Induced gain and modified absorption of a weak probe beam in a strongly driven sodium vapor

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We present the results of an experimental and theoretical study of the gain (or absorption) experienced by a weak probe beam propagating through a sodium vapor in the presence of an intense pump field that is nearly resonant with the $3s \rightarrow 3p$ atomic transition. The probe-transmission spectrum includes three distinct features that result from the modification of the atomic-level structure by the ac-Stark effect. Two of these features can lead to amplification of the probe wave. We measured the dependence of the probe spectrum on the detuning of the pump beam from resonance and on the pressure of a helium buffer gas. The experimentally obtained spectra are in good agreement with the predictions of a theoretical model based on the solution of the density-matrix equations of motion for a two-level atom and including the effects of Doppler broadening. The maximum gain measured in these experiments occurs at one of the Rabi sidebands and leads to a 38-fold increase in the intensity of the probe wave. Under some experimental conditions, we also observed two additional resonances, which are due to stimulated Raman scattering involving the sodium ground-state hyperfine levels.

1. INTRODUCTION

There is great interest in nonlinear optical processes capable of transferring energy from one coherent optical field to another. In this paper we investigate the transmission of a weak probe wave through an atomic vapor that is strongly driven by an intense, nearly resonant laser field and show that the resulting interaction can transfer energy from the pump beam to the probe beam.

Multiwave mixing in atomic systems has been used to generate a variety of resonances that may be exploited for beam coupling. These resonances include several Raman-type phenomena[1–5] that occur in three-level systems as well as induced resonances in strongly driven two-level atoms.[6–12] Gain sufficient to support oscillation has been achieved in sodium vapor.[5,7]

The interaction of nearly resonant fields in a system of stationary two-level atoms was originally formulated according to density-matrix equations of motion by Bloembergen and Shen.[8] Mollow[9] calculated the rate of absorption of a weak probe wave by a strongly driven atom and predicted a profoundly modified line shape that permits the possibility of gain. Wu *et al.*[10] observed the theoretically predicted spectral features by using a sodium atomic beam. The theoretical work, which treats homogeneously broadened media, has been extended to include phase-matched coupling in four-wave mixing.[11,12]

To obtain high atomic-number densities, experiments are often carried out in vapor cells[13–16] or heat pipes,[17] where atomic motion and, in the case of heat pipes, buffer-gas broadening are unavoidable. Analytic solutions for the weak-probe-wave absorption profile in Doppler-broadened media have been presented for some limiting cases, including the inhomogeneously broadened limit[18,19] and a Lorentzian distribution of velocities.[19,20] In addition, several authors[21–23] have shown the importance of including the effects of Doppler broadening when calculating the angular dependence of the phase-conjugate signal in degenerate and nearly degenerate four-wave-mixing schemes. As a result of atomic motion, the reflectivity depends sensitively on the pump–probe crossing angle.

In Section 2 of this paper, we review the theoretical predictions for the absorption response of a strongly driven two-level atom in the homogeneously broadened limit. In Section 3 we extend this result to include an arbitrary ratio of collisional broadening to Doppler broadening for an arbitrary crossing angle between the pump and probe waves. The effects of atomic motion are introduced by numerically integrating the absorption response over a Gaussian distribution of thermal velocities. Experimental results are presented in Section 4, where several resonances are identified and studied. In particular, we investigate the influence of the pump detuning from resonance and the collisional-dephasing rate on the absorption spectrum and find both subtle and dramatic modifications of the spectral profiles. The agreement between theory and experiment is very good.

2. GAIN AND ABSORPTION FOR HOMOGENEOUS BROADENING

The absorption spectrum of a weak wave interacting with a strongly driven two-level atom can be understood qualitatively in terms of the energy-level diagrams shown in Fig. 1. Consider first the interaction of a weak laser field at frequency ω_1 with a two-level atom having ground state $|a\rangle$ and excited state $|b\rangle$ with transition frequency ω_{ba} [Fig. 1(a)]. If the detuning of the laser from resonance is $\Delta = \omega_1 - \omega_{ba}$ ($|\Delta| \ll \omega_{ba}$), then the laser in effect creates a pair of virtual levels (shown by dashed lines), one at an energy $\hbar\omega_1$ above the ground state, the other at an energy $\hbar\omega_1$ below the excited state. In accordance with the conditions of the experiments described below, the pump detuning in Fig. 1 is negative.

