Narrowing of a Dark Resonance in a Cell with Anti-Relaxation Wall Coating

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***Abstract*—This paper is dedicated to the investigation of CPT resonance narrowing in a coated cell without buffer gas. We found that there are two different mechanisms of this narrowing: the laser-induced narrowing for lasers with narrow spectral width and the Dicke narrowing for wide laser spectrum width and gas cell length less than the microwave length.**

I. INTRODUCTION

Secondary quantum frequency standards have numerous applications in various fields such as the navigation systems (GALILEO, GLONASS, GPS), telecommunications, et.c. The space-borne frequency standards should have specific frequency stabilities but also reduced size, weight and consumption. The gas-cell standards based on Coherent Population Trapping (CPT) effect is one of the promising options for such standards (see [1] and references therein). One of its advantage is the absence of the microwave cavity. It allows to reduce the volume and mass of the standard.

Let us consider the principle of operation of CPT-based frequency standard. The main element of its quantum discriminator is the gas cell with alkali metal vapors irradiated by the two-frequency laser radiation with frequencies ν_1 and ν_2. These components are tuned in resonance with optical transitions from different hyperfine components of the ground state on the excited state. When we scan the two-photon detuning $\delta_R = \nu_1 - \nu_2 - \omega_{hfs}$ near the zero, a narrow peak (frequently called the *dark resonance*) in transmission spectrum is observed. Here ω_{hfs} is the ground state hyperfine splitting frequency. The nature of this peak is the existence (for $\delta_R = 0$) of the *dark state* — the coherent superposition of ground state sublevels that doesn't interact with laser radiation.

It should be noted that usually, in addition to the active atoms, the cell also contains a buffer gas with several mbar pressure. Because of the frequent collisions with the buffer-gas atoms, the alkali-metal atoms can only slowly diffuse in a gas cell. On the one hand it leads to the decreasing of the wall collision (or the time-of flight broadening if the laser beam diameter is less than the gas cell). On the other hand it eliminates the residual Doppler broadening of the CPT resonance line width. The last effect named the Dicke narrowing [2] in buffered gas cell has been investigated both experimentally [3] and theoretically [4].

Another option is a cell without buffer gas but with anti-relaxation wall coating. It should be noted that this method helps to deliver from the broadening and temperature–depended shift of clock transition due to collisions between buffer and active atoms. The wall coating can remain effective for at least several decades after the cell is fabricated [5]. On the one hand, the anti-relaxation wall coating decreases the depolarization in a wall collisions. On the other hand it saves the atoms in coherent state in a volume of a gas cell and if this volume is quite small (less than microwave line length λ) we can expect the Dicke narrowing in a coated cell.

The Dicke narrowing for double radio optical resonance in coated cell with optical pumping by the isotopic-filtered light was shown experimentally [6] and explained theoretically in density matrix formalism [7]. It occurs that the anti-relaxation wall coating can provide the Dicke narrowing if the gas cell length is less than the microwave length. In this paper we investigate the CPT resonance narrowing in a coated cell.

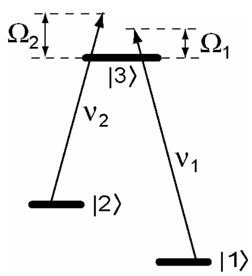

Figure 1. Λ-system excitation scheme.

II. MATHEMATICAL MODEL

A. Set of equations

In this paper we will consider a cell filled with the vapor of an active element (describing the atoms of this element in three-level Λ-system approximation, see Fig.1). Let us write down a set of quantum kinetic equations for the density matrix $\hat{\rho}(\vec{r},\vec{\upsilon},t)$ of the active atoms allowing for all the main interactions of these atoms in adiabatic approximation [8]:

