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Coherent dark states of rubidium 87 in a buffer gas using pulsed laser light

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Abstract. The coherent dark resonance between the hyperfine levels $F = 1$, $m_F = 0$ and $F = 2$, $m_F = 0$ of the rubidium ground state has been observed experimentally with the light of a pulsed mode-locked diode laser operating at the D1 transition frequency. The resonance occurs whenever the pulse repetition frequency matches an integer fraction of the rubidium 87 ground state hyperfine splitting of 6.8 GHz. Spectra have been taken by varying the pulse repetition frequency. Using cells with argon as a buffer gas a linewidth as narrow as 149 Hz was obtained. The rubidium ground state decoherence cross section $\sigma_2 = 1.1 \times 10^{-18}$ cm^2 for collisions with xenon atoms has been measured for the first time with this method using a pure isotope rubidium vapor cell and xenon as a buffer gas.

PACS. 42.50.Gy Effects of atomic coherence on propagation, absorption, and amplification of light – 42.62.Fi Laser spectroscopy

1 Introduction

In the last few years there has been an increasing interest in experiments on coherent population trapping (CPT) or "dark resonances" because of the variety of interesting effects connected with CPT like "electromagnetically induced transparency", "lasing without inversion" and precision experiments using the ground state hyperfine structure of alkali atoms. The topic has been reviewed recently by Arimondo [1]. Recently Brandt *et al.* [2] measured a linewidth below 50 Hz by means of a cw-laser CPT experiment using cesium vapor and two diode lasers which were phase locked to each other. CPT experiments can also be performed with a pulsed laser where the pulse repetition frequency matches an integer fraction of the ground state splitting. This method was used first in a time-resolved pump-and-probe experiment by Mlynek *et al.* [3] and later in an experiment where the pulse repetition frequency of a pulsed diode laser was scanned over an integer fraction of the rubidium ground state splitting while the fluorescence light was observed [4,5]. Using helium and argon as a buffer gas, linewidths down to 52 Hz were reported. Our work shows that CPT experiments similar to the one in [4] can be used to study the interaction of rubidium 87 atoms with heavy noble gas atoms serving as a buffer gas. We find that the linewidth of the dark resonance strongly depends on the kind and on the pressure of the buffer gas. The results are consistent with conventional radio-frequency measurements [6]. For the first time using our method we measure the ^{87}Rb ground state decoherence cross section for collisions with xenon atoms and the pressure shift of the (0–0) transition caused by xenon as a buffer gas. Further we could confirm the extreme linewidth reduction with an appropriate choice of the kind and pressure of the buffer gas, as is shown in [2,4,5].

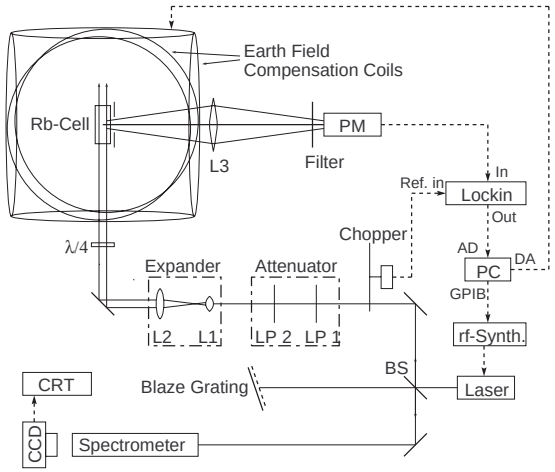

Fig. 1. Experimental setup. L=Lens, LP=Linear polariser, BS=Beam splitter, PM=photomultiplier.

2 Experiment

The experimental setup is shown in Figure 1. We use a SONY SLD 201 laser diode with a "home made"

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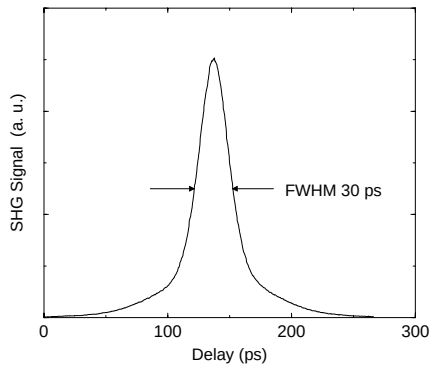

Fig. 2. Autocorrelation of typical pulses.

