupling) field in a Λ configuration of the two ground-state levels $5S_{1/2}F = 1, 2$ and the excited $5P_{1/2}F = 2$ state of ^{87}Rb as shown in Fig. 1. We operate in the power-broadened regime in which $\Omega_d > \sqrt{\gamma\gamma_{bc}}$, where Ω_d is the drive laser Rabi frequency, γ is the radiative decay rate as defined above, and γ_{bc} is the decay rate of ground-state coherence. The resulting ground-state coherence gives rise to a narrow EIT resonance of the probe field in the vicinity of two-photon resonance ($\delta = 0$). This coherence still exists even for one-photon detunings (Δ) comparable with or somewhat greater than the inhomogeneous Doppler width of the medium.

Because of the way our probe field is generated (discussed below), a second Λ configuration consisting of a weak Stokes component and a drive field (see Fig. 1) is also present. However, this system is detuned far from resonance and does not significantly affect our system. (Nonetheless, the propagation properties of the Stokes

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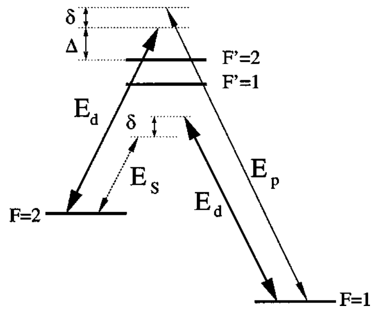

Fig. 1. Three-level interaction scheme of three laser fields with ^{87}Rb atoms. The long-lived coherence is created on the hyperfine split ground-state sublevels with a strong driving field E_d and a weak probe field E_p. A weak Stokes field E_s is also present as a by-product of the generation of the probe field. Δ is the one-photon detuning of the drive and the probe lasers from their respective atomic transitions, and δ is the two-photon detuning, which is scanned.

component are rather interesting and will be discussed elsewhere; see on-line paper at http://arxiv.org/abs/quant-ph/0309171.)

Our experimental setup is shown schematically in Fig. 2. We used an external cavity diode laser tuned to the vicinity of the $5S_{1/2} \rightarrow 5P_{1/2}(D_1)$ transition of ^{87}Rb. The laser is referred to as the driving field (E_d) in Fig. 1. Detuning of the drive laser (Δ) changes from 0 to 2 GHz and is always positive (for any $\Delta > 0$ detuning from upper levels $F' = 1, 2$ is larger then for zero detuning as shown in Fig. 1). Sidebands are generated on the drive laser by an electro-optic modulator (EOM), which is tunable in the vicinity of the 6.835-GHz ground-state splitting. The drive laser is tuned to the $F = 2 \rightarrow F' = 2$ transition so that the upper sideband serves as the probe field and is tunable in the vicinity of the $F = 1 \rightarrow F' = 2$ transition. The lower sideband (Stokes component) is far off resonance. After passing through the EOM, all fields are focused into single-mode optical fiber to obtain a clean Gaussian spatial mode. The laser is collimated to a diameter of 7 mm and circularly polarized with a $\lambda/4$ wave plate right before the cell. The cell itself is surrounded by a three-layer magnetic shield that suppresses the laboratory magnetic field. The cell is heated to 60–70 °C to control the density of ^{87}Rb vapor.

Before entering the EOM, part of the drive laser is split from the main beam, shifted in frequency by a small amount (60 MHz) with an acousto-optic modulator, and deflected around the cell. This shifted beam is recombined with the light transmitted through the cell and all the fields are detected on a fast photodiode. This (heterodyne) procedure allows us to detect the transmitted probe and Stokes fields separately.[2]

We measured the EIT spectrum (transmission as a function of two-photon detuning δ for various one-photon detunings of driving field (Δ). We also measured the group delay of a pulse that propagates through the cell, which was done with a programmable pulse synthesizer that modulates the microwave generator for the EOM. We thus produced a Gaussian (temporal) pulse in the power of the drive field sidebands (the probe field). We extracted the delay time by data taking and analyzing software on a computer that collects the separate probe and reference signals.

We first measured various transmission spectra for an ^{87}Rb cell with no buffer gas. Our measurements in Fig. 3(a) show that the EIT resonance maintains its transmissionlike shape as the drive and probe are detuned from one-photon resonance. When the lasers are far from resonance [bottom trace in Fig. 3(a)], the lasers interact with a very small number of atoms, and the width of the EIT resonance is determined by power broadening with the drive laser (width $\approx \Omega^2/\gamma$, where Ω is the drive Rabi frequency and γ is the Doppler width). When the lasers are on resonance, the effective optical density is much higher, and the EIT width is much lower than the power-broadened width.[21–23]

Next we replace the vacuum cell with a cell with 30 Torr of Ne buffer gas. Results are shown in Fig. 3(b). We find that the resonance spectra for large one-photon detuning (Δ) are changed dramatically from the vacuum case just described. We observe that, as the one-photon detuning increases, the EIT resonance passes through a dispersionlike shape into an absorptionlike shape.