If the applied laser field is sufficiently strong, the atomic energy-level structure is profoundly modified by the ac-Stark effect,[24,25] leading to dressed states,[26] which are the stationary states of the combined laser-field-and-atom

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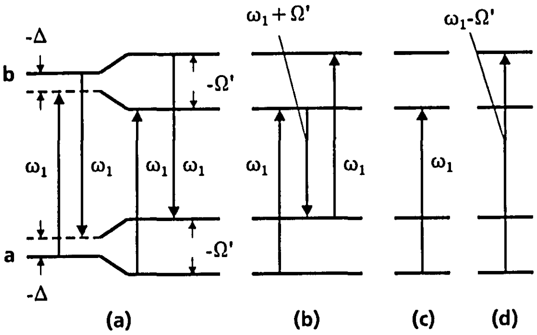

Fig. 1. (a) A laser field at frequency ω_1 that is detuned from resonance by frequency Δ creates a pair of virtual atomic levels, shown as dashed lines. At high laser intensities, the energy-level structure is modified by the ac-Stark effect to become pairs of dressed levels separated by the generalized Rabi frequency Ω', as shown at the right. The possible transitions among these levels are illustrated in (b)–(d). (Both the pump-laser detuning from resonance and the generalized Rabi frequency are negative in this figure.) (b) By simultaneously absorbing two pump-laser photons and emitting a photon with frequency $\omega_1 + \Omega'$, the atom makes a transition to the excited state. As a consequence of this three-photon effect, an incident wave at frequency $\omega_1 + \Omega'$ experiences gain. (c) When the difference between the pump- and probe-wave frequencies is comparable with the inverse of the atomic response time, the strong interaction of the waves' beat frequency with the populations of the atomic levels results in a nearly degenerate absorption feature with a dispersive line shape. (d) The ac-Stark effect shifts the atomic resonance frequency to $\omega_1 - \Omega'$.

Hamiltonian. Each pair of dressed states is split by the generalized Rabi frequency $\Omega' = (\Delta/|\Delta|)(\Delta^2 + \Omega^2)^{1/2}$, where $\Omega = |\mu_{ba}|E_1/\hbar$ is the Rabi frequency, μ_{ba} is the atomic-dipole moment matrix element, and E_1 is the real amplitude of the strong laser field.

The structure of the modified atomic states can be determined by measuring the absorption spectrum with a probe wave whose intensity is weak enough that it does not itself dress the atomic states. For the moment, we ignore the effects of atomic motion. Starting with the density-matrix equations of motion[8,9,27,28] and including the influence of the strong optical field to all orders, while retaining that of the probe field to only first order, results in the following amplitude absorption coefficient for the weak probe wave at ω_3 (Ref. 12)

$$\alpha(\delta) = \frac{2\pi N\omega_3|\mu_{ba}|^2(\rho_{bb} - \rho_{aa})^{dc}}{\hbar n_3 c} \times \mathrm{Im}\left[\frac{(\delta + i/T_1)((\delta - \Delta + i)/T_2)(\Delta - i/T_2) - (\Omega^2\delta/2)}{(\Delta - i/T_2)D(\delta)}\right], \tag{1}$$

where $\delta = \omega_3 - \omega_1$ is the probe–pump detuning, T_1 and T_2 are the population lifetime and the dipole-dephasing time, respectively, N is the number density of atoms, n_3 is the index of refraction of the atomic medium at frequency ω_3, and Im[ ] denotes the imaginary part. In Eq. (1), $(\rho_{bb} - \rho_{aa})^{dc}$ is the steady-state population inversion induced by the strong laser field:

$$(\rho_{bb} - \rho_{aa})^{dc} = \frac{(1 + \Delta^2 T_2^{\,2})(\rho_{bb} - \rho_{aa})^0}{1 + \Delta^2 T_2^{\,2} + \Omega^2 T_1 T_2}, \tag{2}$$

where $(\rho_{bb} - \rho_{aa})^0$ is the equilibrium population inversion in the absence of the optical fields, and $D(\delta)$ is a cubic polynomial in δ given by

$$D(\delta) = (\delta + i/T_1)(\delta + \Delta + i/T_2)(\delta - \Delta + i/T_2) - \Omega^2(\delta + i/T_2). \tag{3}$$

