Dipole-Dipole Broadened Line Shape in a Partially Excited Dense Atomic Gas

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We have experimentally investigated the resonance line shape of a partially excited dense atomic rubidium vapor at a density where the binary collision approximation starts to break down. The overall spectral shape is well represented by a simple expression for the dielectric function that includes an excitation-dependent linewidth and shift. The results for the excitation dependence of the linewidth agree with an estimate based on a quasistatic picture of the collisions. [S0031-9007(96)01439-1]

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The optical properties of a dense atomic gas at its resonance frequency are of fundamental importance for astrophysics, plasma physics, and in a variety of nonlinear optical concepts such as lasers without inversion [1], optical bistability [2,3], and self-induced transparency [4]. The dominant interaction in such a gas is the resonant dipole-dipole interaction between excited and unexcited atoms giving rise to self-broadening of the resonance line. The range of the interaction is given by the Weisskopf radius $\rho_w = \mu/\sqrt{v_0}$; here μ is the resonance transition dipole moment and v_0 is the most probable atomic velocity in the gas. At room temperature one has $\rho_w \approx 20$ nm, a value that is of the order of the average interatomic distance at densities ($N > 10^{17}$ cm^{-3}) that are relevant for applications in nonlinear optics [1,2,4]. In that case, every atom is permanently in a state of collision and the usual models (impact [5], quasistatic [6], unified line broadening [7]) for the line shape cannot be applied since they all assume that the collisions are binary. At these densities a multiperturber approach is clearly required. Recently, it has even been argued that for the case of self-broadening many-particle effects have to be taken into account at all densities [8].

Nevertheless, since multiperturber theory is extremely difficult [8,9] one usually chooses to apply the binary collision approximation anyway and to use the impact model. The optical response of the dense atomic gas then results in a Lorentzian spectral line having a self-broadened width $\Gamma = N v_0 \rho_w^2$ proportional to the atomic density N. The center of the atomic resonance line of the dense gas is shifted due to local-field effects. The size of this Lorentz local-field shift $\Delta\omega_L$ [10–12] is predicted to depend on the degree of excitation; this dependence lies at the heart of the current interest in nonlinear optics of dense atomic vapors (see, for instance, Ref. [1], and references therein).

To our knowledge only a single experiment has been performed on a dense atomic vapor with a substantial fraction of excited particles [12]. In that experiment the Lorentz and collisional shifts were determined for atomic densities $N < 1.5 \times 10^{17}$ cm^{-3} in a four-wave mixing experiment with a single laser for the pump and probe beams. In the present Letter we report that novel information can be obtained by using *independent* light sources for exciting and probing the vapor.

In our experiment we probe the reflectivity of a partially excited high-density Rb vapor on the $^2S_{1/2} \rightarrow {}^2P_{3/2}$ transition. The self-broadened width (HWHM) of this transition is given by $\Gamma = 2\pi \times 5 \times 10^{-8} N$ cm^3 s^{-1} [5]. We choose the vapor density high enough ($N \gtrsim 10^{17}$ cm^{-3}) so that hyperfine structure, Doppler broadening, isotopic composition, and effects of nonlocal polarization [13] are irrelevant. Since $N\rho_w^3 \gtrsim 1$ for $N \gtrsim 10^{17}$ cm^{-3}, we exceed the limit of the binary approximation. As shown in Fig. 1 we excite the Rb vapor with the output of a Ti:sapphire laser ($P = 200$ mW) that is tuned into the far wing of the D_2 line (detuning $\Delta/2\pi \approx 100$ GHz) so that the vapor is quasihomogeneously excited (absorption length $\gg \lambda$). The reflectivity of the vapor is probed using a single-frequency AlGaAs diode laser tunable over 80 GHz. Our cell has an evacuated section in front that serves to avoid turbulence and thus to maintain the quality of the optical beams, while the rear section contains the Rb vapor and is heated. The laser beams are superposed at the thin sapphire window that separates the two