Second, we find that this absorptionlike resonance is accompanied by steep anomalous dispersion that results in a large negative propagation delay of the probe pulse with respect to a reference pulse that is not delayed by the medium. Sample pulses are shown in Fig. 4. These data show a -300-μs delay produced by a 2.5-cm-long cell, implying a superluminal group velocity of $v_g = -84$ m/s.

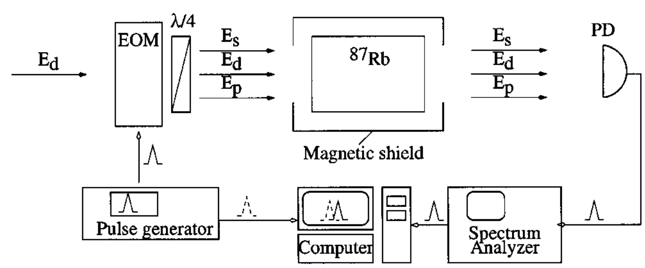

Fig. 2. Schematic of the experimental setup.

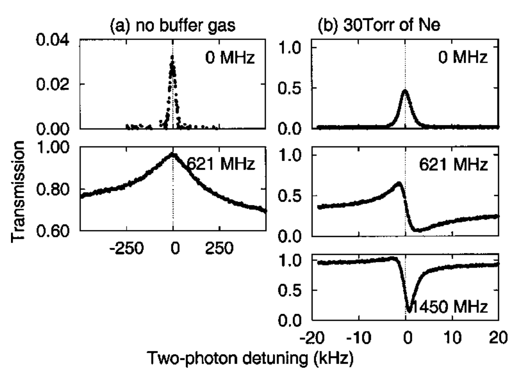

Fig. 3. Transmission of the probe field as a function of two-photon detuning δ (measured in kilohertz) for various one-photon detunings Δ. (a) ^{87}Rb cell with no buffer gas (vacuum) and (b) ^{87}Rb cell with 30 Torr of Ne. The vertical scales are normalized in such a way that 0 corresponds to zero transmission and 1 corresponds to zero absorption (or total transparency). Experimental parameters: (a) $T = 66.4$ °C, $N = 4.2 \times 10^{11}$ cm^{-3}; total power into cell 630 μW, out of cell 40 μW; 4.7-cm cell length. (b) $T = 67.7$ °C, $N = 4.7 \times 10^{11}$ cm^{-3}; total power into cell $\approx$400 μW; 2.5-cm cell length.

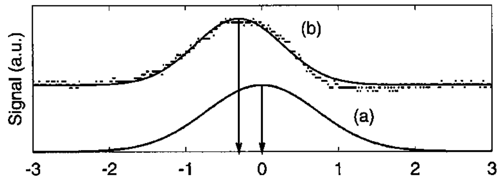

Fig. 4. Demonstration of negative pulse delayed time: (a) the probe field has a Gaussian temporal pulse shape and (b) the transmitted probe field showing a negative time delay before transmission. Experimental conditions are the same as for Fig. 3(b).

To gain physical insight into this new phenomenon we have made numerical simulations based on the density matrix equations coupled with a Maxwell equation description of propagation effects in steady state for low intensity of the probe field. For numerical simulation we use a three-level atomic model coupled with probe and drive fields (Λ scheme).

The density matrix equations are the following:

$$\dot{\rho}_{bb} = i\Omega_p^*\rho_{ab} - i\Omega_p\rho_{ba} + \gamma_r\rho_{aa} - \gamma_{bc}\rho_{bb} + \gamma_{bc}\rho_{cc}, \tag{2}$$

$$\dot{\rho}_{cc} = i\Omega_d^*\rho_{ac} - i\Omega_d\rho_{ca} + \gamma_r\rho_{aa} - \gamma_{bc}\rho_{cc} + \gamma_{bc}\rho_{bb}, \tag{3}$$

$$\dot{\rho}_{ab} = -\Gamma_{ab}\rho_{ab} + i\Omega_p(\rho_{bb} - \rho_{aa}) + i\Omega_d\rho_{cb}, \tag{4}$$

$$\dot{\rho}_{ca} = -\Gamma_{ca}\rho_{ca} + i\Omega_d^*(\rho_{aa} - \rho_{cc}) - i\Omega_p^*\rho_{cb}, \tag{5}$$

$$\dot{\rho}_{cb} = -\Gamma_{cb}\rho_{cb} - i\Omega_p\rho_{ca} + i\Omega_d^*\rho_{ab}, \tag{6}$$

where $\Omega_d = \wp_{ac}E_p/\hbar$ and $\Omega_p = \wp_{ab}E_d/\hbar$ are the Rabi frequencies of the drive and probe fields. The generalized decay rates are defined as

$$\Gamma_{ab} = \gamma + i(\Delta + \delta), \tag{7}$$

$$\Gamma_{ac} = \gamma + i\Delta, \tag{8}$$

$$\Gamma_{cb} = \gamma_{bc} + i\delta. \tag{9}$$

Here $\gamma = \gamma_r + \gamma_p$ is the polarization decay rate, γ_r is the radiative decay rate of the excited state, γ_p is the dephasing rate, and γ_{bc} is the inverse lifetime of the coherence between ground states $|b\rangle$ and $|c\rangle$. Here we note that the presence of a buffer gas affects values of both γ_{bc} and γ. On the one hand, as mentioned above, it allows preservation of the ground-state coherence longer; on the other hand it broadens the optical transition since γ_p grows linearly with buffer gas pressure.[24]

