PHYSICAL REVIEW A VOLUME 57, NUMBER 1 JANUARY 1998

Coherent spectroscopic effects in the propagation of ultrashort pulses through a two-level system

Jinendra K. Ranka, Robert W. Schirmer,* and Alexander L. Gaeta
School of Applied and Engineering Physics, Cornell University, Ithaca, New York 14853
(Received 25 February 1997)

A theoretical and experimental investigation is made of the spectrum of a near-resonant femtosecond laser pulse propagating through a two-level atomic system. Measurements made using a high-resolution Fabry-Perot microcavity show that at the transition frequency the spectrum of the transmitted pulse acquires a feature whose structure depends sensitively on the pulse area, the pulse detuning, and the absorption path length. Our observations are in excellent agreement with the predictions of the Maxwell-Bloch equations. In the thin-sample limit, these results show that from the shape of the spectral feature it is possible to infer the quantum state of the atomic system excited by the pulse. [S1050-2947(98)50801-3]

PACS number(s): 42.50.Md, 42.62.Fi, 42.65.Re, 32.70.Jz

The interaction of coherent optical pulses with a collection of two-level atoms is one of the most fundamental problems in quantum optics [1]. This system forms the basis for understanding the interaction of pulses with multilevel systems. For the case of a single pulse tuned to a two-level resonance, the McCall-Hahn area theorem [2] shows that the temporal evolution of the pulse depends on the pulse area $\theta=(2\mu/\hbar)\int A(z,t)dt$ where μ is the dipole transition matrix element and $A(z,t)$ is the slowly varying amplitude of the electric field. The stimulated absorption and emission induced by the pulse as it propagates through the system result in a temporal reshaping in which the pulse area becomes an integral multiple of 2π. The effects predicted by this theorem include self-induced transparency and pulse compression, both of which were clearly demonstrated in a series of landmark experiments [3]. In the small-area regime, the formation of 0-π pulses has been studied theoretically [4] and experimentally [5,6]. Additional studies [7–11] have investigated various aspects of the temporal dynamics of the optical pulse and the atomic system. In most theoretical analyses, the effects of relaxation of the atomic system are neglected since the durations of the pulses are much smaller than the decay times T_1 and T_2 of the inversion and the dipole moment, respectively.

Despite the extensive work in this area, only a few theoretical studies have investigated the changes in the spectrum of a pulse as it propagates through a resonant atomic medium. Although it is well understood how the spectrum of an incoherent source is modified as the radiation passes through an atomic or molecular system, the spectral behavior of a broadband coherent source is not well understood, even in the thin-sample limit. Diels and Hahn [12] showed that a spectral feature does appear at the transition frequency and that for larger pulse areas significant deviation from a simple Lorentzian dip can result. Miklaszewski [13] performed numerical simulations of a homgeneously broadened system and demonstrated that for relatively large values of the absorption path length the resulting spectral feature can become quite complex and exhibit an oscillatory structure.

In this Rapid Communication, we describe a theoretical and experimental study of the spectral modification of a near-resonant femtosecond pulse propagating through a two-level system. Through the use of a high-finesse scanning Fabry-Perot microcavity, we perform measurements of the pulse spectrum with sub-GHz resolution over an 8-THz bandwidth. In the optically thin regime, our results show that the shape of the resulting spectral feature at the transition frequency depends on the pulse area and the detuning. This behavior can be understood as the interference of the incident pulse with the radiation emitted by the excited atoms undergoing free-induction decay. From the shape of the feature in the transmitted pulse spectrum, one can infer the quantum state of the two-level system excited by the pulse, obviating the need of probing the system. At higher atomic densities, propagation effects play an important role and the resulting spectral feature develops a complicated oscillatory structure. All our experimental results are well described by analytical and numerical solutions of the Maxwell-Bloch equations.

