Light shift of coherent population trapping resonances

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ABSTRACT

Although the frequency position of the non-coupled state in coherent population trapping does not depend on light intensities this is not true for the experimentally observed dark resonance minimum. For precision applications this shift constitutes a major systematic effect. We have measured the light shift in an atomic cesium vapor as a function of buffer gas pressure. The experimental data for the unbuffered vapor can be reproduced quantitatively with a simple model taking into account the atomic velocity distribution and the excited state hyperfine structure. The buffered vapor requires a more complicated model.

Keywords: Coherent population trapping, dark resonance, light shift, AC Stark shift

1. INTRODUCTION

When in an atomic three-level system with two low-lying and long-lived levels (a so-called Λ-system) the difference frequency of two driving laser fields matches the splitting of the two lower levels (Fig. 1a), $\omega_b - \omega_a = \omega_{ab}$, the atomic population can become trapped in a coherent superposition state of the two lower levels that is no longer coupled to the light fields (coherent population trapping, CPT).[1,2] Since one-photon absorption and resonance fluorescence in the trapping state are strongly reduced the resonance is called a "dark resonance". The two-photon resonance can be very narrow,[3] with the current record standing at 42 Hz.[4] The use of such resonances for the construction of atomic radio frequency standards[5] or applications for precision measurements, for instance in magnetometry,[6] has been suggested, and first steps in this direction have been taken.[7] It is an essential requirement for such precision applications to understand the influence of experimental conditions on the position and shape of the resonance under investigation.

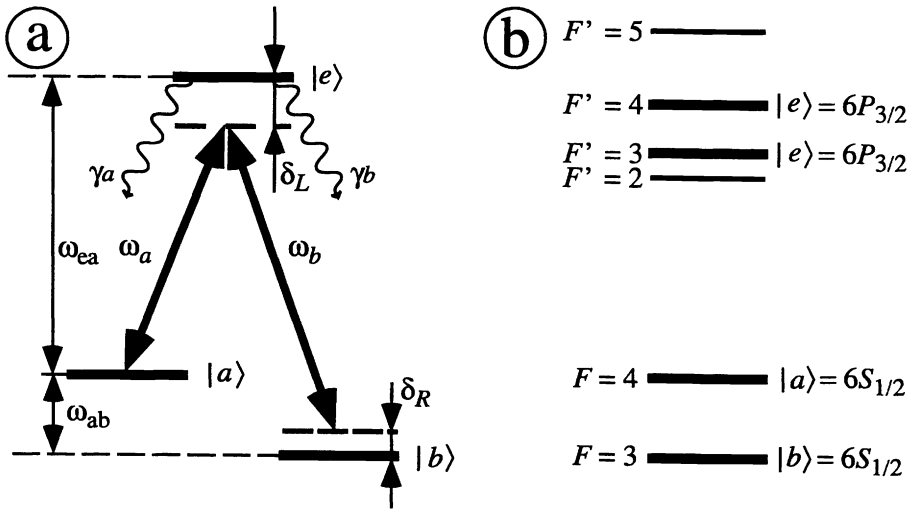

Figure 1. a) Important parameters in a three-level Λ-system. ω_a, ω_b denote the frequencies of the two laser fields. b) Relevant cesium atomic levels for D_2 line excitation.

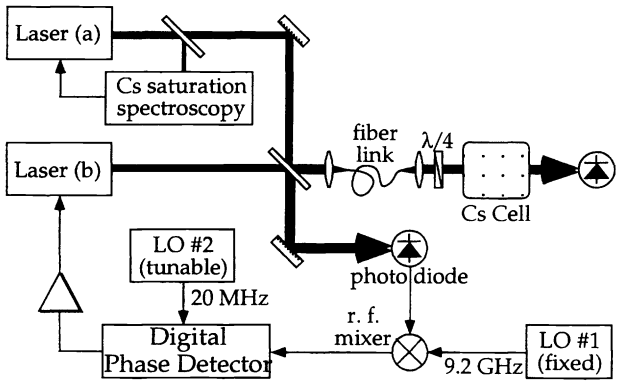

Figure 2. Experimental setup for the sensitive detection of intensity-dependent frequency shifts of the coherent population trapping resonance in cesium vapor.

While the position of the non-coupled state corresponds to zero Raman detuning $\delta_R = \omega_b - \omega_a - \omega_{ab} = 0$ and does not depend on the intensity of the driving fields the minimum of the absorption coefficient does not necessarily coincide with this frequency,[8,2] and in experiments a shift of the center of the resonance is readily observed, as was discussed, for instance, for an atomic beam.[9] Here we report measurements of the light shift of the two-photon resonance coupling two hyperfine ground state levels of the cesium atoms in a thermal vapor as a function of buffer gas pressure. The experimental results for an unbuffered vapor are compared with theoretical calculations that take into account the velocity distribution of the atoms as well as the influence of additional atomic levels on the two-photon resonance.

