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Power broadening and Doppler effects of coherent dark resonances in Rb

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Using a phase-locked laser pair we have observed dark resonances with linewidths below 30 Hz in a rubidium cell filled with neon as buffer gas. A model allowing for pressure broadening correctly reproduces the dependence of the width on the laser intensity. Consideration of velocity changing collisions reveals the absence of Doppler effects in the position and width of the dark resonance at high buffer-gas pressure.

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The quantum interference phenomenon of coherent population trapping (CPT) has been studied in multilevel atomic systems in a great variety of laser-atom combinations [1]. The trapping of population into the coherent ground-state superposition is frequently detected as a decrease in absorption or fluorescence of the sample. In a three-level Λ system, as shown in Fig. 1, the dark resonance (DR) corresponds to the condition where the frequency difference of the two lasers coincides with the ground-state energy spacing, $\omega_{L1}-\omega_{L2}=\omega_{HF}$. Several studies have investigated the parameters which control the width of the dark resonance and its contrast. In a low-pressure vapor cell or in an atomic beam the resonance width is primarily limited by time-of-flight decoherence [1]. In experiments employing two laser fields and neon as buffer gas a spectacular narrowing of the Cs dark resonance width to below 50 Hz has been reported [2], comparable to what is achieved in microwave excitation [3]. Similarly, resonance widths of only 150 Hz were reported for Rb [4]. The narrowing optimizes at a critical buffer-gas pressure, indicating that time-of-flight and pressure broadening control the width at low laser fluence [2]. Theoretical models predict a much steeper increase in power broadening of the dark resonance than is observed experimentally [5]. When applied to the experiment of Cs in Ne [2], theory predicts the broadening to be two orders of magnitude faster than is observed. In addition, Doppler effects on the structure, position, and width of the DR are conspicuously absent in the presence of buffer gas. Theoretical models for the velocity and pressure broadening [6] predict CPT widths incompatible with the observation of ultranarrow resonances in cells with medium gas pressure.

Relaxation widths of only 1.3 Hz were achieved in paraffin-coated cells without buffer gas [7,8]. In these experiments only a single light beam was used to establish the coherence among degenerate Zeeman levels, hence Doppler effects played no role. As we will show below, Doppler effects should be significant for nondegenerate ground states in a Λ system.

Here we report a study of the width and contrast of the dark resonance by comparing theoretical models with experimental data obtained for rubium in a neon-buffered cell. Our experiment (see Fig. 2) employs two phase-locked diode lasers to drive transitions of the $D2$ line that connect two hyperfine ground-state levels with the hyperfine manifold of the $^2P_{3/2}$ state. A master laser (rms bandwidth of 60 ± 4 kHz [9]) is actively frequency locked to a Rb crossover line. A second laser is kept at a variable detuning from the master by employing a digital phase-lock loop as introduced by Prevedelli *et al.* [10]. The difference frequency of the lasers can be continuously tuned while being stabilized to an rms-phase error of better 0.4 rad over periods of hours.

The two laser beams were merged in a short fiber, circularly polarized, and propagated collinearly through the magnetically shielded room-temperature Rb absorption cell containing 6.5 kPa neon. Configurations with $1/e^2$ laser-beam diameters of $2r_0=1.6$ cm and $2r_0=2.9$ cm were investigated [11]. The second laser was phase modulated at 1 kHz with a modulation index of $\Phi_M/2\pi=1/12$ and a lock-in amplifier monitored the modulated transmitted intensity. Measurements of the Zeeman splitting of the DR features revealed a residual field of ≈5 μT. All following data refer to the DR observed for the ground-state Zeeman components with $m_F=0$.

A typical dark resonance trace recorded at $I_0=3$ μW/cm^2 is shown in Fig. 3. The two traces give the in phase and the quadrature component of the frequency modulation signal. The resonance positions were found to be shifted from their nominal values commensurate with the pressure shift predicted in the literature for ^{87}Rb in Ne [3]. DRs were recorded at intensities ranging from $I_0=1$ to 75 μW/cm^2 to deduce the dependence of the linewidth and the contrast of the dark resonance on the laser intensity. These

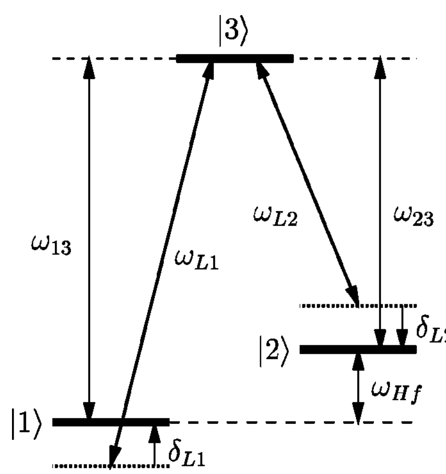

FIG. 1. Notation of the frequencies of the Λ scheme. In our experiments on ^{85}Rb the labels $|1\rangle$, $|2\rangle$, and $|3\rangle$ refer to the states $|^2S_{1/2},F=2,m_F=0\rangle$, $|^2S_{1/2},3,0\rangle$, and $|^2P_{3/2},3,\pm1\rangle$, respectively.

