Abstract: The amplitude of Λ resonance in alkali atoms is limited by perturbing cycling transitions in the case of D2 line or by existence of additional trapping states in the case of D1 line. We propose to eliminate these extra trapping states by using two counter-propagating bichromatic fields of orthogonal circular polarizations. The experiment is in accordance with the theoretical proposal. The result refers to small-size cells and is important for applications in miniaturized atomic clocks.



The dependence of the CPT signal amplitude on the position of the mirror with respect to the vapor cell

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High-contrast dark resonance in $\sigma_+ - \sigma_-$ optical field

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1. Introduction

As communications facilities develop and informationcontent transfer grows, compact frequency references are becoming increasingly important for a variety of military and civilian applications [1,2]. For many of these applications, a clock with small device volume, low power consumption, low market cost and long-term stability is required. Widely available quartz-crystal oscillators have a small size, low power consumption and good short-term stability. The main disadvantage of this kind of oscillator is poor long-term stability. Efforts to decrease the long-term instability of quartz crystals inevitably make them larger and more expensive. The most stable commercial quartz oscillators have a fractional frequency instability of about 10^{-10} per day, a volume of ~ 10^2 cm³, and an energy consumption that exceeds 3 W. These last two parameters are greater for atomic clocks, which on the other hand have good accuracy and long-term stability. In recent years, a number of companies have carried out systematic efforts to decrease the volume of atomic clocks and to make them less expensive and simpler. Commercial devices based on a vapor cell have reached volumes about 100 cm³ [3].

The effect of coherent population trapping (CPT) in alkali-metal atoms has opened a new path for the practical implementation of atomic frequency standards. Using this approach, it becomes possible to develop a simple, miniaturized device, due to absence of a microwave cavity. Efforts to design and build CPT-based chip-scale atomic clocks were undertaken in a few scientific groups [3–5].

For application to atomics clocks, it is important to have both large amplitude and small width of the CPT resonance. In this paper, we show that the contrast and amplitude of the dark resonance can be significantly increased

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in a miniature buffer-gas cell by the use of a $\sigma_+ - \sigma_-$ field configuration and excitation of the D₁ line of alkali-metal atoms. First experiments carried out with Cs vapor confirm the theoretical predictions and show that the contrast of the resonance in the CPT-based chip-scale atomic clock can be greatly increased with only minor design modifications of the device.

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2. Qualitative theoretical consideration

As in conventional vapor-cell frequency references, the CPT atomic clock is based on magnetically-insensitive Zeeman substates of the hyperfine-split ground state of alkali-metal atoms. In the usual CPT excitation geometry, the coherence between these states is induced by a unidirectional, circularly polarized, bichromatic light field

$$\mathbf{E}(t) = (E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t})\mathbf{e}_{\pm 1} + c.c.$$
(1)

in the presence of a static magnetic field directed along the direction of light propagation. Here $\mathbf{e}_1 = \mp (\mathbf{e}_x \pm i \mathbf{e}_y)/\sqrt{2}$ are the spherical orts, corresponding to the right (e_{+1}) or left (e_{-1}) circular polarizations, $E_{1,2}$ are scalar amplitudes of the field components. In a typical approach, the bichromatic field consists of the two co-propagating running waves, with the spectroscopic signal being the total absorption versus frequency difference of laser fields $(\omega_1 - \omega_2)$. When this difference is tuned across the groundstate hyperfine splitting Δ_{hfs} , a narrow dip is observed in the absorption (the so-called "dark resonance"). The width of the dark resonance is governed by the ground-state relaxation rate, that is by collisions and either by the time of free flight of atoms though the laser beam or by the time of their diffusion to the walls if a cell is filled with a buffer gas (in the weak-field limit). The use of vapor cells containing buffer gas in addition to alkali-metal vapor has narrowed the resonance linewidths to less than 50 Hz [6].

For practical application of the dark resonance in atomic clocks both the amplitude and the amplitude-towidth ratio of the output signal should be as large as possible. This can be done by a proper choice of the excitation scheme. For example in the case of the D2 line of alkali-metal atoms the contrast of resonance does not typically exceed a few percent (we define contrast as the ratio (B - A)/B, where A is the absorption in the exact twophoton resonance $\omega_1 - \omega_2 = \Delta_{hfs}$ and B is the absorption far off two-photon resonance but still on the single-photon resonance) [7]. This limit is related to: (1) the destructive interference of coherences that appear in the two available lambda schemes distinguished by upper states (F' = 3, 4); (2) the additional mechanism of relaxation of the groundstate ("0-0") coherence due to absorption on cycling transitions $(F = 4 \rightarrow F' = 5, F = 3 \rightarrow F' = 2$ for Cs). The hyperfine splitting of the P state does not help here since small cells imply high pressure of the buffer gas, which broadens the optical transitions, preventing the excited-state hyperfine splitting from being resolved. The use of the D1 line