2. EXPERIMENTAL SETUP

The experimental setup[4] consists of two grating-tuned diode lasers tuned to the cesium D_2 resonance line at 852 nm wavelength that are electronically phase-locked to each other with a difference frequency of 9.2 GHz, corresponding to the cesium ground state hyperfine splitting (Fig. 2). The phase-locked loop ensures that the difference frequency and phase of the two laser fields are precisely determined by the stable and tunable 9.2 GHz frequency reference. Both laser beams are sent through the same stretch of single-mode fiber in order to reduce the residual Doppler shifts due to non-parallel beams. Behind the fiber the beam is collimated to a diameter of 7 mm before it passes through a cesium vapor cell of length 2 cm and is detected on a photo diode.

Laser frequency ω_a is stabilized to the Doppler-broadened absorption profile in an auxiliary cesium vapor cell, at a frequency position near the cross-over transition from the $F = 4$ ground state to the excited $F' = 3$ and $F' = 4$ levels. Therefore the one-photon detuning $\delta_L = \omega_a - \omega_{ea}$ (see Fig. 1) remains constant. Laser frequency ω_b (tuned to the resonances starting from the $F = 3$ ground state) is scanned through the dark resonance by detuning the 9.2 GHz oscillator, i.e., δ_R. A longitudinal magnetic flux density of 24 μT was applied to the cell so that the dark resonance splits into seven components[10] because of the Zeeman structure of the cesium ground state. Only the dark resonance corresponding to the coupling of $|6S_{1/2}, F = 3, m_F = 0\rangle$ and $|6S_{1/2}, F = 4, m_F = 0\rangle$ was considered. This resonance is shifted by the magnetic field by only 50 Hz from the position at zero field.

The intensity of the laser starting from the $F = 4$ ground state was varied from 0.6 μW/cm^2 to 1.2 mW/cm^2 (about the saturation intensity of the optical one-photon transitions) while the intensity of the other laser was always a factor of 2/3 lower. These intensities correspond to the most interesting range for precision applications because for higher intensities the sensitivity is reduced by power broadening of the resonance.[4] The 9.2 GHz reference frequency was modulated with 1 kHz frequency and 1 kHz amplitude and the photo diode signal demodulated with a dual-phase lock-in amplifier. The center of the measured frequency modulation (FM) line shape can be found from a fit of a calculated curve (assuming a Lorentzian absorption coefficient[11]) with a statistical uncertainty of typically a few hertz (Fig. 3).

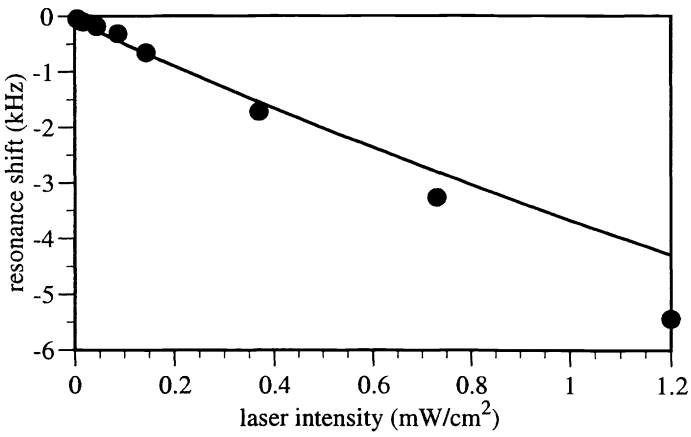

Figure 3. Comparison of the theoretical dependence (solid line) of the dark resonance shift on the laser intensity of the stronger laser with the experimental data points.

3. THEORETICAL MODEL

The shape of the transmission curve for a low-density vapor as a function of Raman detuning δ_R is basically given by the excited state population $n_e(\delta_L, \delta_R)$ which can be calculated from the steady-state solutions of the density matrix equations for the three-level system.[12] With the relaxation rates of the system γ_a and γ_b for the excited state and Γ_a and Γ_b for the dephasing of the corresponding coherences and the one-photon Rabi frequencies $g_a = d_{ae}E_a/\hbar$ and $g_b = d_{be}E_b/\hbar$ (d_{ie} are the dipole moments on the respective transitions) one obtains an analytical solution for n_e in the rotation wave approximation

$$n_e = \frac{A\delta_R^2 + B}{C\delta_R^4 + 2C\delta_L\delta_R^3 + (D + E\delta_L^2)\delta_R^2 + F\delta_L\delta_R + G\delta_L^2 + H} \tag{1}$$