The propagation of an optical pulse in a two-level medium with an atomic density N can be modeled using the Maxwell-Bloch equations

$$\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)\Omega=i\frac{\alpha_0}{T_2}\sigma, \tag{1a}$$

$$\frac{\partial\sigma}{\partial t}=-\left(\frac{1}{T_2}-i\Delta\right)\sigma-\frac{i\Omega}{2}w, \tag{1b}$$

$$\frac{\partial w}{\partial t}=-\left(\frac{1+w}{T_1}\right)+i(\Omega\sigma^*-\Omega^*\sigma), \tag{1c}$$

where w is the inversion, σ is the slowly varying, off-diagonal density-matrix component, $\Omega(z,t)=2\mu A(z,t)/\hbar$ is the Rabi frequency for the pulse envelope, Δ is the detuning of the pulse from the atomic transition frequency ω_0, and $\alpha_0=4\pi\omega_0 N|\mu|^2T_2/\hbar c$ is the line-center absorption coefficient. Doppler broadening can be included by averaging the atomic variables over a Doppler profile of width $2/T_2^*$, where T_2^* is the inhomogeneous dephasing time. In all of our analysis, we consider the regime similar to that of our ex-

*Permanent address: Department of Physics, Clark Hall, Cornell University, Ithaca, NY 14853.

periment in which the pulse duration τ_p is much shorter than any of the characteristic time scales (T_1, T_2, T_2^*) of the atomic medium and in which the pulse does not undergo significant pulse reshaping (i.e., $\alpha_0 L \tau_p / T_2 \ll 1$). As a result, the qualitative behavior of the system does not depend on the relative values of these decay constants.

In the optically thin limit ($\alpha_0 L \ll 1$), we can predict analytically the shape of the feature that should appear in the transmitted pulse spectrum. The in-quadrature part $v(t) = 2\,\mathrm{Im}(\sigma)$ of the dipole moment is proportional to the in-phase part of the dipole field and is responsible for the exchange of energy between the field and the atoms. We assume that a pulse of area θ excites the two-level atom to a point on the Bloch sphere at time $t=0$, and the atom then undergoes free-induction decay. For the case of a square pulse of width τ_p, the Fourier transform of $v(t)$ is then given by

$$\tilde{v}(\delta\omega) = -\frac{1}{2[1+(\delta\omega T_2)^2]}\left[\frac{\theta}{\theta'}\sin\theta' - (\delta\omega T_2) \times \frac{\theta \Delta\tau_p}{\theta'^2}(1-\cos\theta')\right], \quad (2)$$

where $\delta\omega$ is the frequency offset relative to the atomic frequency and $\theta' = [\theta^2 + (\Delta\tau_p)^2]^{1/2}$. In the thin-sample limit, the transmitted pulse spectrum is the input pulse spectrum plus a term resulting from the interference of the input pulse and the dipole field, which is proportional to the expression above for $\tilde{v}(\delta\omega)$. We concentrate our attention on those cases in which $|\Delta\tau_p| < 1$ such that the pulse spectrum overlaps appreciably with the atomic transition frequency. For $\Delta = 0$, the second term on the right-hand side does not contribute, and the spectral feature will be a Lorentzian dip ($\theta < \pi$) or peak ($\pi < \theta < 2\pi$) of width $2/T_2$ at the atomic frequency. When $\Delta \neq 0$, the behavior is more complicated. For pulse areas such that $\theta < \pi/2$, the first term on the right-hand side is dominant, which results again in a dip at ω_0. For a pulse area $\theta \sim \pi$, the second term on the right-hand side is dominant, and a dispersive feature is predicted for the spectrum. Note that this feature flips its orientation depending on the sign of Δ. For $3\pi/2 < \theta < 2\pi$, the first term dominates once again but with a change in sign resulting in a spectral peak at the transition frequency. For the case in which the atoms are Doppler broadened, the predicted spectral shapes are similar but with widths given by $\sim 2/T_2^*$.