The probe-wave absorption coefficient $\alpha(\delta)$ for a typical case, shown in Fig. 2, has three features. The ac-Stark-shifted atomic resonance at $\omega_3 = \omega_1 - \Omega'$ corresponds to the transition between the lowest and highest dressed levels [Fig. 1(d)]. Because the ground state is more highly populated than the excited state, the probe wave is strongly attenuated at this frequency.

The central feature in the absorption spectrum occurs where the probe–pump detuning is less than the inverse of some characteristic atomic response time [Fig. 1(c)]. The line shape of this nearly degenerate feature appears dispersive whenever the pump is detuned from resonance. In the high-intensity, large-detuning limit ($\Omega, \Delta \gg 1/T_2$), which corresponds to our experimental conditions, Eqs. (1)–(3) simplify somewhat in the vicinity of the nearly degenerate resonance to give

$$\alpha_{nd}(\delta) \approx A\,\frac{A_1 + A_2\dfrac{\delta}{\Gamma_{nd}} + A_3\dfrac{\delta^2}{\Gamma_{nd}^{\,2}}}{1 + \delta^2/\Gamma_{nd}^{\,2}}, \tag{4}$$

where

$$A = \frac{2\pi N\omega_3|\mu_{ba}|^2}{n_3 c\hbar T_1}\,\frac{1}{\left[\dfrac{1}{T_1} + \dfrac{1}{T_2}(\Omega/\Delta)^2\right]^3}, \tag{5}$$

$$A_1 = \frac{1}{\Delta^2 T_1 T_2}\left[\frac{1}{T_1} + \frac{1}{T_2}(\Omega/\Delta)^2\right], \tag{6}$$

$$A_2 = \frac{\Omega^2\Gamma_{nd}}{2\Delta^3}\left\{\frac{1}{T_2}[2 + (\Omega/\Delta)^2] - \frac{1}{T_1}\right\}, \tag{7}$$

$$A_3 = \frac{\Gamma_{nd}^{\,2}}{\Delta^2 T_2}, \tag{8}$$

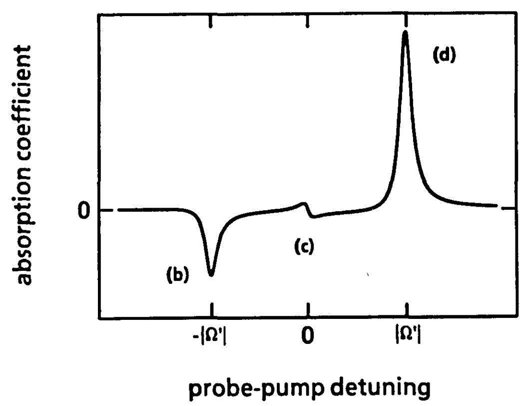

Fig. 2. The probe absorption spectrum as modified by the intense pump-laser field for $T_2/T_1 = 2$, $\Omega T_2 = 20$, and $\Delta T_2 = -5$. (b)–(d) Correspondence between the three spectral features and the transitions illustrated in Fig. 1.

and

$$\Gamma_{nd} = \frac{\frac{1}{T_1} + \frac{1}{T_2}(\Omega/\Delta)^2}{1 + (\Omega/\Delta)^2} \tag{9}$$

is the linewidth of the resonance. The linewidth Γ_{nd} lies between $1/T_1$ and $1/T_2$; the exact value depends on the ratio Ω/Δ of the Rabi frequency to the pump detuning from resonance. The nearly degenerate resonance shares a common origin with the "spectral hole due to population oscillations,"[12,27,29-33] that is, the decreased absorption experienced by a probe wave in the presence of a strong pump wave in a different limit (Δ, $\Omega \ll 1/T_2$, $T_2 \ll T_1$, and $\delta \lesssim 1/T_2$).