anti-reflection coating on one side in an external cavity with a mode separation of 525.7 MHz. Pulsed mode-locked operation can be achieved by modulating the laser current at this frequency. The light pulses have a duration of 15 ps (FWHM) and a spectral width of approximately 100 GHz (FWHM) which is about 85 GHz bigger than the Fourier limit for such pulses. The pulse width and shape can be monitored by means of autocorrelation measurements using a SHG correlator. The autocorrelation of typical pulses as used in the experiment is shown in Figure 2. In order to monitor the laser operation on-line we observe the laser spectrum with a grating spectrometer and a CCD camera. Typically the information obtained by the spectrum is sufficient to optimize the mode locking while autocorrelation measurements are only necessary for reference purposes. The pulse repetition frequency can be scanned over several tens of kilohertz by varying the modulation frequency of the laser diode without affecting the laser spectrum and pulse width. The laser is operating at 795 nm, the D1 transition of rubidium. Transitions out of both hyperfine ground state levels are induced because of the large spectral width of the laser.

In order to minimize power broadening and to increase the mean interaction time of the rubidium atoms with the laser light the chopped beam passes an attenuator and a beam expander. Then the light is circularly polarized and the fluorescence light from the rubidium vapor cell is detected by a photomultiplier with lock-in detection. The lock-in bandwidth is 1/3 Hz at a chopper frequency of 22.8 Hz. The pulse repetition frequency is scanned over an integer fraction of the ground state level splitting. In our case the mean pulse repetition frequency of 525.7 MHz corresponds to 1/13 of the hyperfine splitting of 6.835 GHz.

The cell is in the center of three mutually perpendicular Helmholtz coil pairs. Two of these pairs are used to compensate static transversal fields. The field component parallel to the beam can be adjusted by means of the third Helmholtz coil. This field causes a Zeeman splitting of the ground state hyperfine levels. Typically we used fields up to 100 μT. Fields of this strength are sufficient to Zeeman shift the magnetic sublevels with $m_F \neq 0$ far enough that only the (0–0) transition, *i.e.* the transition between the levels $F = 1$, $m_F = 0$ and $F = 2$, $m_F = 0$ of $5^2S_{1/2}$ is

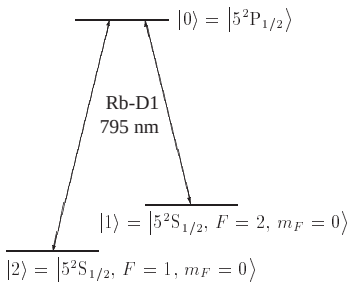

Fig. 3. Rubidium 87 as a Λ-system.

in resonance with the the pulse repetition frequency. The levels with $m_F = 0$ experience only very small shifts in the applied magnetic field and therefore line broadening due to inhomogeneities of this field is negligible.

3 Results

We consider the interaction between the atoms and a train of short pulses resonant to the rubidium D1 transition. For our purposes we describe the ^{87}Rb atom as a three level Λ-system (Fig. 3). It can be shown theoretically [3] that a coherence between the ground state levels builds up when the pulse repetition frequency matches a subharmonic of the level splitting frequency. When the pulse repetition frequency ν_p is scanned over the m-th fraction of the hyperfine splitting frequency ν_{12} the fluorescence signal is given approximately by the Lorentzian [7]

$$S(m\nu_p) \sim \frac{c_{12}/\Gamma_{12}}{1 + \left(\dfrac{2\pi(\nu_{12} - m\nu_p)}{\Gamma_{12}}\right)^2} \tag{1}$$

where Γ_{12} is the ground state coherence relaxation rate. So the linewidth only depends on Γ_{12} that reads [8]

$$\Gamma_{12} = AD_0\frac{p_0}{p} + N_0\bar{v}_r\sigma_2\frac{p}{p_0} \tag{2}$$

in the presence of a buffer gas at pressure p. A is a factor that only depends on the geometry of the cell, D_0 is the diffusion constant of the rubidium in the buffer gas, $p_0 = 1013$ mbar is the reference pressure, N_0 is Loschmidt's constant, $\bar{v}_r$ is the relative velocity of the rubidium and the buffer gas atoms and σ_2 is the decoherence cross section, *i.e.* the cross section for collisions producing a loss of coherence in the ensemble. The first term of (2) describes the relaxation caused by the diffusion of the rubidium atoms through the buffer gas to the cell walls and the second term describes decoherence by collisions between rubidium atoms and buffer gas atoms.