Solving these equations in the steady-state regime and assuming $|\Omega_p| \ll |\Omega_d|$, we obtained the following expression for the linear susceptibility of the probe field:

$$\chi_{ab}(\Delta) = i\eta \frac{\Gamma_{cb}(\rho_{aa}^{(0)} - \rho_{bb}^{(0)}) + \dfrac{|\Omega_d|^2}{\Gamma_{ca}}[\rho_{aa}^{(0)} - \rho_{cc}^{(0)}]}{\Gamma_{ab}\Gamma_{cb} + |\Omega_d|^2}, \tag{10}$$

where $\eta = 3/8\pi N\lambda^2\gamma_r$, N is the ^{87}Rb density, λ is the wavelength of the probe field, and $\rho_{aa}^{(0)}, \rho_{bb}^{(0)}, \rho_{cc}^{(0)}$ are the solutions of the above equations when $\Omega_p = 0$. To take into account propagation and Doppler averaging, the Maxwell equation given by

$$\frac{\partial\Omega_p}{\partial z} = -i\Omega_p \int \chi_{ab}(\Delta + kv)\mathrm{d}v \tag{11}$$

has been solved numerically, where k is the wave number of either the probe or the drive field.

We model the effect of the buffer gas by adding additional homogeneous broadening for optical transitions γ_p using cross sections to broaden[25] and to decrease the time-of-flight limit for the hyperfine coherence decay rate γ_{bc} based on atomic diffusion in the buffer gas.[25] For our simulation of EIT in the absence of buffer gas, we used $\gamma_p = 0$ and $\gamma_{bc}/2\pi = 10$ kHz. For our simulation of the effects of buffer gas, we used $\gamma_p/2\pi = 120$ MHz and $\gamma_{bc}/2\pi = 1$ kHz.[25] The results of these simulations are shown in Fig. 5 and agree well with the experimental results of Fig. 3.

Previous measurements involving one-photon detuning in EIT[18] differ from ours in that low laser power was used on the D_2 line of cesium, which includes a closed cycling transition and also an upper-state hyperfine structure that is covered by the Doppler width. Our numerical model shows the absence of the absorption resonance when an additional homogeneous broadening parameter (γ_p) is small and at low probe and drive intensity. This could be part of the reason the absorption resonance was not observed by Knappe *et al.*[18] Because of the limited sensitivity of our setup we were unable to check this experimentally. The lowest total power for which we have data is 50 μW, which is still in the power-broadened regime.

In summary, we have observed a new, isolated, narrow, two-photon absorption resonance that appears under EIT conditions when a buffer gas is used to narrow the two-photon resonance line and the one-photon detuning is comparable with or greater than the inhomogeneous Doppler linewidth. The effect occurs when homogeneous broadening (due to collisions with buffer gas) is of the order of inhomogeneous (Doppler) broadening, and the effect does not occur in the absence of buffer gas at room temperature. Although we note that Doppler broadening can also be of the order of or even much smaller than the homogeneous broadening for cold atoms, and, in this case, similar absorption resonances should be observable. This technique could provide a new tool to study Bose–

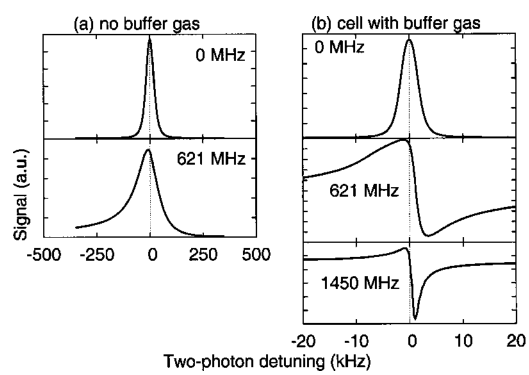

Fig. 5. Calculated transmission of the probe field as a function of two-photon detuning δ (measured in kilohertz) for various one-photon detunings Δ: (a) ^{87}Rb cell with no buffer gas (vacuum) and (b) ^{87}Rb cell with 30 Torr of Ne. The vertical scales are arbitrary.

Einstein condensation. Unlike previous observations of electromagnetically induced absorption, this resonance does not require that the degeneracy of the ground state be lower than that of the excited state. The absorption resonance reported here is accompanied by steep anomalous dispersion giving rise to a large negative group delay.

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