The three-photon effect occurs when the atom makes a transition to the excited state by simultaneously absorbing two laser photons with frequency ω_1 and emitting a photon with frequency $\omega_1 + \Omega'$ [Fig. 1(b)].[22] By the corresponding stimulated process, the probe wave experiences gain at frequency $\omega_3 \approx \omega_1 + \Omega'$, giving rise to the third spectral feature. (The atom subsequently deexcites by spontaneously emitting a photon at the ac-Stark-shifted atomic resonance frequency $\omega_1 - \Omega'$.) In the high-intensity, large-detuning limit ($\Omega, \Delta \gg 1/T_2$), the probe absorption line shape [Eqs. (1)–(3)] near the three-photon resonance reduces to a Lorentzian centered at $\delta = \Omega'$:

$$\alpha_{3p}(\delta) \approx A_{3p}\frac{1}{1 + (\delta + \Omega')^2/{\Gamma_{3p}}^2}, \tag{10}$$

where

$$A_{3p} = \frac{2\pi N\omega_3|\mu_{ba}|^2}{\hbar n_3 c}\,\frac{T_2(\Omega'/\Delta)}{T_2 + T_1(\Omega/\Delta)^2} \times \frac{(\Omega'/\Delta) - (1/2)(\Omega/\Delta)^2 - 1}{(1/T_1)(\Omega/\Delta)^2 + (1/T_2)[(\Omega/\Delta)^2 + 2]} \tag{11}$$

is the maximum three-photon gain and

$$\Gamma_{3p} = \frac{(1/T_1)(\Omega/\Delta)^2 + (1/T_2)[2 + (\Omega/\Delta)^2]}{2[1 + (\Omega/\Delta)^2]} \tag{12}$$

is the linewidth, which has a value between $1/T_2$ and $(1/2)[1/T_1) + (1/T_2)]$, depending on the ratio of Ω to Δ.

3. EFFECTS OF ATOMIC MOTION

The general expression for the absorption coefficient in the presence of atomic motion is obtained by averaging the single-atom absorption coefficient over a Maxwellian distribution of atomic velocities:

$$\alpha^D(\omega_3, \omega_1) = \left(\frac{m}{2\pi k_B T}\right)^{3/2} \iiint_{-\infty}^{+\infty} \mathrm{d}^3\mathbf{v}\, \alpha(\omega_3 + \mathbf{k}_3\cdot\mathbf{v}, \omega_1 + \mathbf{k}_1\cdot\mathbf{v}) \exp\left(-\frac{m|v|^2}{2k_B T}\right), \tag{13}$$

where m is the atomic mass, k_B is the Boltzmann constant, T is the absolute temperature, and $\mathbf{k}_1$ and $\mathbf{k}_3$ are the wave vectors of the optical waves at frequencies ω_1 and ω_3, respectively. If the two waves are copropagating, the triple integral simplifies to become

$$\alpha^D(\omega_3, \omega_1) = \frac{1}{(\pi {T_2}^*)^{1/2}} \int_{-\infty}^{+\infty} \mathrm{d}\omega_D \alpha(\omega_3 + \omega_D, \omega_1 + \omega_D) \times \exp[-(\omega_D {T_2}^*)^2], \tag{14}$$

where ${T_2}^* = (\lambda/2\pi)(m/k_BT)^{1/2}$ is the effective Doppler dephasing time.