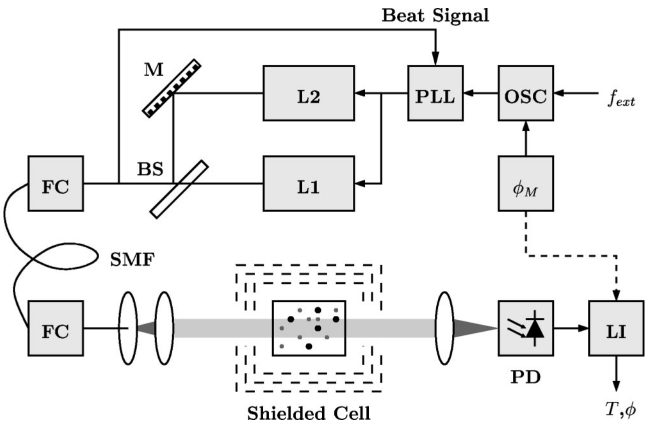

FIG. 2. Schematic of the experimental setup. Two external-cavity diode lasers (L1 and L2) are phase-locked (PLL). The modulated (Φ_M) oscillator (OSC) is externally tuned (f_{ext}) while a lock-in amplifier (LI) monitors the transmitted intensity (M, mirror; BS, beam splitter; FC, fiber coupler; SMF, single-mode fiber).

results are shown in Figs. 4 and 5 for ^{85}Rb. Similar results were obtained for the ^{87}Rb isotope. No noticeable shift in the resonance position with laser intensity appeared. A linear intensity dependence of the resonance width

$$\Gamma_{\mathrm{DR}}=\Gamma_R+\alpha I_0 \tag{1}$$

was found. The dependence of the resonance height, defined as the transmitted intensity at the DR peak relative to the Doppler broadened absorption curve, was fitted as

$$H_{\mathrm{DR}}=CI_0^n. \tag{2}$$

At low intensities a quadratic dependence ($n=2$) is expected due to the linear dependence of the DR susceptibility on intensity. The fit parameters for Eqs. (1) and (2) to these data are listed in Table I.

As we show in a separate paper [12] a theory that includes Dicke-type narrowing [13] and laser fluctuations predicts for the DR width at low intensity

$$\Gamma_{\mathrm{DR}}=\Gamma_R+\frac{2\Omega^2}{\gamma+\Delta_{13}^c+\Delta_{23}^c}. \tag{3}$$

Here Γ_R is the DR width due to ground-state coherence relaxation and γ refers to the optical decay rate from level $|3\rangle$.

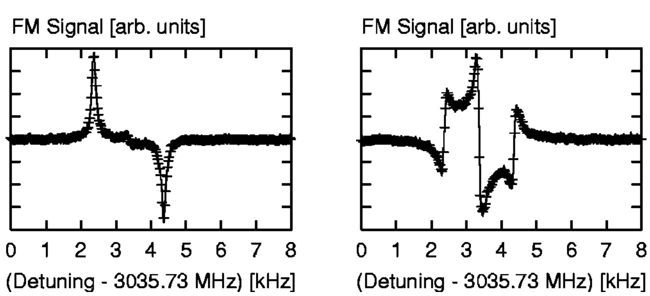

FIG. 3. Dark resonance signal observed at $I_0=3$ μW/cm^2 using the small diameter beam (left, in-phase signal; right, quadrature component).

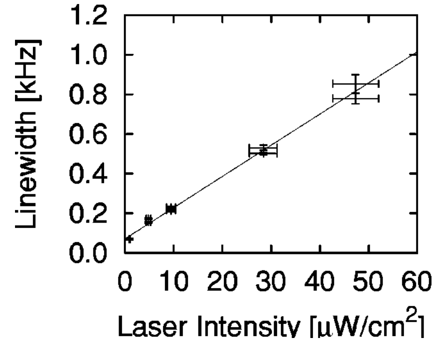

FIG. 4. Dependence of the dark resonance width on laser intensity for ^{85}Rb (small-diameter beam).