with

$$\begin{aligned}
A &= 8\Gamma_a\Gamma_b g_a^2 g_b^2 \\
B &= 2\Gamma_{ab} g_a^2 g_b^2 (\Gamma_a g_a^2 + \Gamma_b g_b^2 + 4\Gamma_{ab}\Gamma_a\Gamma_b) \\
C &= 16 g_a^2 \gamma_b \Gamma_a \\
D &= -8\gamma_b\Gamma_a g^4 + 16\Gamma_a\Gamma_b(\gamma_b\Gamma_b g_a^2 + \gamma_a\Gamma_a g_b^2) + 16\Gamma_{ab}^2\gamma_b\Gamma_a g_a^2 + 4(\Gamma_{ab}\gamma_b - \Gamma_a(\gamma_a + \gamma_b - 6\Gamma_b))g_a^2 g_b^2 \\
E &= 16(\gamma_b\Gamma_a g_a^2 + \gamma_a\Gamma_b g_b^2) \\
F &= 4[2\gamma_b\Gamma_a g_a^4 - 2\gamma_a\Gamma_b g_b^4 - 8\Gamma_{ab}^2\gamma_b\Gamma_a g_a^2 + g_a^2 g_b^2((\gamma_a + \gamma_b)(\Gamma_a - \Gamma_b) - \Gamma_{ab}(\gamma_a + 3\gamma_b))] \\
G &= 8\Gamma_{ab}(2\Gamma_{ab}(\gamma_b\Gamma_a g_a^2 + \gamma_a\Gamma_b g_b^2) + (\gamma_a + \gamma_b) g_a^2 g_b^2) \\
H &= (\Gamma_a g_a^2 + \Gamma_b g_b^2 + 4\Gamma_{ab}\Gamma_a\Gamma_b)(\gamma_b g_a^4 + \gamma_a g_b^4 + 4\Gamma_{ab}(\gamma_b\Gamma_b g_a^2 + \gamma_a\Gamma_a g_b^2) + (\gamma_a + \gamma_b + 6\Gamma_{ab}) g_a^2 g_b^2) \\
&= \gamma_b\Gamma_a g_a^6 + \gamma_a\Gamma_b g_b^6 + 8\Gamma_{ab}\Gamma_a\Gamma_b(\gamma_b g_a^4 + \gamma_a g_b^4) + 16\Gamma_{ab}^2\Gamma_a\Gamma_b(\gamma_b\Gamma_b g_a^2 + \gamma_a\Gamma_a g_b^2) \\
&\quad + g_a^2 g_b^2[(\gamma_b\Gamma_b + \Gamma_a(\gamma_a + \gamma_b + 6\Gamma_{ab}))g_a^2 + (\gamma_a\Gamma_a + \Gamma_b(\gamma_a + \gamma_b + 6\Gamma_{ab}))g_b^2 \\
&\quad + 4\Gamma_{ab}\Gamma_a\Gamma_b(\gamma_a + \gamma_b + 6\Gamma_{ab}) + 4\Gamma_{ab}(\gamma_a\Gamma_a^2 + \gamma_b\Gamma_b^2)].
\end{aligned}$$

The *only* approximations made are that there is no population transfer between the two ground states directly ($\gamma_{ab} = 0$, otherwise the expression gets much more complicated) and that each laser only interacts with one of the lower states.

For $\delta_L \neq 0$ the line shape is asymmetric with respect to Raman detuning and the minimum can be shifted from $\delta_R = 0$. We should note here that this shift is not simply given by the difference of the AC Stark shifts of the two optical transitions, as one might intuitively assume. Instead it is essential to include the ground state coherence into the calculation.

We have not been able to find a completely general formula like Eq. (1), valid for arbitrary intensities and relaxation rates, in an extensive body of literature on three-level systems.[2,9,13] However, many approximate solutions were derived.[2] For example, for the case of a weak field on the a–e transition an expression for the susceptibility was given in[14]; starting from there we find the position of the local minimum of their line shape at $\delta_R = \delta_L \Gamma_{ab}/\gamma_a$, which is just the limit obtained from our Eq. (1) for $g_a^2 \ll g_b^2$.

In a vapor cell the situation is complicated by the distribution $f_D(\delta)$ of atomic velocities v, causing a Doppler shift of the detunings $\delta_L \to \delta_L + \delta$, $\delta = kv$, for the copropagating light fields. In the resulting line shape $L_D(\delta_L, \delta_R)$

$$L_D(\delta_L, \delta_R) \propto \int f_D(\delta) n_e(\delta_L + \delta, \delta_R) d\delta \tag{2}$$

the line shape asymmetry found for a stationary atom (Eq. (1)) gets washed out. A small asymmetry remains because the shape of L_D depends on the offset δ_L of n_e with respect to the center of the Doppler distribution: the curves $n_e(\delta_L)$ effectively sit on a sloping background.