The analysis above gives a qualitative description of the predicted spectral shapes for values of $\alpha_0 L < 10$, as confirmed by numerical integration of Eqs. (1). In our numerical simulations, the shape of the input pulse is given by $\Omega(0,t) = (\theta/\pi\tau_p)\,\mathrm{sech}(t/\tau_p)$. In Figs. 1(a)–(d) we plot the transmitted pulse spectrum near the transition frequency for several different pulse areas. The parameters for all four graphs are $\alpha_0 L = 2$, $T_2/T_1 = 2$, $\tau_p/T_2 = 10^{-3}$, and $\Delta\tau_p = -0.4$. As demonstrated by the inset to Fig. 1(a), which shows the total pulse spectrum for $\theta = \pi/2$, only the frequency components near the transition frequency are affected by the interaction. For this case, the spectral feature has the form of a dip of width $\sim 2/T_2$. For the larger pulse areas, the feature evolves into a dispersive-looking feature

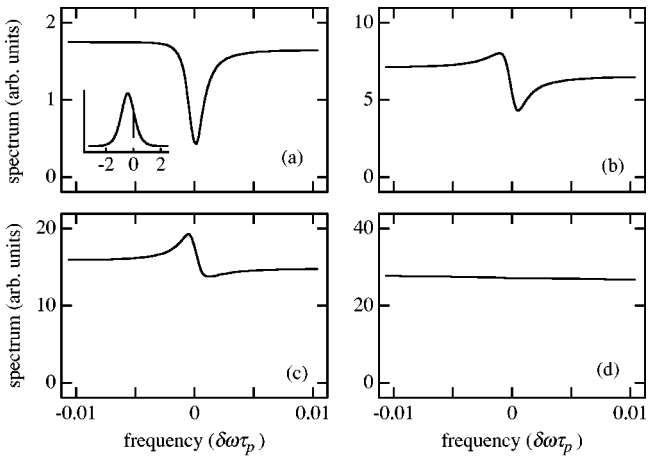

FIG. 1. Theoretical predictions of the transmitted pulse spectrum near the atomic resonance frequency (i.e., $\delta\omega = 0$) for various pulse areas. In all cases, $\alpha_0 L = 2$, $T_2/T_1 = 2$, $\tau_p/T_2 = 10^{-3}$, and $\Delta\tau_p = -0.4$. The pulse areas are (a) $\theta = \pi/2$, (b) $\theta = \pi$, (c) $\theta = 3\pi/2$, (d) $\theta = 2\pi$. The total pulse spectrum is shown in the inset to (a) for $\theta = \pi/2$.

for $\theta = \pi$ [Fig. 1(b)] and into a peak for $\theta = 3\pi/2$ [Fig. 1(c)]. This behavior is in direct contrast to the common belief that a dip should always appear in the spectrum of a broadband radiation propagating through an absorbing medium. For $\theta = 2\pi$ [Fig. 1(d)] the spectrum shows little change from the input, as expected for a near-soliton pulse. If the pulse is tuned above resonance, the feature flips its orientation, and when tuned exactly to resonance, only a dip or a peak is observed, again as predicted by the analytical expression [Eq. (2)]. In the time domain, the transmitted pulse shape is nearly identical to the input pulse, except for the presence of a very weak and slowly decaying tail that represents the dipole field. These predictions show that for a two-level system, the quantum state (i.e., the position of the Bloch vector) excited by the input pulse can be inferred by the shape of the spectral feature that appears at the atomic frequency.

We experimentally verified these predictions using potassium vapor as the two-level medium. Pulses of duration (full width at half maximum) $T_p = 415$ fs and with an energy of 15 nJ were generated at a 90-MHz repetition rate from a mode-locked Ti:sapphire laser. The central frequency of the pulses was tuned near the $4\,^2S_{1/2}$–$4\,^2P_{1/2}$ (770.1-nm) transition. A Pockels cell external to the laser was used to slice pulses out of the pulse train at a repetition rate f sufficiently small that multiple-pulse effects in the atomic system were minimized. No change in the transmitted spectra was observed as the repetition rate was varied over a range of values $f < 10$ MHz. For all the results shown here, the repetition rate was 9 MHz. The pulses were focused into a potassium vapor cell [14] using a 10-cm-focal-length lens. The position of the lens was adjusted to place the focus near the input face of the cell. The interaction length of the vapor cell is 2 mm, and the vapor density could be varied between 10^8 and 10^{14} cm^{-3} by changing the temperature of the potassium reservoir.

