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Buffer-gas-induced linewidth reduction of coherent dark resonances to below 50 Hz

S. Brandt, A. Nagel, R. Wynands, and D. Meschede
Institut für Angewandte Physik, Universität Bonn, Wegelerstraße 8, 53115 Bonn, Germany
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When neon is introduced as a buffer gas the interaction time of cesium atoms in a vapor cell with resonant laser beams is drastically increased. Using a pair of phase-locked lasers we have observed linewidths as narrow as 42 Hz for coherent dark resonances in a cesium vapor cell. We study the influence of power and pressure broadening and systematic shifts of the resonance frequency. Our experiments demonstrate that coherent dark resonances could rival direct radio-frequency precision measurements, which have a wide range of applications in physics. [S1050-2947(97)50108-9]

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Coherent population trapping has been known since 1976 [1] and was recently reviewed by Arimondo [2]. In alkali-metal atoms this effect can occur when a coherence between the two hyperfine-split ground-state levels is established through a two-photon process. Experimentally this can be achieved with two lasers that each couple one of the ground states near resonantly to a common excited state, e.g., on the D_1 or the D_2 transition. When the laser difference frequency exactly matches the lower level splitting the atoms are optically pumped into a coherent superposition of the two lower states that does not absorb the light anymore [2]. Because a direct transition between the two ground states is dipole forbidden the ground-state coherence has an extremely long radiative lifetime. As was pointed out by Scully [3] this leads to potentially very narrow resonance lines that exhibit steep normal dispersion at vanishing absorption, a point that was demonstrated experimentally for rubidium [4] and cesium [5]. An interesting application is a different type of magnetometer of extremely high sensitivity [6].

Recent advances in the stabilization of large laser difference frequencies have allowed high-resolution studies of dispersion and absorption in the vicinity of coherent dark resonances. In a pure vapor cell, the smallest reported resonance width is 30 kHz and is mostly limited by time-of-flight broadening [5]. Ezekiel conducted a series of experiments with sodium and cesium atomic beams where resonance widths of 1.3 kHz could be seen in a Ramsey-type geometry with interaction-zone separation of up to 30 cm [7]. While Raman experiments with atomic beams allow long effective interaction times at reduced Doppler broadening they are inherently bulky, and atomic densities are much lower than in a typical vapor cell.

In the 1950s it was discovered that time-of-flight broadening of rf transitions between alkali-metal-atom ground states can largely be eliminated when a buffer gas at several mbar pressure is added (e.g., He, Ne, Ar) [8]. Because of

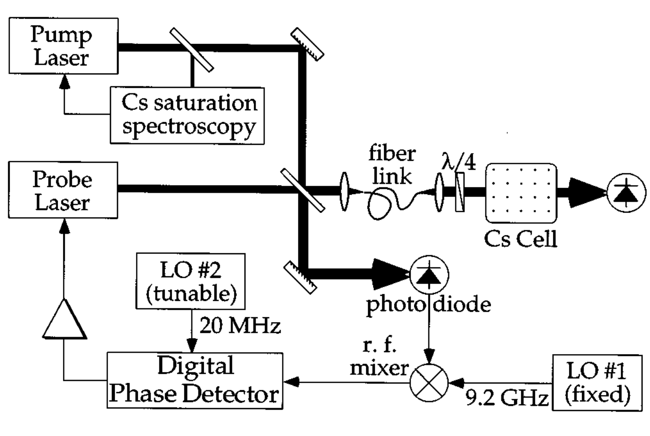

FIG. 1. Experimental setup for the observation of narrow coherent dark resonances.