This formula is similar to that given by Arimondo [5], but it includes the terms Δ_{13}^c and Δ_{23}^c that characterize the collisional broadening of the $1\leftrightarrow2$ and $2\leftrightarrow3$ transitions. Owing to their smallness, we neglect in the denominator of the second term in Eq. (3) additional terms, such as fluctuations of the laser frequency difference and laser bandwidth. The collisional broadening of the $D2$ lines of Rb at 6.5 kPa of Ne is $\Delta_{13}^c=\Delta_{23}^c=2\pi\times263$ MHz [14]. Taking into account an approximate value of 1/4 for the Clebsch-Gordan coefficients for the $|^2S_{1/2},F=2,m_F=0\rangle\leftrightarrow|^2P_{3/2},3,\pm1\rangle\leftrightarrow|^2S_{1/2},3,0\rangle$ transitions of the ^{85}Rb Λ scheme, we obtain for the Rabi frequency

$$\Omega^2=(2\pi)^2\times5.45\times10^9\times\langle I\rangle\frac{1}{s^2}, \tag{4}$$

where we have introduced a spatially averaged intensity $\langle I\rangle$, given in units of μW/cm^2. Inserting this value into Eq. (3) we obtain

$$\Gamma_{\mathrm{DR}}=\Gamma_R+2\pi\times20.7\times\langle I\rangle\mathrm{Hz}. \tag{5}$$

Considering that $\langle I\rangle<I_0$, the predicted magnitude of the intensity dependence is in good agreement with the fitted values of $\alpha/(2\pi)$ (see Table I). Previous models for the intensity dependence [5] did not include the effect of pressure broadening on the effective rate of optical pumping and hence predicted a much steeper increase of the DR width with laser intensity.

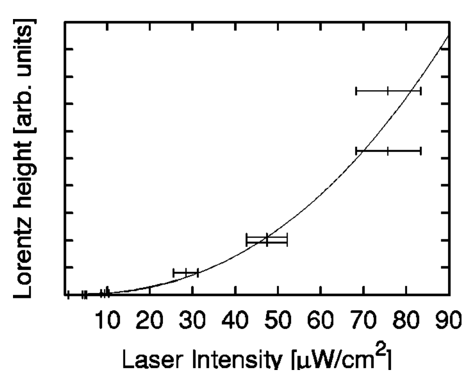

FIG. 5. Dependence of the dark resonance height on laser intensity for ^{85}Rb (small-diameter beam).

TABLE I. Fit parameters of Eqs. (1) and (2) obtained for ^{85}Rb and ^{87}Rb using the small-diameter beam profile.

	^{85}Rb	^{87}Rb	Dim.
$\Gamma_R/(2\pi)$	70.3 ± 11.2	69.1 ± 27.1	Hz
$\alpha/(2\pi)$	15.7 ± 0.4	13.9 ± 0.9	Hz cm$^2/\mu W$
n	2.35 ± 0.32	2.12 ± 0.15	

A second series of studies was carried out with the large beam configuration. In this case the width observed at the lowest intensity ($I_0 = 300\,\mathrm{nW/cm^2}$) was found to lie near 30 Hz. The measurement of this DR was repeated on five consecutive days yielding widths of 28 ± 2, 36 ± 4, 43 ± 3, 27 ± 2, and 37 ± 3 Hz, respectively, under nominally identical conditions. As an example we show one result in Fig. 6. Since Γ_R is primarily determined by diffusive loss of coherence from the observation region, the experimental value from Table I can be scaled to the large-diameter beam experiment, predicting a dark resonance width of 27.4 ± 3.7 for the conditions used in Fig. 6.

In the following we quantify Doppler effects that govern the width and position of the DR for a pure Rb sample at room temperature. The absence of these effects in our experimental data is finally connected to Dicke-type narrowing due to collisions with the buffer gas. The detunings of the laser frequencies from the atomic frequencies ω_{12} and ω_{23} are denoted by δ_{L1} and δ_{L2}. The Raman condition $\delta_{L1} = \delta_{L2}$ determines the position of the DR peak for *stationary* atoms. Considering the Doppler effect due to the thermal motion of the Rb atoms at room temperature, it can be shown [12] that for detunings <1 GHz from resonance the major contribution to the observed DR-signal intensity is from atoms whose Doppler-shifted laser frequencies fall within the natural linewidth, $\gamma = 2\pi\times 6$ MHz. The velocity of resonant atoms is

$$v_r = c\,\frac{\delta_{L2}}{\omega_{\mathrm{opt}}}, \tag{6}$$

where $\omega_{\mathrm{opt}} \approx \omega_{L1} \approx \omega_{L2}$. Since the two lasers experience slightly different Doppler shifts when interacting resonantly

