Dark-line atomic resonances in submillimeter structures

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We present measurements of dark-line resonances excited in cesium atoms confined in submillimeter cells with a buffer gas. The width and contrast of the resonances were measured for cell lengths as low as 100 μm. The measured atomic Q factors are reduced in small cells because of frequent collisions of atoms with the cell walls. However, the contrast of coherent population trapping resonances measured in the small cells is similar in magnitude to that obtained in centimeter-sized cells, but substantially more laser intensity is needed to excite the resonance fully when increased buffer-gas pressure is used. The effect of the higher intensity on the linewidth is reduced because the intensity broadening rate decreases with buffer-gas pressure.

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Compact frequency references have become increasingly important for military[1] and civilian[2] applications in recent years. For many applications in the field of navigation (e.g., global positioning systems) and communication systems, inexpensive miniature references with frequency stabilities in the 10^{-11} range at integration times approaching one day would be of great advantage. It has been predicted[3] that atomic clocks based on coherent population trapping (CPT) spectroscopy in millimeter-sized vapor cells might fulfill these requirements. It is well known that two phase-stable laser fields in resonance with a resonance line of the alkali atoms, and with a difference frequency equal to the hyperfine splitting, induce coherence between the two hyperfine ground states.[4] This gives rise to a narrow resonance in the transmission spectrum with a width determined by the lifetime of the induced coherence. The first CPT-based clock was demonstrated by Thomas *et al.*[5] using a beam of sodium atoms. When used as a frequency reference, the signal-to-noise ratio as well as the Q factor of the resonance determines the frequency stability. Here we present measurements of the resonance linewidth and amplitude as a function of vapor cell size and we discuss the implications of the measurements for the short-term stability of a small-cell frequency reference.

At the heart of the experiment is a stainless-steel vacuum cube filled with Cs vapor and a buffer gas. The cube has a window on one side, and a mirror inside the cube is parallel to this window and can be moved by an actuator, thus changing the distance between the interior surface of the window and the mirror (see Fig. 1). A bichromatic light field was produced by frequency modulating (at 4.6 GHz) the injection current of a single-mode vertical-cavity surface-emitting laser (VCSEL) at 852 nm, transferring approximately 60% of the optical power into the two first-order sidebands. The circularly polarized 3-mm-diameter beam was sent through the window of the cube, reflected off the mirror, and the returned power was detected on a photodiode. A longitudinal magnetic field of a few microteslas was applied to separate the Zeeman levels. Furthermore, the cube was placed into a magnetic and thermal shield to reduce stray magnetic fields and to stabilize the temperature at 75 °C. The laser was locked to the center of the optical Doppler- and pressure-broadened absorption line of the atoms in the same small cell to minimize the noise.[6] By scanning the rf frequency (4.6 GHz) and measuring the transmitted power, we recorded the 0–0 CPT resonance (Fig. 1). The resonance is frequency shifted from that of the unperturbed atom by collisions with buffer-gas atoms. The shift is proportional to the buffer-gas pressure, and its temperature coefficient, which affects the long-term stability of the frequency reference, can be controlled by use of buffer-gas mixtures. A Lorentzian line (solid line) was fitted to the data and the width and amplitude of the resonance were extracted. Since the resonance amplitude scales linearly with Cs density for optically thin media and therefore depends on the cell temperature, it was normalized to the Doppler absorption background. The contrast defined in this manner is a measure of the fraction of atoms in the coherent state and is related to the signal that determines the frequency stability. The optimum amplitude can be adjusted by tuning the atom density by varying the cell temperature.

As the size of the vapor cell is reduced, the lifetime of the ground-state coherence becomes shorter because of collisions of the atoms with the cell walls. A buffer

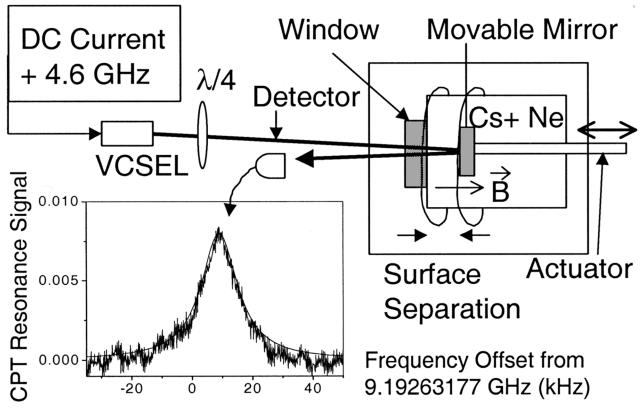

Fig. 1. Experimental setup. The resonance signal is normalized to the Doppler background absorption, measured at 1.1 mW/cm^2 and a gap size of 580 μm.