$$\begin{aligned}
&\frac{\partial\rho_{11}}{\partial t}+\vec{\upsilon}\cdot\nabla\rho_{11}=-\left[\left(G\frac{V_1^2}{\gamma'}+\Gamma\right)\rho_{11}+\right.\\
&\left.+i\frac{V_1V_2}{\gamma'}F\left(\rho_{21}-\rho_{12}\right)-\left(G\frac{V_2^2}{\gamma'}+\Gamma\right)\rho_{22}\right],\\
&\frac{\partial\rho_{22}}{\partial t}+\vec{\upsilon}\cdot\nabla\rho_{22}=-\left[\left(G\frac{V_2^2}{\gamma'}+\Gamma\right)\rho_{22}-\right.\\
&\left.-i\frac{V_1V_2}{\gamma'}F\left(\rho_{21}-\rho_{12}\right)-\left(G\frac{V_1^2}{\gamma'}+\Gamma\right)\rho_{11}\right],\\
&\frac{\partial\rho_{12}}{\partial t}+\vec{\upsilon}\cdot\nabla\rho_{12}=-\frac{V_1V_2}{\gamma'}\left[G(\rho_{11}+\rho_{22})-iF(\rho_{11}-\rho_{22})\right]+\\
&+\rho_{12}\left[-\Gamma-G\frac{V_1^2+V_2^2}{\gamma'}+i\left(\delta_R-F\frac{V_1^2-V_2^2}{\gamma'}+\vec{q}\cdot\vec{\upsilon}\right)\right],
\end{aligned}\tag{1}$$

where Γ is the relaxation rate in a ground state, γ' is the relaxation rate of optical coherence, V_1 and V_2 are the Rabi frequencies of light induced transitions $|1\rangle\rightarrow|3\rangle$ and $|2\rangle\rightarrow|3\rangle$ respectively, $\vec{q}=\vec{k}_1-\vec{k}_2$ is the difference between wave vectors of laser field components (we suppose that these components are co-propagated), $\vec{\upsilon}$ is the atomic velocity. Coefficients *F* and *G* are the following:

$$F=\frac{\gamma'\left(\Omega_L-\vec{k}\cdot\vec{\upsilon}\right)}{\gamma'^2+\left(\Omega_L-\vec{k}\cdot\vec{\upsilon}\right)^2},\qquad G=\frac{\gamma'^2}{\gamma'^2+\left(\Omega_L-\vec{k}\cdot\vec{\upsilon}\right)^2},\tag{2}$$

$\Omega_L=(\Omega_1+\Omega_2)/2$ is the laser detuning, $\vec{k}_1\approx\vec{k}_2\approx\vec{k}$.

B. Boundary conditions

Coating of various types are frequently used to prevent the depolarization effect of the cell walls [7]. Some of these coating, such as long-chain paraffin coating, adsorb active atoms extremely slightly and lead to a slight dispersion of the phase of the wave function in a collision with the wall. Accordingly, a very large number of collisions of active atoms with the wall is required for a complete relaxation of the angular momentum. An idealization of this situation is expressed by boundary conditions of a specular-coherent reflection:

$$\rho_{ij}\left(\vec{r},\vec{\upsilon}\right)\Big|_{\substack{\vec{r}\in S\\ \upsilon_n>0}}=\rho_{ij}\left(\vec{r},\vec{\upsilon}\right)\Big|_{\substack{\vec{r}\in S\\ \upsilon_n<0}}.\tag{3}$$

Another situation apparently prevails in the case of silicone coating, which do not result in a significant adsorption which do introduce a significant dispersion of the phase shift. In this case, ignoring the slight relaxation of population at the wall, we use the boundary conditions of specular-incoherent reflection:

$$\begin{aligned}
&\rho_{ii}\left(\vec{r},\vec{\upsilon}\right)\Big|_{\substack{\vec{r}\in S\\ \upsilon_n>0}}=\rho_{ii}\left(\vec{r},\vec{\upsilon}\right)\Big|_{\substack{\vec{r}\in S\\ \upsilon_n<0}},\\
&\rho_{12}\left(\vec{r},\vec{\upsilon}\right)\Big|_{\substack{\vec{r}\in S\\ \upsilon_n<0}}=0.
\end{aligned}\tag{4}$$

Finally, in the case of uncoated gas cell every collision lead to the total depolarization of atom and therefore this situation can be described by the full quenching condition:

$$\begin{aligned}
&\rho_{11}\left(\vec{r},\vec{\upsilon}\right)\Big|_{\substack{\vec{r}\in S\\ \upsilon_n<0}}=\rho_{22}\left(\vec{r},\vec{\upsilon}\right)\Big|_{\substack{\vec{r}\in S\\ \upsilon_n<0}},\\
&\rho_{12}\left(\vec{r},\vec{\upsilon}\right)\Big|_{\substack{\vec{r}\in S\\ \upsilon_n<0}}=0.
\end{aligned}\tag{5}$$

