Spectroscopy in Dense Coherent Media: Line Narrowing and Interference Effects

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Spectroscopic properties of coherently prepared, optically dense atomic media are studied experimentally and analyzed theoretically. It is shown that in such media the power broadening of the resonances can be substantially reduced. A density-dependent spectral narrowing of the electromagnetically induced transparency (EIT) window and novel, even narrower, resonances superimposed on the EIT line are observed in dense Rb vapor. A nonlinear two-photon spectroscopic technique based on coherent atomic media and combining high resolution with a large signal-to-noise ratio seems feasible. [S0031-9007(97)04278-6]

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In conventional methods of resonant spectroscopy, optically thin ensembles of atoms or molecules are probed with lasers of limited intensity [1]. Low intensities are used in order to avoid power broadening and frequency shifts. In general, as the Rabi frequency of the resonant electromagnetic field exceeds the natural width of the transition under consideration, the width of the resonance increases, resulting in a rapid loss of resolution. This effect is especially profound when transitions between long-lived states are considered, in which case the necessity of low-intensity light fields results in an unfavorably low signal-to-noise ratio [2]. An example of such a resonance is a two-photon transition in a Λ-type atomic configuration [3].

In the present Letter we study experimentally and describe theoretically the spectroscopic properties of *dense* ensembles of atoms coherently prepared and probed by *strong* optical fields. We observe and discuss the narrowing of power-broadened dark resonances [4,5] associated with electromagnetically induced transparency (EIT). Moreover, when the atomic density becomes sufficiently large, we observe novel, even narrower structures superimposed on the EIT line. We find that these resonances are due to interference induced by a new field component arising from resonantly enhanced coherent Raman scattering [6]. We show that, in contrast to simple transmission measurements, the characteristics of these novel spectral features are determined by the *dispersive* properties of the dense phase-coherent medium [7]. In particular, their width can be orders of magnitude smaller than the usual power-broadened limit; that is, power broadening can be compensated to a large extent. This makes feasible a new regime of two-photon spectroscopy, in which high resolution can be achieved with higher intensities and therefore with lower noise than in conventional methods.

Before proceeding we note that it has already been recognized that sometimes it is advantageous to do spectroscopy in optically thick samples [8]. As will be seen, the essential new features of the present system are twofold: the absorption cancellation due to quantum interference [5] and the importance of the dispersive and nonlinear optical properties of a coherently prepared medium. The present technique is also related to a coherent anti-Stokes Raman spectroscopy (CARS) [6]. Our method, however, differs qualitatively from the latter since in our case it is essential to operate near the single photon resonance. As a result the spectral resolution is determined mostly by the linear susceptibility rather than the $\chi^{(3)}$ nonlinearity. We further note that the advantage of using the dispersion of EIT resonances for spectroscopic measurements has been pointed out in a recent group delay experiment [9]. Our approach is conceptually different in that it utilizes a combination of linear and nonlinear response on cw fields.

The conceptual foundations of the present work can be understood by considering a simple 3-level Λ system as shown in Fig. 1(a). A resonant driving field on the $a \to c$ transition with complex amplitude $E = E_0 e^{-i\omega_d t}$ and Rabi frequency $\Omega = \wp_{ac} E_0 / 2\hbar$ produces symmetric Autler-Townes dressed states with a frequency separation

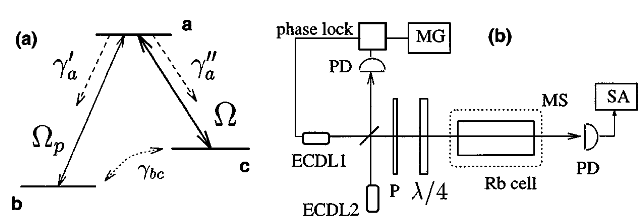

FIG. 1. (a) Simplified 3-level model for EIT. (b) Experimental setup (schematic). ECDL1 and ECDL2 are extended-cavity diode lasers (probe and driving lasers, respectively). PD: photodetector, P: polarizer, MG: microwave synthesizer, and SA: spectrum analyzer. The Rb cell is inside magnetic shields.