In general an analytic expression for the Doppler-averaged absorption line shape cannot be obtained, and we have therefore performed the required integration numerically. [Examples of the resulting transmission spectra are shown below in Figure 4(b).] In each case illustrated, the three spectral features of the single-atom response are still present. Atomic motion broadens the three-photon resonance and the ac-Stark-shifted atomic resonance (to roughly $1/{T_2}^*$). In addition, the peak of the three-photon gain feature is shifted toward the pump frequency. This shift occurs because those atoms whose velocities bring them closer to the pump frequency produce larger three-photon gain than do those atoms whose velocities shift them away from the pump frequency. In contrast, the nearly degenerate feature is relatively unaffected by the effects of atomic motion. This insensitivity occurs because the nearly degenerate feature is centered at $\delta = \omega_3 - \omega_1 = 0$ for each atom. In the limiting case of a zero beam-crossing angle, the pump and probe waves are Doppler shifted by the same amount, leading to a resonance that is centered on the pump frequency for each velocity group of atoms. Furthermore, in the limiting cases when Ω/Δ is either much smaller or much greater than unity for all velocity groups of atoms, the width of the resonance is also independent of the atomic velocity, as can be seen from Eq. (9). However, for nonzero values of the beam-crossing angle, the beat frequency δ becomes velocity dependent, leading to a broadening of the nearly degenerate line shape. This effect can lead to a substantial (threefold) broadening of the line shape even for beam-crossing angles as small as 1.6°, as discussed below in connection with Fig. 6.

4. EXPERIMENTAL RESULTS

A Coherent Innova 100 argon-ion laser is used to excite two Coherent CR699-21 continuous-wave, frequency-stabilized ring dye lasers. Both lasers operate in a single longitudinal mode tuned near the sodium D_2 resonance line and have the same linear polarization. The output from one of the lasers is focused with a lens used at $f/360$ to produce a pump beam whose intensity is about 200 W/cm^2. The corresponding Rabi frequency is approximately 1.3 GHz, which is comparable with the Doppler width and with the ground-state hyperfine splitting. The pump laser is tuned to the low-frequency side of the D_2 resonance line to avoid beam breakup owing to the self-focusing effects that occur on the high-frequency side. A weak probe field is produced by attenuating the output from the other dye laser by a factor of 10^3 and then focusing with a lens used at $f/130$. This procedure yields a probe intensity of less than 1 W/cm^2 and a spot size about three times smaller than that of the pump field.

The two laser fields are brought to a common focus in a 7-mm-long sodium-vapor cell consisting of a stainless-steel body and sidearm. The sidearm temperature is maintained at about 250 °C to produce a sodium number density of $\sim 3 \times$

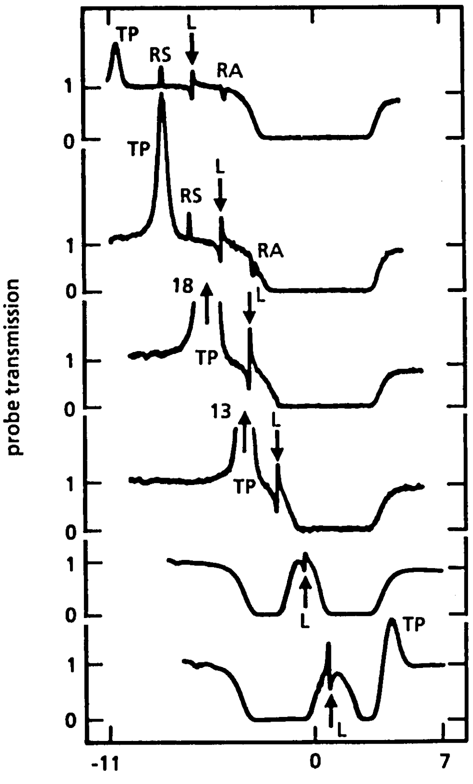

Fig. 3. Dependence of the probe transmission spectrum on pump detuning from resonance. In successive spectra, the pump-beam frequency is increased by 1.5 GHz. The resonances in the spectra are the three-photon gain (TP), the Raman Stokes gain (RS), the nearly degenerate resonance near the pump-laser frequency (L), the Raman anti-Stokes loss (RA), and the Stark-shifted atomic absorption. In the bottom spectrum, the nearly degenerate resonance has changed sign, and the three-photon gain appears on the high-frequency side of the pump frequency.

10^{13} atoms/cm^3 and a corresponding self-broadened weak-field line-center intensity absorption of $\alpha_0 L \approx 300$. Polarization distortions are minimized by using zero-degree-oriented sapphire windows at near-normal incidence.