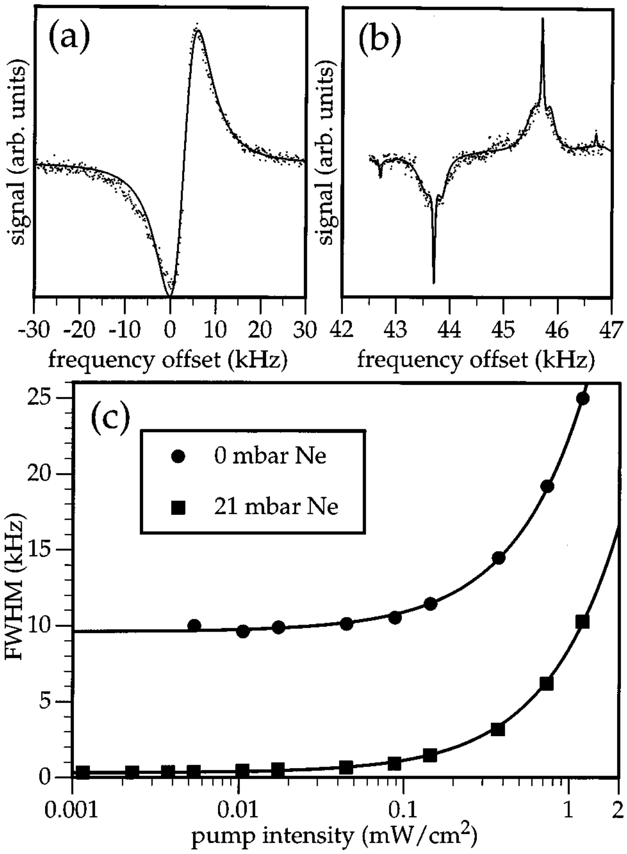

FIG. 2. Line shapes of the 0-0 resonance (a) without buffer gas, FWHM = 10.4 kHz; and (b) with 87-mbar neon buffer-gas pressure, FWHM = 42 Hz. (c) Power-broadened linewidth with and without buffer gas. The solid lines are linear fits to the data points.

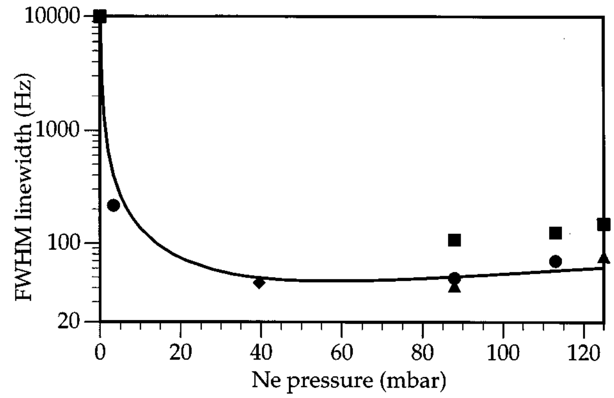

FIG. 3. Measured coherent dark resonance linewidth vs neon pressure for very low laser intensity (squares, 17 $\mu W/cm^2$; triangles, 11 $\mu W/cm^2$; circles, 6 $\mu W/cm^2$; diamond, 1 $\mu W/cm^2$). The solid line is calculated using values measured by direct excitation of rf transitions between cesium ground-state levels [16], and does not involve any adjustable parameters.

frequent collisions with buffer-gas atoms the alkali-metal atoms can only diffuse out of the laser beams and therefore allow much longer interaction times, i.e., narrower linewidths. We find it fascinating that with properly chosen buffer gas the ground-state coherence can survive more than 10^7 collisions without decay, and since the lifetime of the coherence should not depend on the way in which it is created one should expect the same beneficial effects also for coherent dark resonances.

However, there have only been two published reports on the use of buffer gas in coherent dark resonance experiments [9,10]. Ezekiel *et al.* [9] achieved a 1.6-kHz linewidth in a sodium vapor cell with a helium buffer gas. There is also an unpublished experiment by Xu [11], where helium reduced the linewidth of dark resonances in sodium from 1.3 to 0.4 MHz. Here we report on our systematic study of buffer-gas effects in Cs coherent population trapping. We are using the D_2 line at a 852-nm wavelength to excite a coherence between the two cesium ground states with a hyperfine splitting of 9.192 631 77 GHz. We find linewidth reductions by three orders of magnitude to values below 50 Hz and study systematic line shifts and residual broadening effects.

The experimental setup consists of two extended-cavity diode lasers (Fig. 1). The laser beams (called pump and probe for easier reference) are superposed on a beam splitter, and one of the output beams is focused onto a photodiode that is fast enough to detect the 9.2-GHz beat frequency between the two lasers. The beat signal is amplified and mixed with a 9.172 631 61–GHz radio frequency signal from a dielectric resonance oscillator that is locked to the 183rd harmonic of a 50.123 67–MHz synthesizer. The mixer output signal of about 20 MHz is compared with the 20-MHz output of a second, computer-tunable rf synthesizer in a digital phase and frequency detector [12]. In this way the servo loop stabilizes the phase of the probe laser through its injection current such that the laser difference frequency is exactly the sum of the synthesizer frequencies. The servo loop remains locked for many hours. Both rf synthesizers are locked to the same 10-MHz frequency reference.