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of 2Ω. $\wp_{ac}$ is the dipole moment of the $a \rightarrow c$ transition and ω_d the drive-field frequency. Interference of transitions from the ground state to the two dressed states causes a reduction of absorption of a probe field E_p tuned to the center of the $b \rightarrow a$ transition. This is the essence of dark resonances [4] and EIT [5]. The EIT window in the single-atom response is power broadened and its spectral width approaches 2Ω. As we will now demonstrate, this conclusion is not valid in the case of transmission through a thick atomic ensemble [10].

The spectroscopic properties of a dense vapor of coherent Λ-type Rb atoms were studied in the experiment indicated in Fig. 1(b). Two extended-cavity diode lasers, a drive laser and a probe laser, were phase locked with a frequency offset ($\omega_0 \approx \omega_{\mathrm{HFS}} = 6.83$ GHz) determined by a tunable microwave-frequency synthesizer. Two collinear laser beams were passed through a 5 cm long cell containing natural Rb, and the transmitted power was detected by a fast photodetector (PD). Both beams were of identical circular polarization. The powers of the drive and probe beams in the cell ranged from 5 to 10 mW and 0.05 to 0.1 mW with spot sizes of 5 and 3 mm, respectively. The frequency of the drive laser was tuned to the center of the $S_{1/2}, F = 2 \rightarrow P_{1/2}, F = 2$ transition of the Rb D_1 line, while the frequency of the probe laser was scanned across the $S_{1/2}, F = 1 \rightarrow P_{1/2}, F = 2$ transition by tuning the synthesizer frequency. In such a configuration probe and drive fields create a Λ-type system [Fig. 1(a)] within the Rb D_1 manifold.

In the present experiment the signal was recorded by monitoring the amplitude of the time-dependent component of the detector current at frequency $\omega_0 = \omega_p - \omega_d$ corresponding to the difference frequency between probe and drive fields. In the case of a dilute medium the heterodyne signal at the photo detector is proportional to the transmitted probe-field amplitude.

At relatively low values of the atomic density the observed signal shows the usual absorptive features corresponding to EIT [Fig. 2(a)]. When the beat-note frequency of the lasers matches the resonant frequency of the $b \rightarrow c$ transition, the medium becomes transparent displaying an EIT-type resonance [curve *i* in Fig. 2(a)] with a nearly symmetric line shape. The peculiar feature of this resonance is that its width *decreases* with the atomic density. At higher densities the transparency window becomes substantially asymmetric. Within this window, additional few kilohertz-wide resonances [Fig. 2(b)] were clearly resolved. Their characteristics such as position, width, and amplitude depend on the strength of the drive field and on the atomic density. Furthermore their position was found to be very sensitive to atomic level shifts, induced, for example, by weak magnetic fields. Under the conditions of dense media, resonances as narrow as 3 kHz were observed with a high signal-to-noise ratio. For the data presented here, the estimated Rabi frequencies of drive and probe fields were correspondingly on the order of 10 and 2 MHz.