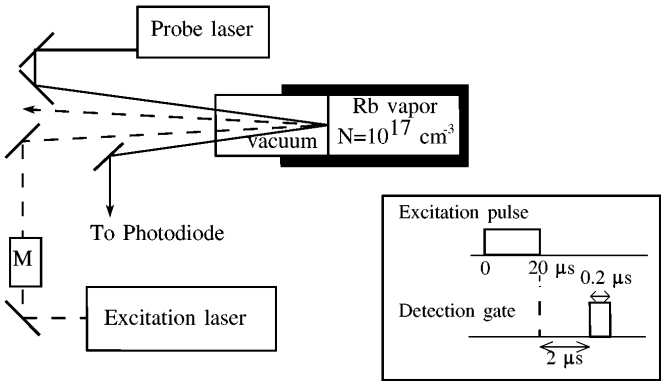

FIG. 1. Experimental setup to probe the excitation-modulated reflection spectrum of a high-density atomic vapor. The inset shows the timing of the excitation pulse and the detection gate. *M* indicates a shutter.

sections of the cell, impinging at near-normal angles of incidence [14]. The intensity of the excitation laser at the interface can be varied between 1 and 30 W/cm^2 [15]; the intensity of the probe at the interface is kept far below the saturating value.

Typical experimental results are shown in Fig. 2 for a Rb density of 8×10^{16} cm^{-3}. The various features of the reflectivity spectrum persist throughout the range of atomic densities ($8 \times 10^{16} < N < 2 \times 10^{17}$ cm^{-3}) that were probed in the experiment. Curve (*a*) shows the low-density Rb absorption spectrum and provides an absolute frequency reference. The conventional reflection spectrum of the high-density vapor near the interface is shown in curve (*b*). Curve (*c*) shows the excitation-modulated reflection spectrum of Rb with the Ti:sapphire laser detuned 100 GHz into the red wing of the resonance line. Here the probe is delayed (typical delay time ≈ 2 μs) with respect to the excitation laser and the excitation-induced change in reflectivity is sampled in a time interval of 0.2 μs. The cycle of excitation and probing is repeated every millisecond. Because of radiation trapping the modulated reflectivity decays in approximately 5 μs (for this detuning), in rough agreement with the decay time of the fluorescence in a homogeneously excited dense vapor [17]. We estimate that at the time we probe the vapor a few percent of the Rb atoms are still in the excited state. As can be seen the modulated reflectivity spectrum displays a surprising "dip" at the center of the resonance. Since the exciting laser is tuned 100 GHz into the wing of the resonance line this feature is not a result of spectral hole burning nor can it reflect a coherent artifact [19] since the probe is delayed relative to the exciting laser. If the excitation laser is coincident with the probe laser and tuned into the far wing of the resonance, the probe spectrum is *identical* with that obtained with delay [Fig. 2(c)], implying that coherent effects are unimportant also in this case.

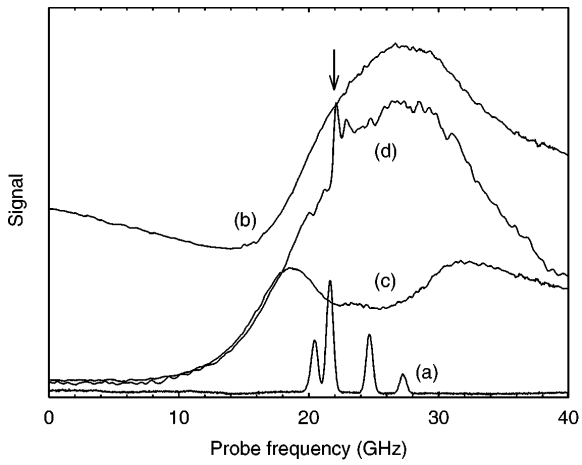

FIG. 2. Reflectivity spectra on the D_2 line for a high-density ($N = 8 \times 10^{16}$ cm^{-3}) Rb vapor. For details see text.