The experimental situation for cesium vapor is further complicated by the hyperfine structure in the cesium $6P_{3/2}$ state (Fig. 1b). While the ground states $|a\rangle$ and $|b\rangle$ of Fig. 1a can be identified with the cesium $6S_{1/2}, F = 3, 4$ states, the two excited states $6P_{3/2}, F' = 3, 4$ can both play the role of state $|e\rangle$. This is taken into account by an incoherent sum of two three-level resonance lines:

$$n''_{D,\mathrm{tot}}(\delta_L, \delta_R) = n''_D(\delta_{L3}, \delta_R) + n''_D(\delta_{L4}, \delta_R), \tag{3}$$

where δ_{L3} and δ_{L4} correspond to the laser detunings δ_L with respect to the upper levels $F' = 3$ and $F' = 4$ of an atom at rest. It has been shown previously that such an incoherent sum adequately describes the situation in a vapor cell without buffer gas.[15,10] In our case $\delta_{L3} - \delta_{L4} = \omega_{43}$, where ω_{43} is the hyperfine splitting of the upper levels $F' = 4$ and $F' = 3$. The $6P_{3/2}, F' = 2, 5$ levels lead to off-resonant one-photon excitation that produces additional dephasing of the ground state coherence. For the calculation a total dephasing rate of $\Gamma_{ab} = 2\pi \cdot 10\,\mathrm{kHz}$ was used, corresponding to the minimum linewidth expected for transit time broadening at our laser beam diameter.

Special care has to be taken to map the relaxation rates and Rabi frequencies of the theoretical three-level model to the two three-level subsystems of the cesium multilevel system. This is explained in more detail in Ref.[12] The result of the numerical Doppler integration of $L_{D,\mathrm{tot}}(\delta_L, \delta_R)$ (solid line in Fig. 3) shows good agreement with the experimental data even without the use of any free parameters. Given the well-known experimental difficulties when trying to determine an accurate value of intensity it is surprizing that there is only such a small discrepancy. For instance, no effort was made to account for the transverse Gaussian intensity profile of the laser beam. An additional effect is the dependence of the total shift on optical detuning δ_L. In the experiment δ_L was held constant only to within a few megahertz, causing a small uncertainty in the slope of the curve of a few percent.

4. BUFFERED CESIUM VAPOR

In the precision applications mentioned above one clearly wants as narrow a linewidth as possible. For instance, by adding neon as a buffer gas linewidths of less than 50 Hz are obtained for neon pressures of 40–90 mbar.[4] Since the frequent collisions with buffer gas atoms drastically broaden the optical transition to several 100 MHz the two upper levels $F' = 3, 4$ not only become less and less resolved but also is the residual asymmetry after Doppler integration reduced.

Using a resealable cesium vapor cell we have measured the position of the dark resonance as a function of intensity for a series of neon pressures. For each pressure a curve similar to the experimental points in Fig. 3 was obtained. For simplicity, a straight line was fitted to the data points and its slope, the light shift rate, plotted versus neon pressure (Fig. 4).

As expected, the light shift rate strongly decreases with increasing buffer gas pressure. In the interesting pressure range around 60 mbar the rate is already one order of magnitude lower than for an unbuffered vapor—good news for any precision application of coherent dark states.

Currently we are in the process of adapting our theoretical model for the case of a buffered vapor. Since now the two upper levels are no longer fully resolved one can no longer add one three-level system solution each for each of the two excited states, as before. Instead, the corresponding solution for a four-level system consisting of two ground and two excited states in the presence of two resonant laser fields has to be derived.

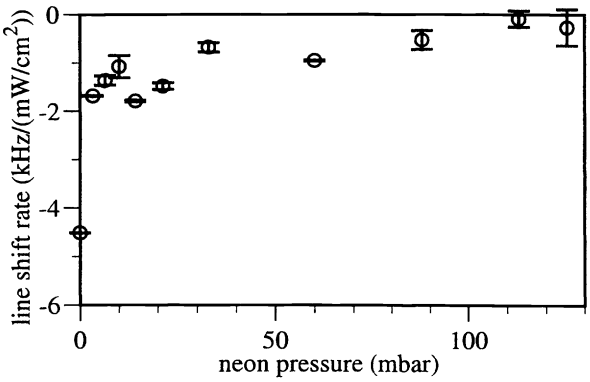

Figure 4. Light shift rate of the dark resonance in cesium vapor as a function of neon buffer gas pressure. In the interesting range around 60 mbar the rate is already strongly reduced.

5. CONCLUSION

The dependence of the position of the coherent population trapping resonance on laser intensities is quite substantial. For envisioned precision applications this effect has to be taken into account, and we have shown that it can be modeled rather accurately in an unbuffered vapor. Using the theoretical model one can predict a reduction of the light shift by more than a factor of 50 when the lasers are tuned 85 MHz to the blue side of the transitions to the excited $F' = 4$ state. Furthermore, the light shift is automatically strongly reduced in a cell filled with an additional buffer gas.

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