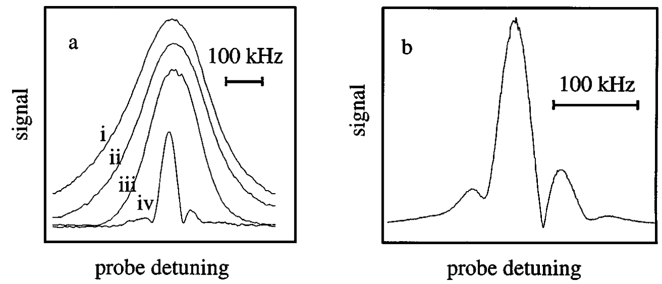

FIG. 2. Measured signal as a function of a relative laser detuning for various atomic densities. (a) Curves *i–iv* correspond to atomic densities $N \sim 6 \times 10^{10}$, 1×10^{11}, 2×10^{11}, and 6×10^{11} cm^{-3}, respectively. Estimated Rabi frequencies are 10 and 2 MHz for drive and probe. (b) Extra resonances in optically dense Rb vapor. Drive-field Rabi frequency is 13 MHz and atomic density is $\sim 10^{12}$ cm^{-3}. Scan time was a few seconds. The estimated time-of-flight broadening in the probe and drive beams is $\sim$15 and $\sim$8 kHz, respectively. There is no external magnetic field. Natural linewidth of the Rb D_1 line is 5.4 MHz. Doppler broadening is $\sim$500 MHz.

The narrowing of the dark resonances with increasing density has a nature similar to that of Ref. [8] and can easily be understood from an *idealized* 3-level model in which probe and drive interact solely with the respective transitions $a \rightarrow b$ and $a \rightarrow c$. For simplicity let us assume, in addition, a spatially homogeneous Rabi frequency of the drive field. The intensity transmission of a monochromatic probe of wavelength λ through a vapor cell of length L is described by $T = \exp\{-\chi'' kL\}$. Here $k = 2\pi/\lambda$ and χ'' is the imaginary part of the susceptibility χ, which for the idealized, homogeneously broadened 3-level Λ system and a weak ($\Omega_p < \Omega$) field is given by [5]

$$\chi = \eta\, \gamma_a' \frac{i\gamma_{bc} - (\Delta - \Delta_d)}{[\gamma_{ab} + i\Delta][\gamma_{bc} + i(\Delta - \Delta_d)] + \Omega^2}, \quad (1)$$

where $\eta = 3N\lambda^3/4\pi^2$, N is the atomic density, Δ is the detuning of the probe laser, and Δ_d is the detuning of the drive laser. γ_a', γ_a'' are the radiative decay rates as per Fig. 1(a), and $\gamma_{\mu\nu}$ are the decay rates of the respective coherences.

In the low density limit ($\eta kL \ll 1$) the spectral features of the transmission coincide exactly with those of χ''. In the high density limit, however, only spectral components which are very close to the center of the transparency are transmitted. In order to estimate the effective width of the EIT window, we expand χ'' around the point of maximum transmission [$\Delta = (1 - \gamma_{bc}/\gamma_{ab})\Delta_d$] assuming the usual EIT conditions: $\Omega^2 \gg \gamma_{ab}\gamma_{bc}, \Delta_d\gamma_{bc}, \Delta_d^2\gamma_{bc}/\gamma_{ab}$, and $\gamma_{ab} \gg \gamma_{bc}$. The resulting transmission spectrum is a Gaussian function of the detuning. Its characteristic half width

$$\Delta\omega_{\mathrm{trans}} = \frac{\Omega^2}{\sqrt{\gamma_{ab}\gamma_a'}} \frac{1}{\sqrt{\eta kL}} \quad (2)$$

scales inversely with the square root of the density-length product and can become substantially smaller than the single-atom power-broadened width.

Let us now turn to the additional resonances. We show that these resonances are due to a new field generated by resonantly enhanced coherent Raman scattering [see Fig. 3(a) and Ref. [11]] and that they reflect the *dispersive* properties of the medium. The mechanism of this process can be understood if we note that the strong driving field couples the excited state to both ground state hyperfine levels. The interaction of the drive and probe fields with the resonant transitions generates coherence between the ground states. In the presence of this coherence, the interaction of the drive with the off-resonant transition $S_{1/2}, F = 1 \rightarrow P_{1/2}, F = 2$ leads to a nonlinear generation of a new field E_n with frequency $\omega_n = 2\omega_d - \omega_p$. This process is very similar to the recently observed frequency conversion in coherent atomic media in Ref. [12]. For a large density-length product, the output amplitude of the new field E_n can be of the same order as that of the probe field, E_p. At the photodetector, the new field gives rise to an additional component to the beat note at frequency ω_0. Taking this contribution into account we find that the signal power at ω_0 in the present heterodyne detection scheme is given by