Figure 1 The scheme of transitions in ¹³³Cs induced by bichromatic laser light resonant to D1 line. The bold solid lines indicate the transitions from the working states $|F_1, m_1 = 0\rangle$ and $|F_2, m_2 = 0\rangle$ (shown by black circles). The coherence between these states are formed by the two-photon Raman transitions on the frequency Δ_{hfs} . The trap state $|pump\rangle = |F_2, m = F_2\rangle$ is indicated by an asterisk

allows for a significant increase of the resonance amplitude [8]. The corresponding physical reasons are as follows. In the case of D1-line excitation, there exists a dark (i.e., non-absorbing) state $|dark\rangle$ at the exact two-photon resonance $(\omega_1 - \omega_2) = \Delta_{hfs}$ even in the presence of strong collisional broadening of the optical transition (since two coherences distinguished by upper F'' = 3, 4 states interfere constructively and there are no cycling transitions). This state is a coherent superposition of the states $|F_1, m_1 = 0\rangle$ and $|F_2, m_2 = 0\rangle$ (Fig. 1). In the case of σ_+ circular polarization (\mathbf{e}_{+1}) it takes the form

$$|dark\rangle = |F_1, m_1 = 0\rangle + (E_1/E_2)|F_2, m_2 = 0\rangle.$$
 (2)

Due to optical pumping, atoms are accumulated in this dark state, resulting in a decrease of the absorption. The use of the D1 line, however, limits the amplitude of the resonance in another way. It stems from the existence of another nonabsorbing trap state $|pump\rangle = |F_2, m_2 = F_2\rangle$ (Fig. 1), which is insensitive to the Raman detuning $\delta_R = (\omega_1 - \omega_2) - \Delta_{hfs}$. Atoms pumped into this state by circularly polarized light prevent the contrast of the dark resonance from exceeding 50%.

This obstacle can be avoided in small cells by the use of a $\sigma_+ - \sigma_-$ configuration of the bichromatic field, which

Figure 2 A scheme of the light-induced transitions from the working states $|F_1, m_1 = 0\rangle$ and $|F_2, m_2 = 0\rangle$ in the $\sigma_+ - \sigma_-$ configuration of bichromatic field. The numbers on lines give the relative amplitudes of the dipole-moment matrix elements for the D1 line of ¹³³Cs

is formed by counter-propagating waves with orthogonal circular polarizations:

$$\mathbf{E}(z,t) = \left(E_1^{(+)}\mathbf{e}_+e^{ik_1z} + E_1^{(-)}\mathbf{e}_-e^{-ik_1z}\right)e^{-i\omega_1t} + (3) + \left(E_2^{(+)}\mathbf{e}_+e^{ik_2z} + E_2^{(-)}\mathbf{e}_-e^{-ik_2z}\right)e^{-i\omega_2t} + c.c.$$

Here $k_j = 2\pi/\lambda_j$ is the wave vector of the field component with the frequency ω_j (j = 1, 2), z is the coordinate along the direction of propagation of the beam, and z = 0 is arbitrarily defined. In this case the trap state $|pump\rangle$ is absent since at least one of the two counterpropagating fields couples to each Zeeman sublevel. The dark resonance in the $\sigma_+ - \sigma_-$ configuration has been studied in [9], where a spatial variation of the resonance amplitude was observed. This dependence was due to the spatial variation of the difference between the relative phases of the frequency components $2(k_1 - k_2)z$. Thus, the effect of the spatial modulation of the resonance amplitude with the period $\pi/(k_1 - k_2)$ is caused by the difference of the wavelengths λ_1 and λ_2 for the two frequency components. For example, for the ¹³³Cs atom the period $\pi/(k_1 - k_2)$ is 16.3 mm.

Indeed, in the case of exact two-photon resonance the dark state for the σ_+ polarized wave, propagating in the positive direction, has the form:

$$|dark^{(+)}\rangle = |F_1, m_1=0\rangle + \beta e^{i(k_1-k_2)z} |F_2, m_2=0\rangle,$$
 (4)

while for the counter-propagating σ_{-} polarized wave, the dark state differs from (4) in the phase of the second term:

$$|dark^{(-)}\rangle = |F_1, m_1 = 0\rangle - \beta e^{-i(k_1 - k_2)z} |F_2, m_2 = 0\rangle,$$
 (5)

where $\beta = E_1^{(+)}/E_2^{(+)} = E_1^{(-)}/E_2^{(-)}$. The formulae (4) and (5) are obtained with account for the relative amplitudes of the dipole-moment matrix elements shown in Fig. 2. It follows from equations (4),(5) that the dark states $|dark^{(+)}\rangle$ and $|dark^{(-)}\rangle$ coincide (constructive interference of the two-photon transitions) in the planes $z = z_{max}$ where

$$2(k_1 - k_2)z_{max} = (2n+1)\pi, \quad (n = 0, 1, 2, \ldots).$$
(6)

Consequently, the dark state for the total field (3) exists in these planes and the amplitude of resonance attains there a maximum. In all other planes $|dark^{(+)}\rangle \neq |dark^{(-)}\rangle$ and destructive interference occurs, so that the pure dark state for the field (3) does not exist, which leads to a decrease of the amplitude. The minimum of the resonance amplitude is achieved in the planes where $2(k_1 - k_2)z_{min} = 2\pi n$.