We make our calculations for the parameters of Λ-system close to similar parameters of ^{87}Rb atom: $\omega_{21}=\omega_{hfs}=6.8$ $GHz\approx4.3\cdot10^{10}\ s^{-1}$, $\lambda=2\pi c/\omega_{hfs}=4.4\,cm$, and $\omega_{31}\approx\omega_{32}=$ $=\omega_{opt}=2.4\cdot10^{15}\ s^{-1}$. Value of ground state relaxation rate was supposed to be $\Gamma=100\ s^{-1}$. The value of the relaxation rate γ' of the optical coherence is determined by the spectral laser line width Γ_L and the spontaneous relaxation rate $\gamma_{sp}=3.5\cdot10^7 s^{-1}$ [8]:

$$\gamma'=\Gamma_L/2+\gamma_{sp}/2.\tag{6}$$

We consider the one-dimensional gas cell with length a and use the following normalization condition:

$$\rho_{11}\left(z,\upsilon\right)+\rho_{22}\left(z,\upsilon\right)=M\left(\upsilon\right)/a.\tag{7}$$

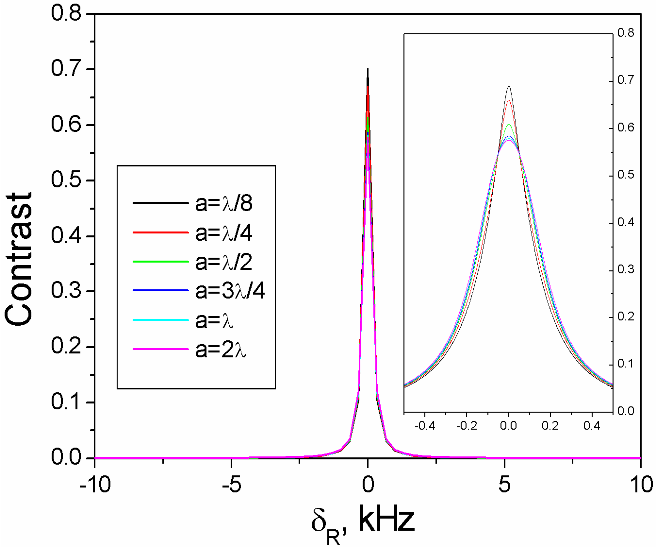

Figure 2. CPT signal line shape for specular-coherent reflection. $\gamma' = 1.8\cdot 10^{7}\ s^{-1}$, $V_1 = V_2 = 10^{5}\ s^{-1}$

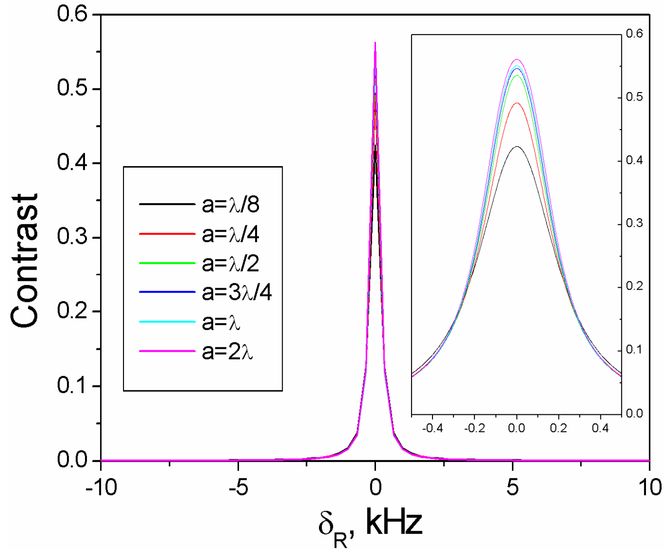

Figure 3. CPT signal line shape for specular-incoherent reflection. $\gamma' = 1.8\cdot 10^{7}\ s^{-1}$, $V_1 = V_2 = 10^{5}\ s^{-1}$

Condition (7) corresponds to the case when the translational degree of freedom of active atoms are in thermal equilibrium. Here $M(\upsilon)$ is the Maxwell distribution function:

$$M(\upsilon) = \frac{1}{\upsilon_T\sqrt{\pi}}\exp\left(-\frac{\upsilon^2}{\upsilon_T^2}\right). \qquad (8)$$

We suppose that $\upsilon_T = 2.5\cdot 10^{4}\ cm/s$ ($T = 50\ ^{\circ}C$). In this case $k\upsilon_T = \omega_{opt}\upsilon_T / c = 315\,MHz = 2\cdot 10^{9}\ s^{-1}$, $q\upsilon_T = \omega_{hfs}\upsilon_T / c = = 5.7\,kHz = 3.56\cdot 10^{4}\ s^{-1}$. Also we suppose that the laser detuning $\Omega_L = 0$.