$$S_{\rm sig}^2 \propto |E_d E_n^* + E_d^* E_p|^2. \tag{3}$$

To understand the nature of the additional resonances, in particular, their relatively narrow width, let us consider again the simplified 3-level model discussed before. We now, however, take the nonresonant couplings and the associated parametric process into account. Let us further assume that the probe and "new" field are much weaker than the drive field, and that the drive field ($\Delta_d = 0$) is undepleted. These assumptions (which will not be used in the later numerical simulations) allow for a simplified discussion of the relevant physics. In this case we arrive at the system of equations

$$\frac{2}{k}\frac{\partial}{\partial z}\begin{bmatrix} E_p \\ E_n^* \end{bmatrix} = \begin{bmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{bmatrix}\begin{bmatrix} E_p \\ E_n^* \end{bmatrix}. \tag{4}$$

The coefficient a_{11} is proportional to the linear susceptibility of the resonant field E_p and a_{22} to that of the off-resonant field E_n. The cross-coupling coefficients a_{12} and a_{21} describe the resonantly enhanced $\chi^{(3)}$-type nonlinearity. Under near-resonance conditions and in the absence of Doppler broadening they are given by $a_{11} = i\chi \approx -i\eta\gamma_a'(\Delta - \Omega^2/\omega_0)/\Omega^2 - \eta\gamma_a'\gamma_{bc}/\Omega^2$, $a_{22} \approx -i\eta\gamma_a''/2\omega_0$, and $a_{12} = a_{21}^* \approx -i\eta\gamma_a'/\omega_0 \times E_d^2/|E_d|^2$. Solving this linear propagation problem and substituting the solution into Eq. (3) we find that the beat note at the photodetector contains an interference term with an amplitude depending on the probe laser detuning via the phase shift $\Delta\phi = \chi' kL/2$, which modulates the usual transmission profile and gives rise to extra resonances. Hence the characteristics of the new resonances are determined by the dispersive properties of the EIT medium [7].

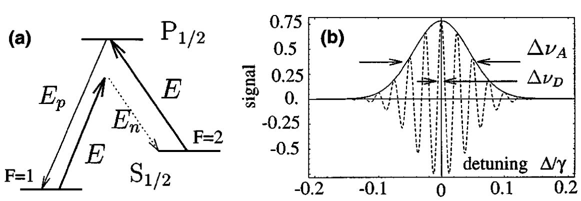

FIG. 3. (a) 3-wave mixing leading to parametric generation of field E_n within the Rb D_1 line. (b) Illustration of the difference between absorption and dispersion spectroscopy based on EIT in optically dense medium. Solid curve (characteristic width $\Delta\nu_A$) illustrates typical absorption signal, while dashed curve (width $\Delta\nu_D$) shows the dispersion signal.

Since the index of refraction close to the point of maximum transmission is a linear function of the probe detuning [7], the phase shift of the transmitted field is also linear in Δ. For a large density-length product the phase shift accumulated over the cell length can easily surpass 2π already for a very small detuning from the two-photon resonance. Hence the interference term ($\sim\exp\{-\chi'' kL/2\}\sin\Delta\phi$) in the beat note at the photodetector rapidly oscillates with Δ as indicated in Fig. 3(b). We define a characteristic width $\Delta\omega_{\rm dis}$

$$\Delta\omega_{\rm dis} = \pi\,\frac{\Omega^2}{\gamma_a'}\,\frac{1}{\eta kL}\,, \tag{5}$$

as the detuning from the line center at which the phase of a probe laser shifts by $\pi/2$. This expression scales inversely with the density-length product itself instead of its square root as in Eq. (2). Thus we conclude that in dense media the width of interferometric fringes ("dispersive" width $\Delta\omega_{\rm dis}$) can be considerably smaller than the characteristic width of the EIT transmission Eq. (2). Because of the parametric generation and the homodyne detection technique both "absorptive" and "dispersive" widths are observed in our experiment.