Figure 3 shows the probe-transmission spectrum for several different choices of the pump-laser frequency. In each case, the spectral features due to the three-photon effect (labeled TP), nearly degenerate resonance near the laser frequency (L), and the Stark-shifted atomic absorption are present. In some cases, two additional features not predicted by the two-level-atom model appear; the Raman Stokes (RS) gain resonance and Raman anti-Stokes (RA) loss resonance are due to stimulated Raman scattering involving the hyperfine levels of the electronic ground state.[34] In some of the spectra, these Raman resonances are not visible because they are hidden under the three-photon gain peak or the atomic absorption. The gain at either the three-photon or the nearly degenerate resonance can be maximized by an appropriate choice of pump-laser detuning. Although the frequency spacing between the three-photon-gain peak and the pump-laser frequency decreases with decreasing pump-resonance detuning, as expected from the dependence of the generalized Rabi frequency $\Omega' = (\Delta/|\Delta|)(\Delta^2 + \Omega^2)^{1/2}$, the Raman shift is ~1.7 GHz, independent of pump-resonance detuning. In the bottom spectrum, the three-photon gain

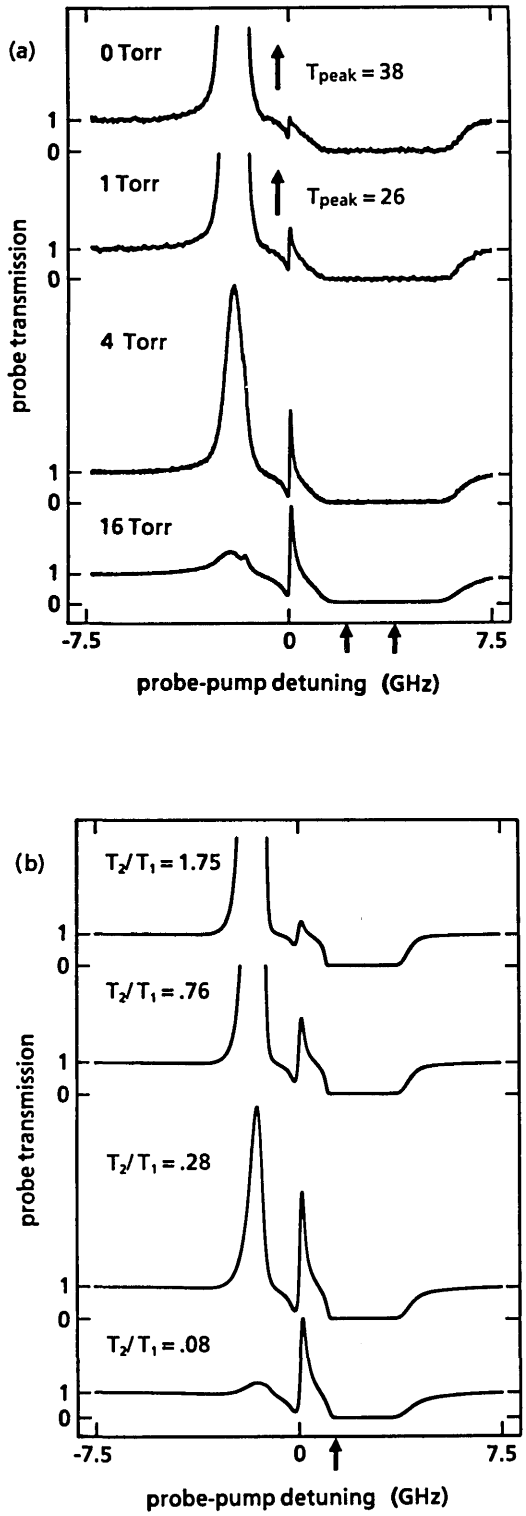

Fig. 4. (a) Measured probe transmission spectra for several values of the helium buffer-gas pressure. As the pressure is increased, the gain due to the three-photon effect decreases, while that of the nearly degenerate feature increases. The left and right arrows below the abscissa indicate the approximate locations of the sodium $3s_{1/2}(F = 2) \to 3p_{3/2}$ and $3s_{1/2}(F = 1) \to 3p_{3/2}$ hyperfine transitions, respectively, for the zero-velocity group of atoms. (b) Theoretically predicted transmission spectra for values of T_2 corresponding to the buffer-gas pressures in (a). The arrow below the abscissa indicates the transition frequency for the zero-velocity group of atoms. The parameter values used for these plots are $T_2{}^* = 0.18$ nsec, $\Omega T_2{}^* = 1.4$, $\Delta_0 T_2{}^* = -1.5$, and a beam-crossing angle of 7.5°.