If the exciting laser is tuned near resonance and the probe is delayed, the modulated reflectivity is buried in the noise. If, for this tuning of the exciting laser, the two lasers interact simultaneously with the vapor, the spectrum of Fig. 2(d) results. The line shape is now quite similar to that of the reflection spectrum of the unexcited vapor [Fig. 2(b)] except for a narrow resonance at line center (see arrow); the latter reflects the coherent interaction between the two laser beams. The qualitative differences between the spectra for small and large values of the detuning Δ of the exciting laser are related to the thickness of the excited layer ℓ compared to the wavelength and to the decay time of the excited-state population. For $\Delta \gg \Gamma$ we have $\ell \gg \lambda$, and for the calculation of the reflectivity we can treat the system as spatially homogeneous. For this case, the excited-state population decays slowly as a result of radiation trapping. In the other limit we have $\ell \ll \lambda$ and the reflectivity of the sample depends on details of the spatial distribution of excited atoms, which we do not know. This distribution decays rapidly ($\lesssim$150 ns), the decay being governed by higher-order Holstein modes [20]. Since this situation is rather complex we do not pursue it here.

We interpret our experiment as a measurement of the linear response of a partially excited vapor to be described in terms of a dielectric coefficient ϵ. Because self-broadening gives rise to a Lorentzian line shape in both the line core and line wings [8] we write, in the absence of excitation [12],

$$\epsilon(\Delta\omega) = 1 + \frac{kN}{\Delta\omega + \Delta\Omega - i\Gamma}, \qquad (1)$$

with $\Delta\omega$ the probe detuning from resonance, $\Delta\Omega$ the line shift, and Γ the self-broadened linewidth. For $\Delta\Omega$ we can write $\Delta\Omega = \Delta\omega_c + \Delta\omega_w + \Delta\omega_L$ with $\Delta\omega_c$ the collisional shift, $\Delta\omega_w$ the wall shift [21], and $\Delta\omega_L$ the Lorentz local-field shift. The constant k is given by $k = fcr_e\lambda$; here r_e is the classical radius of the electron and f the oscillator strength of the transition. The local-field shift and the collisional width and shift are all proportional to the atomic density; theory gives $\Delta\omega_L = kN/3$ and $\Gamma = kN\sqrt{g_g/g_e}$ [5], with g_g and g_e the degeneracies of the ground and excited state, respectively.

To account for the partial excitation of the vapor we follow the approach of Manassah [22] and replace the atomic density N in both Eq. (1) and in the expression for the Lorentz shift by $(N_g - g_g/g_e N_e)$. Except for a trivial reduction of the reflectivity, partial excitation of the vapor results in only a small distortion of the reflectivity spectrum [10], much too little to account for the features shown in Fig. 2, in particular, the dip. Below we will show that such features can be explained if we assume the *width* to be excitation dependent. First we will argue that such an assumption is quite reasonable within the

binary collision approximation in the context of existing line broadening theories.

In an impact description of self-broadening one does *not* expect a dependence of the linewidth on the degree of excitation since the collisional width is proportional to $N_g + N_e = N$: there is symmetry between the ground and excited states [23]. This symmetry is absent in a quasistatic description of self-broadening since one has to take the different polarizabilities of the ground and excited states into account. For an $S \rightarrow P$ transition the excited state has a much higher polarizability than the ground state so that atoms in the excited state are much more strongly perturbed (by ground-state atoms) than their ground-state counterparts (by the excited-state atoms). When the vapor is strongly excited the fraction of ground-state atoms is diminished and *the linewidth will be reduced.*