gas with a small cross section for collisions that destroy the hyperfine coherence is added to the alkali vapor for a substantial reduction of the CPT linewidth. At high enough buffer-gas pressures the alkali atoms follow a diffusive motion through the buffer gas. In this regime the linewidth ν_{CPT} scales with cell dimension L as $1/L^2$ because of wall collisions.[7] At higher buffer-gas pressures, the broadening that results from collisions between buffer-gas and alkali atoms comes into effect; this broadening is proportional to buffer-gas pressure p.[7] The total linewidth is therefore given by

$$\nu_{\mathrm{CPT}} \propto \xi_p p + \frac{\xi_L}{pL^2}, \tag{1}$$

where ξ_p and ξ_L are constants. A minimum linewidth can be reached for a given cell size at some optimum pressure. This optimum pressure increases as the cell size becomes smaller.

Although in the experiment we varied the cell size in one dimension only by changing the separation between the surfaces, no substantial changes would be expected if the atoms were confined in all three dimensions. This is because in the diffusive regime, i.e., when the mean-free path between two collisions is much smaller than the surface separation, the smallest cell dimension dominates the broadening of the CPT resonance. Furthermore, interference effects between incident and reflected laser beams can be ignored for path lengths much smaller than the wavelength of the hyperfine transition (3.2 cm).[8]

The CPT linewidth was measured as a function of laser intensity for various surface separations and Ne pressures. Because the CPT resonance is subject to optical power broadening, the linewidth was extrapolated to zero laser intensity. The extrapolated results are plotted in Fig. 2 as a function of surface separation for three different Ne pressures. The experimental values (squares) obtained from the Lorentzian fits (as shown in Fig. 1) agree fairly well with Eq. (1) (lines). Figure 2 clearly shows that the resonance width increases with smaller distance for constant Ne pressure. As the Ne pressure is increased, the linewidth is reduced, until the broadening that is due to buffer-gas collisions starts to dominate. In the submillmeter regime this collisional broadening dominates only at Ne pressures above 100 kPa. Nevertheless, CPT resonances were measured for surface separations below 100 μm at lower Ne pressures and the linewidths agree with the theoretical predictions in this regime.

Measurements of the CPT contrast as a function of cell size were also carried out. For an optically thin vapor of ideal three-level atoms with no ground-state decay, the contrast is expected to be 100%.[4] However, for Cs assuming that the atoms are equally distributed over all ground-state Zeeman levels, only one eighth of the atoms contribute to the CPT resonance, reducing the maximum possible contrast to near 10%. Furthermore, for excitation on the D_2 line, the excited states that couple to only one ground state serve as loss channels, reducing the contrast even further.[9,10] As a result, the contrast of resonances, measured in centimeter-sized cells under similar conditions has been near 1%.[11] We note that optical pumping can be used to enhance the atomic population in selected Zeeman states,[12,13] thus increasing the resonance contrast. Because of the added complexity of a setup that requires additional laser beams, the implementation of a small clock based on one of these techniques could be more difficult. Figure 3 shows the CPT resonance contrast as a function of gap size (between mirror and window) for three different Ne pressures (filled symbols). All the spectra were measured at the same temperature of 75 °C and total laser intensity of 350 μW/cm^2. It can be seen that for 4-kPa Ne, the maximum contrast of 0.4% is only slightly lower than the 1% value observed in centimeter-sized cells. For small surface separations as well as for large ones, the contrast drops drastically. In spite of the fact that our system does not fulfill the

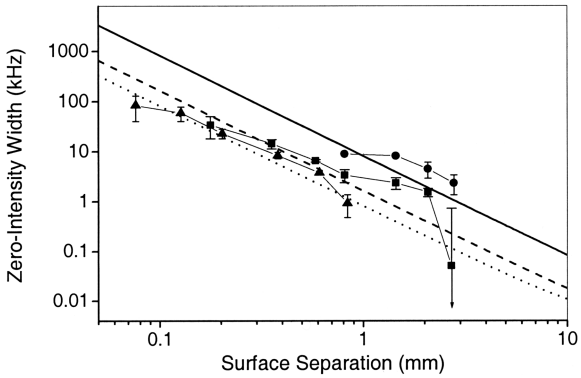

Fig. 2. Full width at half-maximum (as a function of surface separation) of the CPT resonances extrapolated to zero light intensity for three different Ne pressures: 0.8 (circles), 4 (squares), and 8 kPa (triangles). The lines are the theoretical predictions taking into account diffusion and buffer gas collisions for the same pressures: 0.8 (solid), 4 (dashed), and 8 kPa (dotted).