In the case of large cells, where $L \ge \pi/(k_1 - k_2)$, the resonance amplitude in the $\sigma_+ - \sigma_-$ field cannot be significantly increased, because of the spatial averaging of the field over the cell. However, in small cells, where $L \ll \pi/(k_1 - k_2)$, the phase relations are approximately constant throughout the entire volume of the cell. Therefore, if such a cell is placed in the vicinity of the plane $z = z_{max}$ a significant enhancement (of up to 100%) of the contrast of the dark resonance is expected, since the dark state for the total field (3) does exist at $(\omega_1 - \omega_2) = \Delta_{hfs}$, and there is no trap state at $(\omega_1 - \omega_2) \neq \Delta_{hfs}$. In addition, the required counter-propagating field can be created by reflecting the initial field off a mirror after it has passed once through the cell and a waveplate.

The qualitative considerations presented here can be confirmed by theoretical calculations based on the application of the methods developed in [9,10] to the case under investigation. In particular, it can be shown that the amplitude-to-width ratio can be increased by a factor of several, as compared to the case of co-propagating, circularly-polarized waves. Details of the calculations and discussions of their results are published in [11].

3. Experimental setup and results

In order to confirm the theoretical predictions, an experiment was carried out using a ¹³³Cs vapor cell having a diameter of 2 mm and a length of 4 mm; the length was much less than the wavelength of microwave transition, $2\pi/(k_1 - k_2) \approx 32.6$ mm. A neon buffer gas (20 kPa) was added to the cell to reduce the microwave transition linewidth. The cell was placed inside 3-D Helmholtz coils to compensate the transverse magnetic field and to provide a longitudinal magnetic field of 6×10^{-5} T. The radiation





Figure 3 (online color at www.lphys.org) Experimental setup

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Figure 4 The dependence of the CPT signal amplitude on the position of the mirror with respect to the vapor cell

of an extended cavity diode laser tuned to 894.3 nm (D1 line) was modulated at 4.6 GHz with an electro-optical modulator, circularly polarized, and sent through the cell (Fig. 3).

The diameter of the laser beam in the cell was 0.5 mm, and the maximum power was 30 μ W, which was limited mainly by the losses in the modulator and instabilities of the laser at higher currents. After passing through the cell, the laser beam was reflected back on itself with a mirror fixed on a micrometer stage. Either one or two quarter-wave plates were inserted between the cell and the retro-reflecting mirror, depending on the polarization state to be investigated. When one quarter-wave plate was present, σ^+ polarization in the incident beam was transformed to σ^- polarization in the returning beam and the amplitude of the CPT resonance was measured as a function of the mirror displacement (Fig 4). In agreement with theory, the signal changed with a period of 16.3 mm and its maximum value was greater than a single pass signal. When two quarter-wave plates were present, the polarization state of the return beam could be varied by changing the relative orientation of the waveplates' optical axes. In this case, the distance to the mirror was kept constant and equal to $(2n+1)\lambda_{HFS}/4$, and the dependence of CPT signal amplitude on the polarization state of the return beam was measured. The dependence is shown in Fig. 5. The CPT signal attains a maximum for the $\sigma_+ - \sigma_-$ configuration (constructive interference of coherences produced by the counter-propagating beams) and goes to a minimum for the $\sigma_+ - \sigma_+$ configuration (destructive interference), as predicted by theory. The horizontal level at 1.5 mV shows the CPT-signal amplitude when the return beam is not present. It can be seen that the amplitude of the CPT signal taken with $\sigma_+ - \sigma_-$ configuration is almost 35% larger than the corresponding amplitude taken with the σ_+ single-pass scheme. The contrast attains value of 7%.

diction not only qualitatively. The calculations made for parameters (the optical width, the ground-state relaxation

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Figure 5 CPT signal amplitude plotted as a function of the polarization of the reflected beam. The gray horizontal line shows the signal amplitude when the reflected light is blocked

rate, and the laser intensity) corresponding to the experiment show the increase of the resonance amplitude by a factor of 1.4 with respect to the σ_+ polarized running wave of the same intensity. For further increase of the resonance amplitude it is necessary to increase the laser power. We plan to perform similar experiments with more powerful lasers.

4. Conclusion

A scheme to enhance the amplitude of a CPT signal was proposed and confirmed. We have shown that in small cells, the amplitude of a nonlinear dark resonance excited on the D1 line with a counter-propagating, bichromatic $\sigma_+ - \sigma_-$ field can be improved over the usual scheme of co-propagating, circularly polarized laser beams. The results may be important for applications such as highly miniaturized atomic frequency references or magnetometers. Initial experiments confirm the theoretical predictions.

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