In reality the situation is more complex; due to fine-structure coupling we will also populate the $^2P_{1/2}$ state when exciting on the D_2 line. Since the self-broadened widths of the two D lines are different [5] there is an additional channel through which an excitation dependence of the self-broadened linewidth can arise. This channel is already present in the impact limit, yielding an impact linewidth $\widetilde{\Gamma}^i$ [22,23]

$$\widetilde{\Gamma}^i(D_2)/\Gamma^i(D_2) = (N_g + N_{3/2} + \xi^{-1}N_{1/2})/N\,, \quad (2)$$

with $\Gamma^i(D_2)$ the linewidth in the zero-excitation limit and $\xi \approx \sqrt{2}$ the ratio of the widths of the D_2 and D_1 lines in the same limit [5]. At the prevailing Rb densities fine-structure mixing is very efficient (mixing time ≈ 40 ns) and we estimate the ratio $N_{3/2}/N_{1/2} \approx 0.9$. With this ratio fixed the excitation dependence of the linewidth can be written in terms of a *single* parameter $n = N_{3/2}/N$. In the impact limit we then arrive at $\widetilde{\Gamma}^i(D_2)/\Gamma^i(D_2) = (1 - 0.264\,n)$. At saturation we have $N_g = \frac{1}{2}N_{3/2}$ ($n = 0.4$) yielding $\widetilde{\Gamma}^i(D_2)/\Gamma^i(D_2) = 0.9$. So the impact linewidth is reduced by at most 10%.

In the quasistatic regime the perturbation of the excited state by ground-state particles dominates and we can write $\widetilde{\Gamma^{qs}}(D_2)/\Gamma^{qs}(D_2) = N_g/N = (1 - 1.9\,n)$. Again the linewidth will decrease when the vapor is partially excited; at saturation $\widetilde{\Gamma^{qs}}(D_2)/\Gamma^{qs}(D_2) = 0.21$, a much larger reduction than found in the impact limit. Note that in the quasistatic limit the atoms in the $^2P_{1/2}$ state affect the linewidth only through the decrease of the ground-state population resulting from the collisional redistribution over the excited levels.

Independent of a particular collision model, it is thus plausible to write

$$\widetilde{\Gamma}(D_2) = \Gamma(D_2)[1 - \alpha\, n]\,, \quad (3)$$

with α a free parameter and n as defined above. Using this *ansatz* we have fitted the calculated excitation-modulated reflection spectrum of the Rb vapor to the experiment. With $\alpha = 2.0$ we obtain the best fit; this value is very close to the quasistatic value α^{qs} and very different from the impact value α^i. The results for $N \approx 2 \times 10^{17}$ cm^{-3} [$\Gamma(D_2)/2\pi \approx 11$ GHz] are shown in Fig. 3. Clearly, the essence of the spectral behavior in the experiment is reproduced by our model. Note that an excitation dependence of the linewidth as in Eq. (3) is a prerequisite for such agreement.

In order to arrive at the calculated spectrum of Fig. 3 that agrees so nicely with the shape of the experimental spectrum, we also had to introduce into our model an excitation dependence of the *line shift*: $\widetilde{\Delta\Omega} = \Delta\Omega(1 - \beta\, n)$. It is reasonable to do so since one of the components of the line shift, viz., the Lorentz local-field shift is expected to be excitation dependent [22]. Since the relative importance of the various contributions to the line shift (Lorentz shift, collisional shift, wall shift) is not known one cannot predict a value for the parameter β; the experiment yields $\beta = 0.6 \pm 0.3$. For the Lorentz shift on an $S \rightarrow P$ transition theory predicts a twice larger β value [22].

In all our experiments the fraction of excited atoms was small (at most a few percent). In that limit one may argue that the excitation-modulated signal has a magnitude that is proportional to the degree of excitation and that its spectral dependence is independent of the degree of excitation. Experimentally, we have studied the dependence of the amplitude and width of the diplike feature in the modulated reflectivity spectrum on the detuning of the excitation laser from resonance. The results are shown in Fig. 4 for a density $N = 2 \times 10^{17}$ cm^{-3}.