The second output beam from the beam splitter is coupled into a short stretch of single-mode fiber in order to have clean and perfectly overlapping Gaussian modes. This is important because an angle of only 1 mrad between pump and probe laser beams already gives a Doppler contribution of 700 kHz to the width of the coherent dark resonance [13,5]. Both beams are circularly polarized and then pass through the cesium cell. The transmitted light is detected on a photodiode.

The cesium vapor is contained in a glass cell with a radius of 1.8 cm and a length of 2 cm. In one sidearm there is a small cesium reservoir, and another sidearm is sealed off by a glass valve. It can be connected to a gas dosing system so that different buffer-gas compositions can be prepared in the same cell.

The cell is mounted inside an arrangement of three mutually perpendicular current coil pairs that compensate static magnetic fields. A small flux density (24 μT) in the direction of laser beam propagation splits the coherent dark resonance into seven components [5], and in the experiments described here only the central component was considered. It corresponds to a Λ system based on the ($F=3$, $m_F=0$) and ($F=4$, $m_F=0$) states and is shifted by only 24 Hz by the applied magnetic field. Broadening due to field inhomogeneities across the cell is negligible.

To take an experimental spectrum the pump laser is

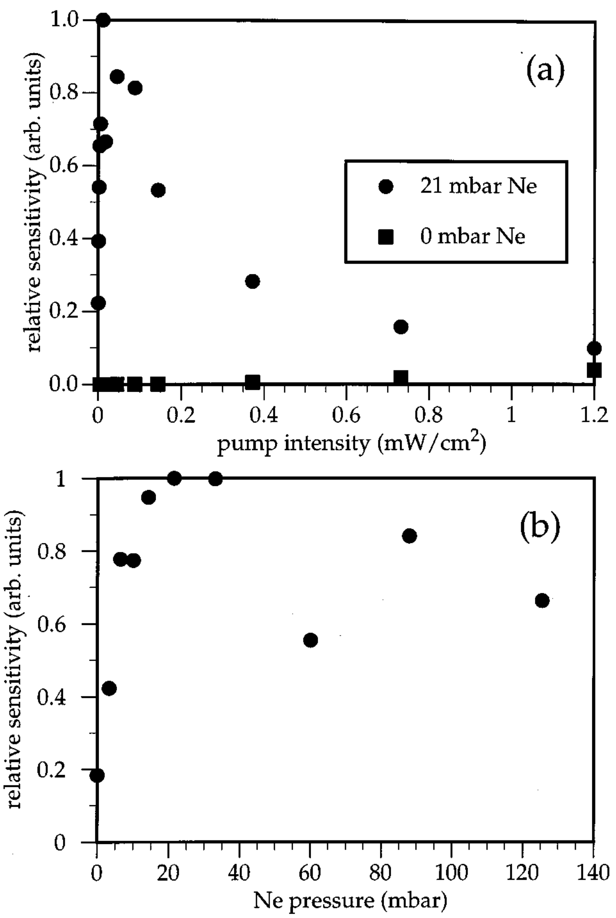

FIG. 4. Estimated sensitivity (resonance height)$^{1/2}$/(resonance width) of a hypothetical precision magnetometer. (a) Intensity dependence for 0 and 21 mbar Ne, and (b) pressure dependence of the maximum sensitivity. The sensitivity can be improved by more than an order of magnitude when a buffer gas is used.

locked to the 3-4 crossover transition of the saturated absorption spectrum in an auxiliary cesium vapor cell, and the 20-MHz synthesizer frequency is stepped so that the probe laser is swept through the resonance. In addition, the rf frequency is modulated with 1-kHz frequency and a modulation index of 1. The transmission signal is demodulated by a digital dual-phase lock-in amplifier. In all experiments the pump laser intensity is a factor of 1.5 higher than the probe intensity.

Figure 2 shows examples of line shapes for the coherent dark resonance at low laser intensities without buffer gas [Fig. 2(a)] and with 87-mbar neon pressure [Fig. 2(b)]. The solid line in Figs. 2(a) and 2(b) is a fit to the experimental data points that not only takes the frequency modulation into account [14] but also an additional amplitude modulation that occurs because the frequency modulation is accomplished through modulation of the probe laser current. This explains the small asymmetries in the line shape. The pedestal around the narrow peak in Fig. 2(b) is due to residual line frequency interference within the servo loop for the laser phase lock, and we are currently modifying the setup in order to reduce the effect. Nevertheless, the main signal component has a full width at half maximum (FWHM) of only