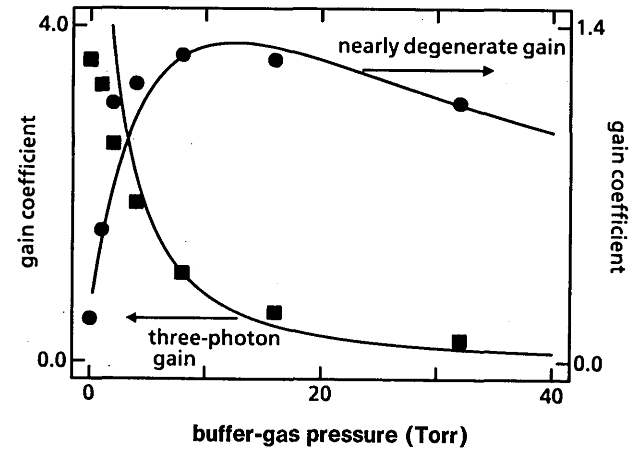

Fig. 5. Measured peak gain coefficient for the three-photon feature (squares) and the nearly degenerate feature (circles) versus helium buffer-gas pressure. The solid curves are fits of Eqs. (11) and (4)–(9), respectively, to the data, with Eq. (15) relating the buffer-gas pressure to the dipole-dephasing time T_2.

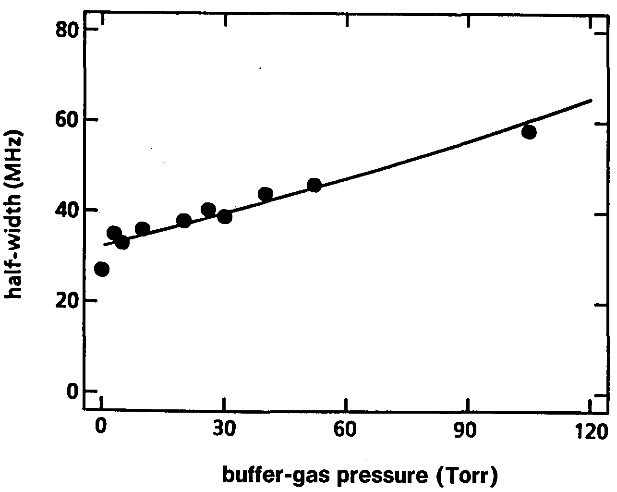

Fig. 6. Half-width of the nearly degenerate feature versus helium buffer-gas pressure. The experimental pump detuning from resonance for the zero-velocity group of atoms is approximately −6.5 GHz. The solid line is a theoretical fit to the data taking $\Delta_0/\Omega = -5$ and a beam-crossing angle of 1.6°.

appears on the high-frequency side of the pump-laser frequency, and the nearly degenerate resonance line shape has changed sign. We define zero detuning from resonance as that pump-laser frequency at which the nearly degenerate resonance changes sign [see Eq. (7)]. Spectra at higher pump-laser frequencies are more difficult to obtain than those used here because the pump beam tends to break up into self-focused filaments.