Light absorption in a gas cell is proportional to the population ρ_{33} in the excited state

$$\rho_{33} = \frac{2G}{\gamma_{sp}\gamma'}\left[|V_1|^2\rho_{11} + \left(V_1V_2^*\right)\rho_{12} + \left(V_2V_1^*\right)\rho_{21} + |V_2|^2\rho_{22}\right] \qquad (9)$$

integrated via the atomic velocity and the cell length:

$$\bar{\rho}_{33} = \iint \rho_{33}(z,\upsilon)\,dz\,d\upsilon. \qquad (10)$$

In this work we are interested in the CPT absorption contrast that we determine by the following way:

$$C(\delta_R) = \frac{\bar{\rho}_{33}{}^{NR} - \bar{\rho}_{33}(\delta_R)}{\bar{\rho}_{33}{}^{NR}}. \qquad (11)$$

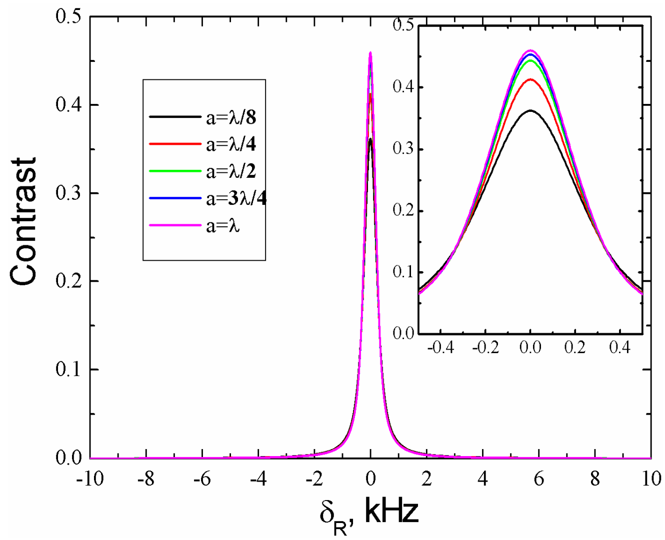

Figure 4. CPT signal line shape for uncoated cell. $\gamma' = 1.8\cdot 10^{7}\ s^{-1}$, $V_1 = V_2 = 10^{5}\ s^{-1}$

where $\bar{\rho}_{33}{}^{NR}$ is the value of $\bar{\rho}_{33}$ out of CPT resonance but under condition of one-photon optical resonance.

III. Results and Discussion

We make our calculations for different types of boundary conditions, for different values of the relaxation rate γ' of the optical coherence and for different length of a gas cell. Results are presented on Figures 2 — 7.

Let us consider the obtained results. Figures 2 — 4 shows that if the relaxation rate γ' of optical coherence is much less than $k\upsilon_T$ the cell size and the type of boundary conditions affects on the CPT line shape weakly. However the dark resonance line width is much less than qv_T. Therefore the line narrowing in this case isn't connected with Dicke narrowing. Here we can observe another mechanism ~~what is~~ called laser-induced narrowing. Laser-induced narrowing was investigated theoretically for infinite cell [9-12] and experimentally for cells without buffer gas [13,14]. The nature of this mechanism is the following: laser light interacts only with the slow atoms, i.e. the atoms whose velocities v satisfy the condition $v \le \gamma'/k$, that is about two orders of magnitude less than v_T. Therefore Doppler broadening and wall collisions play an insignificant role for such atoms and the CPT line occurs to be narrowed.

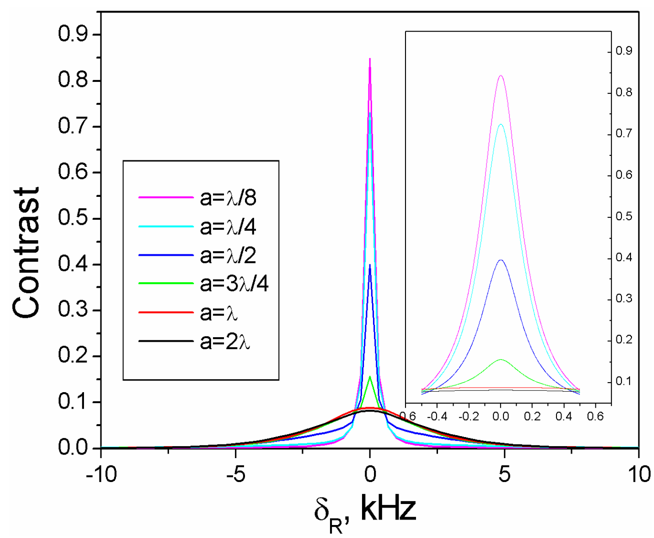

Figure 5. CPT signal line shape for specular-coherent reflection. $\gamma' = 2\cdot 10^9\ s^{-1}$, $V_1 = V_2 = 10^6\ s^{-1}$

