Modification of D₂-line absorption contours of alkali
atoms in antirelaxation coated cells

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Abstract. Absorption features in cells with antirelaxation coating in D₂-lines of alkali metal
atoms due to the competition of cyclic and open transitions lying within the Doppler contour
are studied. In cells of this type ensemble of atoms is optically pumped over all velocities
and the entire volume of the cell during the characteristic time \( T \), which noticeably exceeds
local pumping times related to resonant atomic velocities. If the frequency scanning time
is comparable to \( T \), the retardation effect results in significant distortion of the laser wave
absorption contour and in emergence of the hysteresis phenomenon. The developed theory
for a realistic scheme of atomic levels with one cyclic and two open transitions describes
experimentally observed absorption contours, their dependence on the light power and the
frequency scanning direction.

Cells with antirelaxation coating are widely used in quantum metrology, frequency standards
and magnetometry to obtain narrow metrological resonances [1–4] due to enlarged probability
of preserving the atomic inner state after collisions with the cell walls. As it was shown [5–7]
the frequency corresponding to the maximum of absorption and the maximum value of the
absorption itself in D₂-lines of alkali atoms depend on a sign of the laser frequency scanning
direction, if the scanning time is comparable to characteristic relaxation times due to collisions
with the cell walls. Absorption contour is found to be significantly distorted — absorption is
smaller compared to the cell without coating and this effect is more expressed in the range of
open atomic transitions than in the range of cyclic transition.

Cylindrical cell (50 mm in length, 35 mm in diameter) with \(^{133}\)Cs vapor was used in the
experiment. Line width of the laser with external resonator was of the order of 1 MHz. Light
was linearly polarized and beam size in cross section (5 mm × 2 mm) was smaller by almost
an order of magnitude than the diameter of the cell. Absorption contours obtained in the
experiment for scanning rate of about 50 MHz/ms with increasing (1+, 2+) and decreasing
(1−, 2−) of frequency for two values of light power are shown in Fig. 1.

The simplest 4-level scheme [7] based on a single cyclic and a single open transition
qualitatively explains the characteristic features of the absorption contour — its asymmetry and
a kind of hysteresis effect, that is, the dependence on the sign of frequency scanning. However,
such a model of levels does not allow to describe dependence of the crossing point for different
frequency scanning directions on light power (see the points \( \omega^* \) and \( \omega^\# \) in Fig. 1.)
Figure 1. Absorption contours of the $^{133}$Cs D$_2$-line long-wavelength components for the light power of 20 $\mu$W (curves 1+ and 1−) and 50 $\mu$W (curves 2+ and 2−). Absorption contour in the cell without antirelaxation coating is shown by dashed line for comparison. Scanning rate is 50 MHz/ms. Signs ± correspond to scanning process with increasing and decreasing of the frequency.

In the given paper we consider interaction of the monochromatic laser wave $E_L \cos \omega_L t$ with an atomic system that has one cyclic ($|g\rangle \leftrightarrow |5\rangle$) and two open ($|g\rangle \leftrightarrow |3\rangle, |4\rangle$) transitions (Fig. 2). Such a levels scheme corresponds to the long-wavelength components of an alkali atom D$_2$-line.

Atoms interact with laser radiation during the time $\tau_0$ of flight through the cross section of the beam. Its value is of the order of $\tau_0 \sim 10^{-5}$ s for thermal velocities $v_T$, so that the parameter $\gamma \tau_0 \gg 1$, where $\gamma$ is the natural width of the excited state levels ($\gamma/2\pi \approx 5.2$ MHz for $^{133}$Cs). In order to register the absorption contour, the laser wave frequency is varied in time according to the law $\omega_L = \omega_0 \pm st$ with some finite rate $s = \omega_D/\tau_{sc}$, where $\omega_D = kv_T$ is the characteristic width of the Doppler contour and $\tau_{sc}$ is the scanning time. We consider the latter to be significantly larger than the flight time $\tau_0$, which is, in its turn is noticeably exceeds $\tau_0$. In this case the variation of frequency $\omega_L$ during these times of flight can be neglected and the description of the time evolution is considerably simplified.

Detunings of the laser frequency from working atomic transitions with taking into account the Doppler shift due to longitudinal velocity have the form: $\Delta_3(t, v) = \pm st + \Delta/2 - kv$, $\Delta_4(t, v) = \pm st - \Delta/2 - kv$, $\Delta_5(t, v) = \pm st - \Delta_0 - kv$. As far as the Doppler width is much larger than the homogeneous broadening, $\omega_D \gg \gamma$, the resonant interaction for the transitions under consideration is realized for significantly different longitudinal velocities.