We use cylindrical cells with a length of 5 cm and a diameter of 2 cm. Using 33 mbar of argon as a buffer gas in a cell containing rubidium in the natural isotopic mixture we could measure the dark resonance with linewidths (FWHM) as narrow as 149 Hz. Experimental data and a

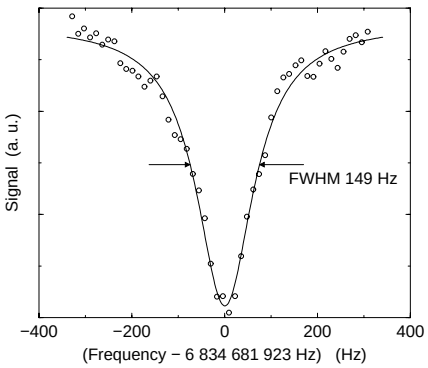

Fig. 4. Typical (0–0) dark resonance in 33 mbar argon as a buffer gas at 34 °C. The longitudinal magnetic field is $B = 84.1\ \mu$T. The solid line is a fitted Lorentzian.

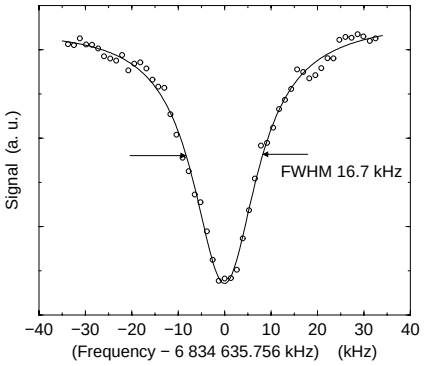

Fig. 5. Typical (0–0) dark resonance in 53 mbar xenon as a buffer gas at 21 °C. The longitudinal magnetic field is $B = 50\ \mu$T. The solid line is a fitted Lorentzian.

fitted Lorentzian are shown in Figure 4. The linewidth increases with laser intensity which is consistent with the results in [2,5]. From (1) and (2) it can be expected that the smallest achievable linewidth using our cells is about 30 Hz. The larger linewidth we get experimentally is due to power broadening, the rather poor beam quality of our laser and possibly to jitter in the output of our rf-synthesizer. Broadening due to inhomogeneities of the magnetic field can be neglected because we only consider coherences between levels with $m_F = 0$. Using 53.3 mbar of xenon as a buffer gas in a pure isotope rubidium cell allowed us to measure the rubidium ground state decoherence cross section [9] for collisions with xenon atoms and the pressure shift [6] of the (0–0) line caused mainly by Van der Waals interaction with the xenon atoms. The experiment yielded a linewidth of 16.7 kHz at 21 °C (Fig. 5). Since the diffusion of rubidium atoms through the xenon buffer gas is very slow only the second term in (2) contributes considerably to the linewidth. Therefore the result can be considered as a direct measurement of the decoherence cross section σ_2. We get $\sigma_2 = 1.1 \times 10^{-18}$ cm^2 which is about one order of magnitude greater than the spin disorientation cross section $\sigma_1 = 1.3 \times 10^{-19}$ cm^2 given by Franz [10]. This is in accordance with experimental results for these cross sections obtained for ^{85}Rb in He, Ne and Ar [9] where the σ_2 cross sections turned out to be up to three orders of magnitudes greater than the respective σ_1 values for ^{87}Rb given in [10]. Since we own only one sealed off resonance cell containing enriched ^{87}Rb and Xe buffer gas we cannot measure the pressure shift by varying the xenon pressure. This shift was obtained by correcting the resonance frequency of Figure 5 for the applied magnetic field and subtracting the hyperfine splitting of the free ^{87}Rb atom [8]. This results in a pressure shift caused by xenon of (-885 ± 128) Hz/mbar. The uncertainty of this value is mainly due to the uncertainty of the exact value of the buffer gas pressure at the time of sealing. The shift compares well with measurements of the frequency shift of ^{87}Rb in Ar and Kr [11] and the frequency shift of ^{133}Cs in Ar, Kr and Xe [12]. We checked the accuracy of our frequency reference (HP-5340A) by calculating the ground state hyperfine splitting of ^{87}Rb from measurements of dark resonances with the buffer gas argon (*e.g.* Fig. 4) taking into account the well-known pressure shift of −51 Hz/Torr [11] for argon and the shift due to the magnetic field and obtained agreement with the value for the hyperfine splitting taken from literature [8] within 200 Hz. Thus the frequency measurement contributes only a very small part to the error of the xenon pressure shift.

Our results show that experiments on dark resonances using pulsed laser light can be applied to study the interaction of alkali atoms with buffer gas atoms. The advantage of this kind of experiment compared to conventional rf measurements and to common CPT experiments using cw-lasers is the relatively simple experimental setup. In our experiment both broadening and shift of the (0–0) dark resonance line of rubidium in xenon buffer gas could me measured. The linewidth yielded the rubidium ground state decoherence cross section for collisions with xenon atoms.

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