The spectrum of the beam transmitted through the cell was analyzed using a high-finesse, scanning Fabry-Perot cavity (FPC) (Newport, model SR-240). The free-spectral range of the cavity is 8 THz, and the high-reflectivity mirrors result in a spectral resolution of 500 MHz at 770 nm. After

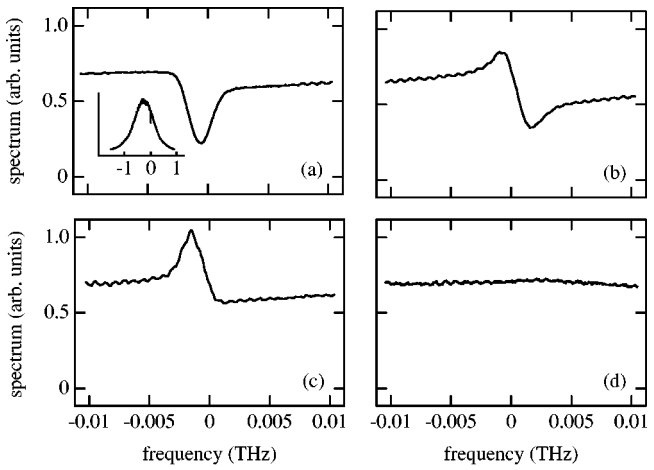

FIG. 2. Experimentally measured spectra of the transmitted pulse near the atomic resonance frequency (i.e., $\delta\omega=0$) for various pulse areas. In all cases, the density of the atomic vapor is estimated to be $N=2\times10^{11}$ cm^{-3}, $\tau_p/T_2=10^{-3}$, and $\Delta\tau_p=-0.4$. The pulse areas are (a) $\theta=0.3\pi$, (b) $\theta=\pi$, (c) $\theta=3\pi/2$, (d) $\theta=2\pi$. The total pulse spectrum is shown in the inset to (a) for $\theta=0.3\pi$.

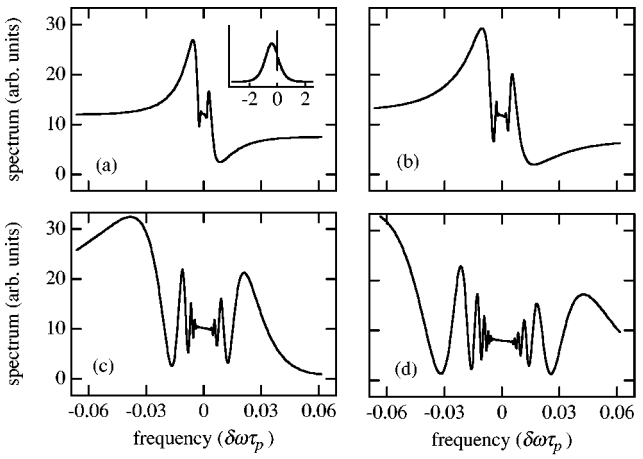

FIG. 3. Theoretical predictions of the transmitted pulse spectrum near the atomic resonance frequency for increasing absorption path length. In all cases, $\theta=0.9\pi$ and $\Delta\tau_p=-0.4$. The absorption path lengths are (a) $\alpha_0L=50$, (b) $\alpha_0L=100$, (c) $\alpha_0L=400$, (d) $\alpha_0L=800$. The total pulse spectrum is shown in the inset to (a) for $\alpha_0L=50$.

passing through the cell, the beam was coupled into a 15-cm-long single-mode fiber, and the output was mode-matched into the FPC with a pair of lenses. The use of the fiber allowed us to make adjustments at the input end without effecting the alignment of the beam into the FPC. The amount of light coupled into the fiber was kept sufficiently small such that self-phase modulation inside the fiber did not lead to any detectable distortions in the measured pulse spectrum. The light transmitted through the FPC was detected using a photomultiplier tube, and the signal was stored on a digital oscilloscope. The scan rate of the cavity was 30 Hz, and several frequency scans of the supercavity were averaged to produce the spectra shown here. The alignment of the FPC was initially performed using a frequency-stabilized Ti:sapphire laser (Coherent 899-21).