The amplitude S of the diplike feature reflects the excited-state fraction at the time that the vapor is probed;

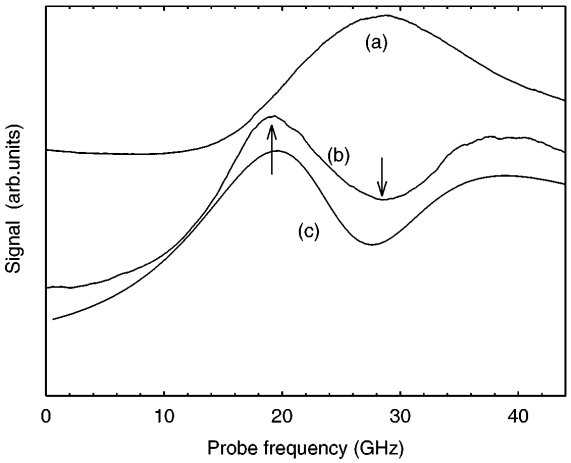

FIG. 3. Reflectivity spectra for $N(\mathrm{Rb}) \approx 2 \times 10^{17}$ cm^{-3} at $\Delta/\Gamma \approx 35$. Curve (*a*) shows the ordinary reflection spectrum as measured by the probe laser. The other curves show the experimental [curve (*b*)] and calculated [curve (*c*)] excitation-modulated spectra. The arrows indicate the points between which the width of the diplike feature is determined.

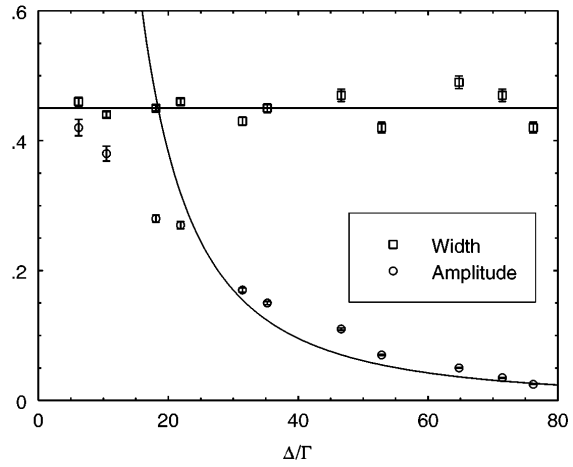

FIG. 4. Amplitude (circles) and width (squares) of the excitation-modulated reflectivity feature as a function of the normalized detuning of the pump laser from resonance. The amplitude is normalized on the response at zero detuning; the width is given as a fraction of 2Γ.

it slowly decreases with increasing detuning Δ. For large detunings ($\Delta \geq 30\Gamma$) the layer of excited atoms is bulklike and the excitation survives beyond the temporal delay between the laser pulses due to radiation trapping, as explained before. In this regime the variation of S with Δ is well described by a Lorentzian $S \propto (\Delta^2 + \Gamma^2)^{-1}$ reflecting the excitation probability. At detunings $\Delta \leq 30\Gamma$ the absorption length is smaller and the distribution of excited atoms is inhomogeneous; this distribution decays rapidly being governed by higher-order Holstein modes [20]. As a result there is a reduction of the fraction of excited atoms and thus of the signal strength. The width of the diplike feature in the modulated spectrum, defined as the frequency separation between the center of the dip and the peak on its short wavelength side (see Fig. 3), is independent of the detuning of the excitation laser (see Fig. 4), its intensity, or its polarization. This indicates that the line shape of the modulated spectrum is indeed independent of the degree of excitation as argued above.

In conclusion, we have experimentally shown that the linewidth of a dipole-dipole broadened line depends on the degree of excitation of the atomic vapor. Our results can be understood in terms of binary, quasistatic collisions, and this description is valid over the full spectral line. This is surprising since the Weisskopf radius is of the order of the interatomic distance at the densities that are used. We have also shown that the line shift has an excitation-dependent part. Our results imply that the various predictions that have been made for a dense vapor with dipole-dipole interactions need to be reinvestigated [1,2,4].

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