To make a detailed comparison with the experiment we consider a theoretical model in which the 3-wave mixing process is included together with Doppler broadening and the full (hyperfine) level structure of Rb^{87}. The propagation equations for the three fields were solved numerically. The atomic polarization was calculated from the density matrix equations with a Floquet ansatz. To truncate the resulting hierarchy, the beat note frequency ω_0 was assumed to be large compared to the relevant decay rates and Rabi frequencies. As shown in Fig. 4 the result of this calculation is in good agreement with the experimental data. In particular, the additional narrow resonances indeed have a width close to the "dispersive" width in a dense medium, Eq. (5). Finally, the observation of the extra resonances coincided with the appearance of a new field component E_n as was verified by making a beat note between the output field and an independent laser.

Before concluding we remark on the possible implications of the compensation of power broadening in dense media. To this end we estimate the theoretical limit of this compensation by considering an *ideal* 3-level Λ-type medium with a large density-length product, such that the probe-field intensity is attenuated at line center by $1/e$. In this case, which corresponds to a maximum

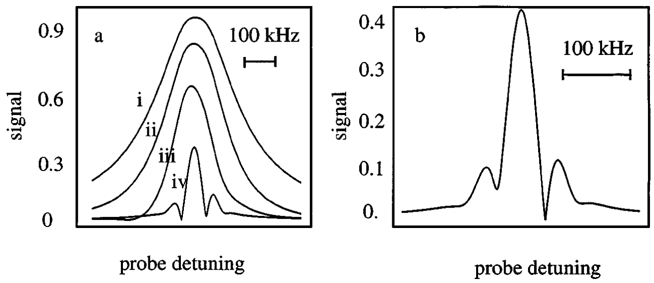

FIG. 4. Calculated signal [Eq. (3)] as a function of probe detuning. (a) Curves *i–iv* correspond to atomic densities of the Fig. 3(a). (b) Extra resonances under the conditions corresponding to those of Fig. 3(b). All parameters correspond to Fig. 3. Relaxation rate of ground state coherences is taken to be 15 kHz, and relaxation rate of ground state populations is 8 kHz. Signal is normalized such that unity corresponds to undepleted probe and drive fields.

signal-to-noise ratio at the output, the "absorptive" and "dispersive" widths approach

$$\Delta\omega_{\rm trans} \to \Omega\sqrt{\frac{\gamma_{bc}}{\gamma_{ab}}}, \qquad \Delta\omega_{\rm dis} \to \pi\gamma_{bc}, \qquad (6)$$

which implies that under *idealized* conditions the effect of power broadening can be completely compensated in a dispersive measurement [13]. For a realistic experimental situation this conclusion is certainly too optimistic due to light shifts from off-resonant levels and/or due to effects of collisional broadening of the ground state transition (e.g., for the maximal laser power used in our experiments residual power broadening of the resonances became observable at about ~20 kHz). In general these effects limit the "useful" laser intensity [14]. We further note that the particular heterodyne detection scheme used here yields a rather complicated, asymmetric line shape and is therefore not ideal for the determination of the atomic line center. It is, however, likely that modification of the detection scheme [15] can lead to more symmetric line shape and therefore to improved precision for the line center determination.

In conclusion, we have shown that a spectroscopic technique based on dense ensembles of coherent atoms can be much less sensitive to power broadening, and thus may combine high resolution with large intensities resulting in an enhanced signal-to-noise ratio.

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[1] W. Demtröder, *Laser Spectroscopy: Basic Concepts and Instrumentation* (Springer, Berlin, 1996).

[2] J. Vanier and C. Audoin, *The Quantum Physics of Atomic Frequency Standards* (A. Hilger, Bristol, 1989).