In Fig. 2, we plot the measured spectra of the transmitted pulses in the optically thin regime for several values of the input pulse area. The temperature of the potassium vapor was 90 °C, which we estimate corresponds to $N=2\times10^{11}$ cm^{-3}. The center frequency of the input pulse was tuned below resonance ($\Delta T_p=-0.5$). The inset to Fig. 2(a) shows the total transmitted pulse spectrum for $\theta=0.3\pi$. As expected, the spectrum is modified only near the atomic transition frequency. As predicted by our theoretical analysis, the spectral feature evolves from a dip for relatively small pulse areas [$\theta=0.3\pi$, Fig. 2(a)], to a dispersive feature for a π pulse [Fig. 2(b)], to a peak for a near $3\pi/2$ pulse [Fig. 2(c)]. For a near 2π pulse [Fig. 2(d)], the spectrum was essentially unchanged from the input. When the pulse area was increased slightly above 2π, the feature again became an absorptive dip. When the center wavelength of a near π pulse was tuned across the atomic resonance, the spectral feature was observed to flip its orientation, as predicted by the theoretical analysis. In all cases, the width of the spectral feature was comparable to the Doppler width of 1 GHz. We were unable to confirm the shape of the predicted spectral feature for relatively large pulse areas at $\Delta=0$ as a result of jitter (several GHz) in the central frequency of the pulse during the scanning time of the FPC. The sensitivity of our measurement scheme allowed for detection of the spectral feature in the transmitted pulse at atomic densities as low as $N\sim10^8$ cm^{-3}. Additional stabilization of our laser system should allow for even greater sensitivities.

As the the absorption length of the medium increases, propagation effects begin to play an important role, and the shape of the resulting spectral feature becomes considerably more complicated. For an incoherent broad-bandwidth source, the dip in the transmitted spectrum becomes deeper and wider. However, for coherent pulses with relatively large pulse areas, the behavior is very different. In Fig. 3, the theoretically predicted spectrum is plotted for increasing atomic density, for the case in which $\theta=0.9\pi$, $\Delta\tau_p=-0.4$,

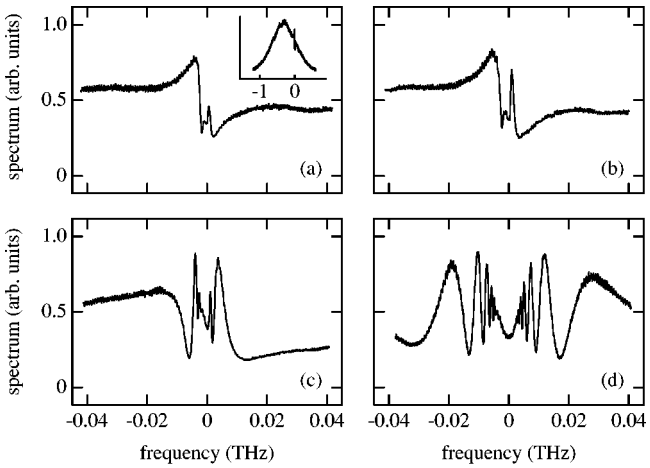

FIG. 4. Experimentally measured spectra of the transmitted pulse near the atomic resonance frequency (i.e., $\delta\omega=0$) for increasing absorption path length. In all cases, $\theta=0.9\pi$ and $\Delta T_p=-0.5$. The atomic densities are (a) $N=2\times10^{12}$ cm^{-3}, (b) $N=9\times10^{12}$ cm^{-3}, (c) $N=3\times10^{13}$ cm^{-3}, (d) $N=2\times10^{14}$ cm^{-3}. The total pulse spectrum is shown in the inset to (a) for $N=2\times10^{12}$ cm^{-3}.