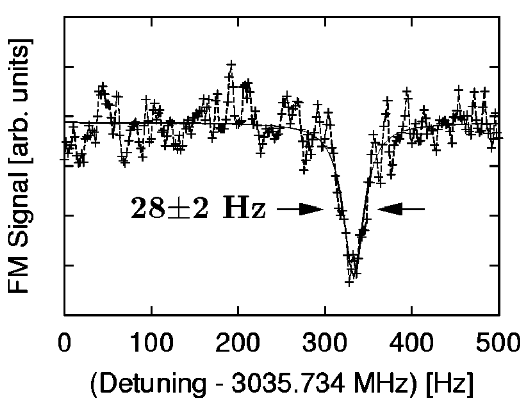

FIG. 6. Dark resonance signal observed at $I_0 = 300\,\mathrm{nW/cm^2}$ using the large-diameter beam (the peak corresponds to the resonance at $\omega_{\mathrm{DR}}/2\pi + f_{\mathrm{mod}}$).

with the same class of moving atoms, the resonance condition for moving atoms is different from that for atoms at rest:

$$\omega_{\mathrm{DR}}^{\mathrm{lab}} = \frac{\omega_{HF}}{1-\dfrac{v_r}{c}} \approx \omega_{HF}\left(1+\frac{v_r}{c}\right). \tag{7}$$

In our case the Doppler-broadening, $\Gamma_{\mathrm{Dopp}} = 620$ MHz, is large compared to the natural linewidth. In this high-temperature limit the velocity spread of atoms contributing to the DR is roughly determined by the natural linewidth

$$\frac{\Delta v_r}{c}\,\omega_{\mathrm{opt}} \approx \gamma \tag{8}$$

and should result in a Doppler contribution to the DR width of

$$\Gamma_{\mathrm{DR,Dopp}}^{\mathrm{lab}} = \frac{\gamma}{\omega_{\mathrm{opt}}}\,\omega_{HF} \approx 2\pi\times 47\ \mathrm{Hz}. \tag{9}$$

This high value is incommensurate with the narrowest widths observed here.

A second feature expected from the Doppler effect is the sensitivity of the DR position to the detuning from optical resonance. Applying Eqs. (6) and (7) we expect a shift of the DR position when the absolute frequency of one of the lasers is detuned from the atomic resonance by δ_L,

$$\Delta\omega_{\mathrm{DR}}^{(\mathrm{lab})}/\delta_L = \frac{\omega_{HF}}{\omega_{\mathrm{opt}}}. \tag{10}$$

A shift of 7.9 Hz/MHz in ^{85}Rb and 17.8 Hz/MHz in ^{87}Rb, respectively, is predicted from this equation. We searched for this effect, exploring detunings up to $\delta_{L2} = 800$ MHz. Our experiments show that such shifts could at most amount to 0.5 Hz/MHz.

As a third example of Doppler phenomena the effect inherent in Eq. (10) should give rise to a splitting of the DR feature when the hyperfine splitting in the excited state is taken into account (63 and 157 MHz in the two isotopes, respectively). In Rb at room temperature where the Doppler width is much larger than the exited-state hyperfine splitting, the equivalent of Eq. (10) predicts the appearance of two dark resonances, spaced by 498 Hz in ^{85}Rb and 2795 Hz in ^{87}Rb, regardless of the detuning of one of the lasers from atomic resonance. Also this effect is suppressed in experiments conducted at high-buffer-gas pressure.

Collisions with the buffer gas cause Rb atoms to rapidly change velocity class, yet the ground-state coherence survives for typically $>10^7$ collisions [13]. When the rate of buffer-gas collisions is higher than the typical rate at which the DR develops, the individual nature of a velocity class to which an atom may belong for a short period of time is experienced by the atom merely as fluctuation in the optical frequency. For a velocity distribution with an average velocity of zero, the laser frequency fluctuations ‘‘seen’’ by the atom center around a frequency given by the laboratory frequency of the lasers, hence permitting the development of a single DR line at the laboratory energy of the ground-state hyperfine spacing. While the DR position is severely pressure shifted from the isolated atom energy spacing, the additional damping due to pressure broadening serves to reduce the power broadening of the DR width.

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