We studied the effects of collisional dephasing by introducing varying amounts of buffer gas into the sodium cell. In general, the dipole-dephasing time is given by[35]:

$$T_2 = [\pi(\Delta\nu_{\text{nat}} + \Delta\nu_{\text{self}} + CP)]^{-1}, \tag{15}$$

where $\Delta\nu_{\text{nat}}$ is the radiative linewidth, $\Delta\nu_{\text{self}}$ is the broadening due to sodium–sodium collisions, P is the foreign buffer-gas pressure, and C is a constant that depends weakly on the cell body temperature. For the buffer gas used to introduce collisional dephasing we chose helium because its low atomic mass minimizes the effects of velocity-changing collisions.[36]

In Fig. 4(a) we show a sequence of measured spectra in which the intensity transmission of the probe wave is plotted as a function of the probe–pump detuning for several different buffer-gas pressures. The pump laser is tuned to a frequency that maximizes the three-photon gain. The similarity to the theoretical spectra in Figure 4(b) is striking. The three-photon gain is largest in the absence of buffer gas, the corresponding measured intensity transmission being T = 38 (i.e., 3800%). As the buffer-gas pressure is increased, the amplitude of the three-photon gain decreases, and the gain due to the nearly degenerate resonance increases. [In the bottom spectrum in Fig. 4(a), the three-photon gain is small enough that the Raman Stokes gain peak is visible.]

In Fig. 5 we have plotted the peak value of the gain coefficient for both the three-photon resonance and the nearly degenerate resonance as a function of buffer-gas pressure. The data points are extracted from spectra similar to those in Fig. 4(a). The solid lines represent the gain as predicted by the simple expressions [Eqs. (4)–(9) and (11)] based on a theory that ignores Doppler broadening. In applying the theory to the data of Fig. 4, we have taken $\Delta = -3.2$ GHz, Ω = 1.3 GHz, and the coefficients in Eq. (15) to be $\Delta\nu_{\text{self}} = 5.0$ MHz and C = 10 MHz/Torr. The gain due to the three-photon effect decreases rapidly with increasing buffer-gas pressure because collisional-dephasing effects damp out the Rabi oscillations. Since the three-photon-resonance line shape is affected by thermal motion, agreement between theory and experiment should be only approximate. The gain due to the nearly degenerate resonance grows initially with increasing pressure and subsequently decreases. Since the nearly degenerate profile is insensitive to atomic velocities, the experimental results agree with the predictions of the stationary-atom theory.

The measured width of the nearly degenerate profile is plotted versus helium buffer-gas pressure in Fig. 6 for a pump frequency detuned from the center of the Doppler profile by $\Delta_0 \approx -6.5$ GHz. The solid line shows the theoretical half width in the presence of Doppler broadening for $\Delta_0/\Omega = -5.0$ and a beam-crossing angle of $\theta = 1.6°$. This angle is in approximate agreement with the measured value. At zero buffer-gas pressure, the half-width is about 30 MHz. For the case of exactly copropagating waves (θ = 0), the theory predicts a half-width of 10 MHz. The half-width of this feature broadens rapidly with increasing crossing angle because the effects of Doppler motion on the waves at ω_1 and ω_3 no longer cancel exactly when $\theta \neq 0$.

5. CONCLUSION

We have investigated both theoretically and experimentally the absorption spectrum of a probe wave interacting with a strongly driven atomic vapor when the probe wave is nearly copropagating with the driving field. In particular, we have taken into account the Doppler motion of the atoms and shown how the dephasing collisions produced by a buffer gas affect the absorption spectrum. In this geometry, the three spectral features that occur in the non-Doppler-broadened case are still present when there is thermal motion. The feature near the pump-wave frequency resulting from population oscillations retains its narrow line shape if the probe wave is exactly copropagating with the pump wave; however, even a small nonzero crossing angle causes a substantial broadening. On the other hand, Doppler motion significantly broadens the three-photon-gain feature. In addition, the probe–pump detuning at which the peak three-photon gain occurs is smaller than the generalized Rabi frequency for the zero-velocity group of atoms. Even in the presence of Doppler broadening, we observed a probe-wave amplification of 38. The experimental spectra agree well with theoretical predictions based on a two-level-atom model. We also observed Raman Stokes and anti-Stokes resonances that result from transitions from the ground-state hyperfine levels.

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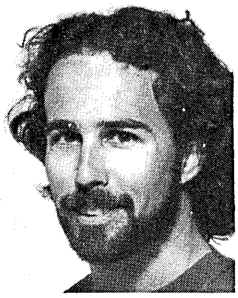

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