$T_2/T_1=2$, and $\tau_p/T_2=10^{-3}$. As the absorption is increased, further structure develops at the atomic frequency and the feature broadens. For values of $\alpha_0 L\tau_p/T_2$ that approach unity, the spectral feature develops an oscillatory structure in which the number of oscillations increases with absorption path length. For pulse areas $\theta<2\pi$, this oscillatory structure is most pronounced for the range $\pi/2<\theta<3\pi/2$. The origin of the oscillations can be traced to the temporally oscillating tail [13] that appears in the transmitted pulse. This tail arises as a result of the pulse shaping that occurs as the pulse propagates through the system and its area stabilizes to the value given by the generalized area theorem [1].

The experimentally observed behavior at these larger absorption lengths mirrors that predicted by our theoretical simulations. Figure 4 shows the spectrum of the pulse near the atomic transition frequency as the density of the atomic vapor is increased for the case in which the pulse area was $\sim 0.9\pi$ and the pulse was detuned below resonance ($\Delta T_p=-0.5$). As predicted by theory, the spectral feature develops oscillations that become more numerous as the the density is increased [(a) $N=2\times10^{12}$ cm^{-3}, (b) $N=9\times10^{12}$ cm^{-3}, (c) $N=3\times10^{13}$ cm^{-3}, (d) $N=2\times10^{14}$ cm^{-3}]. For similar values ($<2\times$) of $\alpha_0 L\tau_p/T_2$ we have good agreement between the theoretically predicted line shapes shown in Fig. 3 and the experimentally observed spectra. We believe that the transverse Gaussian spatial profile of the beam and the relatively large uncertainty in our estimate of the atomic density prevent us from achieving absolute agreement with the plane-wave model.

In summary, we have studied the modification of the spectrum of a femtosecond pulse propagating through a two-level system. We observe a number of coherent effects for pulse areas on the order of π. In thin-sample limit, we find that the shape of the feature that appears on the transmitted pulse spectrum can be used to ascertain the state of the atomic system that had been excited by the incident pulse. These results are the first step in understanding the spectral behavior of pulses in multilevel systems, and are suggestive that the quantum states of these systems could be determined by analyzing the spectrum of the transmitted excitation pulse.

We gratefully acknowledge discussions with J. Eberly and I. Walmsley. This work was supported by the U.S. Army Research Office, Grant No. DAAH04-95-1-0420, and the National Science Foundation, Grant No. PHY-9307511.

[1] See, for example, L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Dover, New York, 1987).
[2] S. L. McCall and E. L. Hahn, Phys. Rev. Lett. **18**, 908 (1967); Phys. Rev. **183**, 457 (1967).
[3] H. M. Gibbs and R. E. Slusher, Phys. Rev. Lett. **24**, 638 (1970); Appl. Phys. Lett. **18**, 505 (1971); Phys. Rev. A **5**, 1634 (1972).
[4] M. D. Crisp, Phys. Rev. A **1**, 1604 (1970).
[5] J. E. Rothernberg, D. Grischkowsky, and A. C. Balant, Phys. Rev. Lett. **53**, 552 (1984).
[6] M. Matusovsky, B. Vaynberg, and M. Rosenbluh, J. Opt. Soc. Am. B **13**, 1994 (1996).
[7] D. Grischkowsky, Phys. Rev. Lett. **24**, 866 (1970); Phys. Rev. A **7**, 2096 (1973).
[8] G. L. Lamb, Jr., Rev. Mod. Phys. **43**, 99 (1971).
[9] D. C. Burnham and R. Y. Chiao, Phys. Rev. **188**, 667 (1969).
[10] J. N. Sweetser and I. A. Walmsley, J. Opt. Soc. Am. B **13**, 611 (1996).
[11] M. Matusovsky, B. Vaynberg, and M. Rosenbluh, Phys. Rev. Lett. **77**, 5198 (1996).
[12] J. C. Diels and E. L. Hahn, Phys. Rev. A **8**, 1084 (1973).
[13] W. Miklaszewski, J. Opt. Soc. Am. B **12**, 1909 (1995); Z. Phys. B **93**, 491 (1994).
[14] M. Y. Lanzerotti, R. W. Schirmer, and A. L. Gaeta, Appl. Phys. Lett. **69**, 1199 (1996).