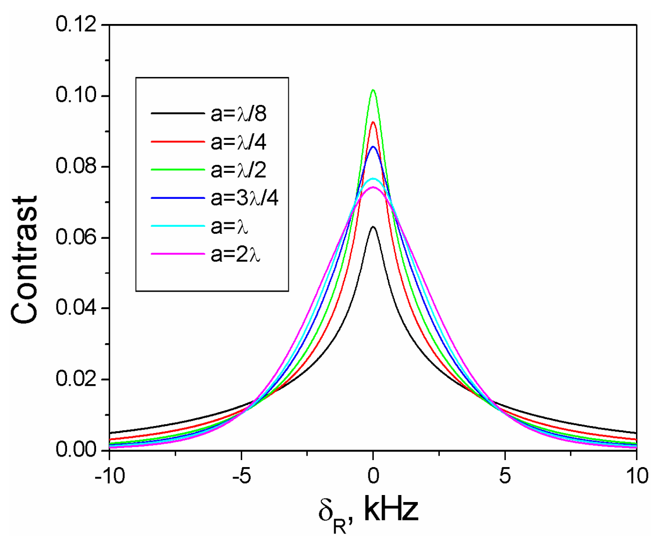

Figure 6. CPT signal line shape for specular-uncoherent reflection. $\gamma' = 2\cdot 10^9\ s^{-1}$, $V_1 = V_2 = 10^6\ s^{-1}$

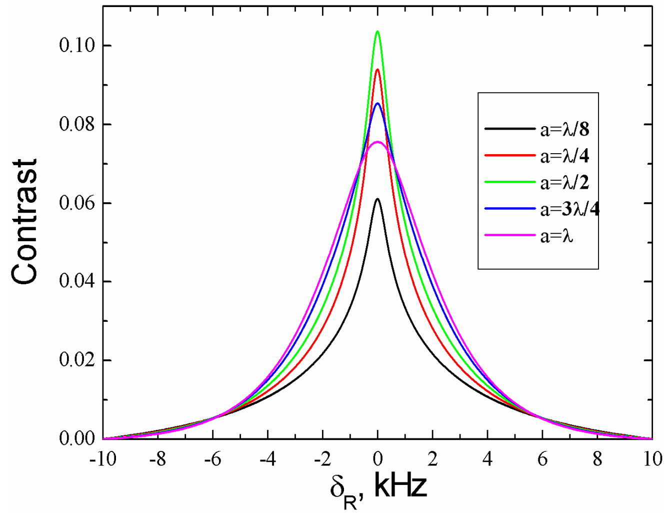

Figure 7. CPT signal line shape for uncoated cell. $\gamma' = 2\cdot 10^9\ s^{-1}$, $V_1 = V_2 = 10^6\ s^{-1}$

Figures 5 — 7 shows that if the relaxation rate γ' of optical coherence is comparable to or higher than (in our example equal) kv_T the cell size and the type of boundary conditions sufficiently affects on the CPT line shape. We can see that only specular-coherent reflection boundary conditions can provide the narrowing if the cell length is less than $\lambda/2$. Moreover the contrast remain high only if the cell length is less than $\lambda/4$. It should be noted that the maximum contrast for specular-incoherent boundary condition and uncoated cell reaches only 0.11 (see Figs. 6 and 7), whereas for specular-coherent conditions it reaches 0.85 for $a = \lambda/8$.

Therefore we can see two different dark-resonance line narrowing mechanisms: light-induced narrowing and Dicke narrowing. Light-induced narrowing takes place if the relaxation rate γ' of optical coherence is much less than kv_T independently of the type of coating of gas cell walls, and the Dicke narrowing takes place if γ' is comparable to or higher than kv_T. Dicke narrowing for CPT resonance takes place only for the coating (such as long-chain paraffin) that saves the atomic polarization state.

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