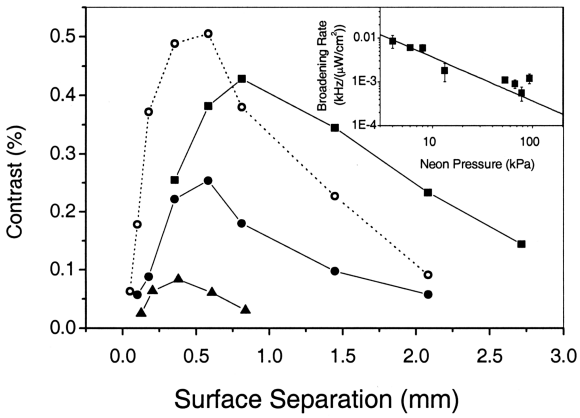

Fig. 3. Contrast of the Lorentzian fits to the CPT resonances as a function of surface separation at 350 μW/cm^2 for three different Ne pressures: 4 (squares), 6 (circles), and 8 kPa (triangles). The open circles correspond to a higher light intensity of 1.4 mW/cm^2 at 6 kPa. Inset: linear broadening rate of the CPT resonance with laser intensity at a 380-μm surface separation as a function of Ne pressure.

assumption of a symmetric three-level system, this behavior is in qualitative agreement with the theory for CPT resonances in optically thick vapors recently published by Godone *et al.*[14] The measured dependence stems from different absorption characteristics of the light on and off CPT resonance. Off resonance the transmitted light drops exponentially with length according to the Lambert–Beers law of attenuation.[15] On resonance, the absorption depends on the coherence between the ground states, giving rise to a different dependence on length. For the longer path lengths, the transmission decreases faster on resonance than off resonance as the cell gets optically dense. For small sizes, the wall collisions diminish the lifetime of the coherence, reducing the contrast. The situation is further complicated by possible effects of radiation trapping arising as a result of our use of Ne as the buffer gas, which effects are not included in Ref. 14. Since the decay rates of the transmission depend on the absorption coefficient of the vapor, i.e., the density of Cs atoms, the cell temperature can be chosen such that the maximum contrast occurs for a given surface separation. Furthermore, spin–exchange collisions between Cs atoms contribute negligibly to the linewidth for our typical parameters but become important at more elevated temperatures.

Increasing the Ne pressure reduces the overall contrast and causes the turnover of the contrast as a function of surface separation to occur at smaller separations. Despite narrowing the linewidth of the microwave CPT transition, collisions of Ne atoms also homogeneously broaden the optical transition to the $P_{3/2}$ state, resulting in less efficient pumping into the dark state. A higher light intensity is therefore required to reach the same contrast (open circles in Fig. 3). We believe it might be possible to reach contrasts similar to those observed in centimeter-sized cells in our submillimeter cells, but the limited intensity available in this experiment did not appear to saturate fully the dark-state population.

In the preceding paragraphs we have seen that, with increased buffer-gas pressure, higher intensities are desirable. In an optically thin medium the CPT linewidth broadens proportional to the laser intensity.[4] Therefore the broadening rate of the resonance with intensity for different buffer-gas pressures is of some importance. The broadening rate of the CPT resonance with laser intensity was measured for a series of Ne pressures at 380-μm surface separation (squares in Fig. 3 inset). Because of the reduced transition probabilities, it should decrease inversely proportional to the pressure-broadened linewidth of the optical transition. The solid line in the Fig. 3 inset is a constant divided by a homogeneous optical linewidth. This means that the higher laser intensity required to excite high-contrast resonances at higher buffer-gas pressure has a weaker effect on the linewidth because of the reduced broadening rate.

The fact that our measurements agree with the theoretical predictions fairly well to gap sizes below 100 μm is encouraging for the development of miniature CPT-based atomic clocks. We have found that smaller cell sizes require higher buffer-gas pressures to optimize the CPT linewidth. Since higher pressure reduced the CPT resonance contrast, a higher laser intensity is also needed to compensate for it. Simultaneously, the broadening rate with intensity decreases for higher buffer-gas pressure. For clocks based on CPT, this raises the important but somewhat complex question of exactly how line broadening (reduction of the Q factor), caused by (a) collisions of the alkali atoms with cell walls, (b) collisions of alkali atoms with buffer-gas atoms, and (c) intensity, affects the optimal clock stability.

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