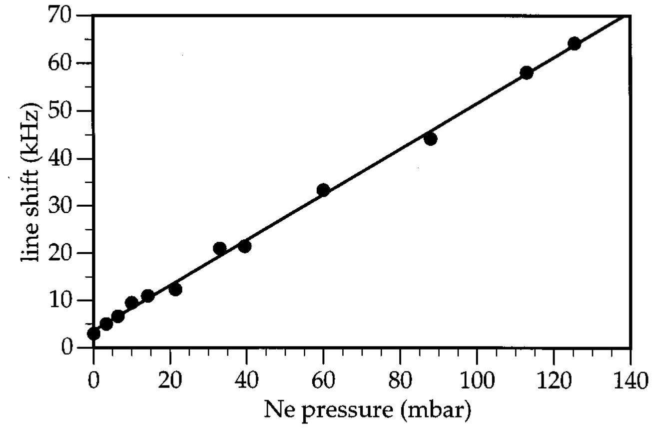

FIG. 5. Shift of the resonance line with neon pressure. The solid line is a linear fit to the experimental points.

42 Hz and thus impressingly shows the narrowing effect of a buffer gas. Our linewidths are now comparable to widths obtained in earlier experiments where the $(F=3, m_F=0) \mapsto (F=4, m_F=0)$ transition was directly driven by microwave excitation [8].

The linewidth shows a linear increase with laser intensity [Fig. 2(c)], as is to be expected from the theory of a three-level system [2]. Without buffer gas the minimum linewidth for low intensity is about 9 kHz, which is mainly due to time-of-flight broadening for our beam diameter of 1 cm.

The linewidth reduction induced by a buffer gas obviously depends on its pressure. From the solution of a diffusion equation [15] one obtains a contribution of time-of-flight broadening that varies inversely proportional to pressure, whereas collisions with buffer-gas atoms increase the linewidth linearly with pressure. Using the relaxation cross sections measured in rf excitation experiments [16] the total resonance linewidth due to time-of-flight broadening and buffer-gas collisions can be calculated as $\mathrm{FWHM}=(1314\ \mathrm{Hz\ mbar})/p+(0.404\ \mathrm{Hz/mbar})p$ and is shown as the solid line in Fig. 3. The dots in that figure are experimental results we obtained for laser intensities far below the optical two-level saturation intensity of $I_0=2.2\ \mathrm{mW/cm^2}$. It is clearly evident that there is quantitative agreement between the theoretical prediction from rf spectroscopy and our optical experiment.

Such narrow linewidths in principle allow precision measurements; e.g., the detection of small Zeeman shifts of the resonance frequency due to an external magnetic field. Accuracies of 10^{-16} T have been predicted theoretically for the case of shot-noise limited detection [6]. In this limit the sensitivity is proportional to the steepness of the slope of the resonance line [i.e., proportional to the quantity (line height)/(linewidth)], while the noise increases with the square root of optical power on the detector. An increase in laser intensity initially increases the height of the signal, but eventually power broadening dominates and reduces the steepness. A simple estimate of the gain in sensitivity with a buffer gas can therefore be obtained through a comparison of the quantity [(line height)/(linewidth)]/(line height)$^{1/2}$ (Fig. 4). For 21-mbar Ne pressure there is an optimum intensity of 88 μW/cm^2, where the sensitivity is higher by a factor of

25 compared to the case without buffer gas [Fig. 4(a)]. 21 mbar is about the optimum neon pressure, as is evident from Fig. 4(b), where the maximum sensitivity is plotted as a function of pressure.

While collisions with buffer-gas atoms do not significantly broaden the coherent dark resonance line, they do, however, shift it. In Fig. 5 the position of the resonance line is plotted as a function of buffer-gas pressure for 21 °C and probe laser intensity of 4 μW/cm^2. A numerical linear fit gives a slope of (480 ± 10) Hz/mbar in the limit of zero laser intensity where the uncertainty is that of the fit. There is reasonable agreement with a value of (452 ± 20) Hz/mbar determined from the pressure shift of the 0-0 microwave resonance [17].

Our results indicate that the use of buffer-gas filled cells can increase the usefulness of coherent dark resonances for precision spectroscopy and measurement, just as it improved rf measurements in the past [8]. The use of optical radiation to excite and probe a ground-state coherence has several well-known advantages. Optical detectors are much more efficient than available detectors in the rf regime. The radiation can be transported to the atoms using optical fibers [9], and also the geometry does not have to be as large as with direct rf excitation because of the much smaller wavelength. Furthermore, the resonance is dark, i.e., it has very low absorption, so that in principle long interaction lengths are feasible. This is also one reason why it is predicted [3] that the detection of magnetic resonances via coherent dark resonances could be superior to traditional rf probe experiments.

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