[3] The narrow features of two-photon transitions were exploited in many experiments for optically thin samples and weak fields: M. S. Feld and A. Javan, Phys. Rev. **177**, 177 (1964); W. Gawlik *et al.*, Phys. Lett. **48A**, 283 (1974); R. P. Hackel and S. Ezekiel, Phys. Rev. Lett. **42**, 1736 (1979); J. E. Thomas *et al.*, Phys. Rev. Lett. **48**, 867 (1982); M. Kailova *et al.*, Phys. Rev. A **32**, 207 (1985); A. Akulshin, A. Celikov, and V. Velichansky, Opt. Commun. **84**, 139 (1991).

[4] G. Alzetta, A. Gozzini, L. Moi, and G. Orriols, Nuovo Cimento **36B**, 5 (1976); E. Arimondo and G. Orriols, Nuovo Cimento Lett. **17**, 333 (1976); H. R. Gray, R. M. Whitley, and C. R. Stroud, Jr., Opt. Lett. **3**, 218 (1978).

[5] K.-J. Boller, A. Imamoğlu, and S. E. Harris, Phys. Rev. Lett. **66**, 2593 (1991); for review see S. E. Harris, Phys. Today **50**, No. 7, 36 (1997).

[6] Y. R. Shen, *The Principles of Nonlinear Optics* (John Wiley & Sons, New York, 1984).

[7] Dispersive properties of coherent media have been studied in M. O. Scully, Phys. Rev. Lett. **67**, 1855 (1991); S. E. Harris, J. E. Field, and A. Kasapi, Phys. Rev. A **46**, R29 (1992); M. Xiao *et al.*, Phys. Rev. Lett. **74**, 666 (1995); R. Moseley *et al.*, Phys. Rev. Lett. **74**, 670 (1995); O. Schmidt *et al.*, Phys. Rev. A **53**, R27 (1996); A. S. Zibrov *et al.*, Phys. Rev. Lett. **76**, 3935 (1996).

[8] S. Svanberg *et al.*, Opt. Lett. **11**, 138 (1986); S. Svanberg *et al.*, J. Opt. Soc. Am. B **4**, 462 (1987); O. Di Lorenzo-Filho *et al.*, Opt. Lett. **16**, 1768 (1991).

[9] A. Kasapi *et al.*, Phys. Rev. A **53**, 4547 (1996).

[10] Various aspects of propagation in dense EIT media were studied in S. E. Harris, Phys. Rev. Lett. **72**, 52 (1994); M. Fleischhauer, Phys. Rev. Lett. **72**, 989 (1994); M. Jain *et al.*, Phys. Rev. Lett. **75**, 4385 (1995); A. Kasapi *et al.*, Phys. Rev. Lett. **74**, 2447 (1995); S. E. Harris and Zhen-Fei Luo, Phys. Rev. A **52**, R928 (1995); M. B. Gorny, B. G. Matisov, and Yu. V. Rozhdestvenskii, Sov. Phys. JETP **68**, 728 (1989).

[11] Y. Li and M. Xiao, Opt. Lett. **21**, 1064 (1996).

[12] M. Jain *et al.*, Phys. Rev. Lett. **77**, 4326 (1996); we also point out the relevant studies of P. Hemmer *et al.*, Opt. Lett. **20**, 982 (1995).

[13] M. O. Scully and M. Fleischhauer, Phys. Rev. Lett. **69**, 1360 (1992).

[14] We note here that our results indicate that the ac-stark shift and accompanying power broadening due the off-resonant ($\Delta_{\rm HFS} \sim 800$ MHz) states of the $F = 1, P_{1/2}$ hyperfine manifold is substantially reduced for the present choice of polarizations, since the dark states associated with two different upper levels $P_{1/2}, F = 1$ and $F = 2$ are identical. For example, for parameters of Fig. 2 a naive estimate yields for shifts and broadening values $\Omega^2/\Delta_{\rm HFS} \sim 200$ kHz, which is more than an order of magnitude larger than the experimentally observed value.

[15] For example, detecting the generated field itself can combine a symmetric line shape with a resolution given by the dispersive width.