When an atom passes through the beam, a change of the local (with respect to velocities) populations $\rho_{gg}(t, v)$ and $\rho_{3\tilde{g}}(t, v)$ of the ground state sublevels

$$\frac{d\rho_{gg}}{dt} = -\frac{d\rho_{3\tilde{g}}}{dt} = -\Gamma(t, v)\rho_{gg}$$

in the low saturation regime is determined by optical pumping rate of the non-absorbing state $|\tilde{g}\rangle$:
Figure 2. Left: Hyperfine structure of the states $6S_{1/2}$ and $6P_{3/2}$ and long-wavelength components of $^{133}$Cs D$_2$-line. Transition $F_g = 4 \rightarrow F_e = 5$ is a cyclic one, transitions $F_g = 4 \rightarrow F_e = 3, 4$ are open ones. Excited state for forbidden transition $F_g = 4 \rightarrow F_e = 2$ is represented by a dashed line. Frequency interval of 9.1926 GHz between levels $F_g = 4$ and $F_g = 3$ is much larger than Doppler width, which is of the order of 380 MHz. Therefore optical pumping of level $F_g = 3$, represented by a dashed line, occurs on the open transitions.

Right: Five-level scheme of transitions considered in the theory.

\[ \Gamma(t, v) = \Gamma_3(t, v) + \Gamma_4(t, v) = \frac{\gamma_3 \Omega_3^2}{\gamma^2/4 + \Delta_3^2(t, v)} + \frac{\gamma_4 \Omega_4^2}{\gamma^2/4 + \Delta_4^2(t, v)}, \]  

where $\Omega_{3,4} = d_{3,4} E_L/2\hbar$ are the Rabi frequencies for the transitions $|g \rangle \Rightarrow |3 \rangle$, $|4 \rangle$, respectively, $\gamma_3$ and $\gamma_4$ are partial spontaneous decay rates of the states $|3 \rangle$ and $|4 \rangle$ to the non-absorbing state $|\tilde{g} \rangle$. $\gamma = \gamma_5 = \gamma_4 + \gamma_3 = \gamma_3 + \gamma_3$. At each moment of time, i.e. for each value of the laser frequency, the two terms in Eq. (2) look like small narrow peaks in the vicinities of the two significantly different resonant velocities.

The probability for an atom with a given longitudinal velocity to transit into the non-absorbing state $|\tilde{g} \rangle$ during one act of flight is $\tau_0 \Gamma(t, v)$. Therefore the equilibrium velocity distribution function $f(v) = \frac{1}{\tau_0 \sqrt{\pi} v^2} e^{-(v/v_T)^2}$ for the states $|g \rangle$, $|\tilde{g} \rangle$ is perturbed in the small vicinities $\delta v \sim \gamma/k \ll v_T$ of the resonant velocities, which are determined by the condition $\Delta_{3,4}(t, v) = 0$. Renewal of the equilibrium velocity distribution for the states $|g \rangle$ and $|\tilde{g} \rangle$ occurs due to collisions with the cell walls in a period of the order of the time $\tau_v$ of flight through the cell, which is noticeably larger than the flight time $\tau_0$ through the beam. In other words collisions with the walls extend local perturbation of the velocity distribution functions for the states $|g \rangle$ and $|\tilde{g} \rangle$ to the entire Doppler contour in a stretch of $\tau_v$. Laser field violates the distribution function $f(v)$ only in a small range near resonant velocities. After integration over all velocities we find that the total populations of the ground state sublevels $n_g(t)$ and $n_{\tilde{g}}(t) = 1 - n_g(t)$ change much slower, namely, during the characteristic time $T$, which has the form...
\[ \frac{\tau_v}{T(t)} = \left(1 - e^{\tau_0 \Gamma(t, v)}\right), \quad (3) \]

and generalizes the result obtained in [5] onto the case of two open transitions. Here the angular brackets denote averaging over velocities with the Maxwellian distribution. It should be noted that according to Eqs. (2), (3) the function \( T(t) \) depends on time via the frequency scanning law \( \omega_L(t) \). This simple result holds true provided the scanning process is slow enough and resonance velocities corresponding to working atomic transitions remain well separated during the relaxation time \( \tau_v \), i.e. \( k_{\text{D}} v \sim \tau_v s \sim \tau_v \omega_D/\tau_{\text{sc}} \ll \Delta, \Delta_0 \ll \omega_D \).

The relaxation of the populations of the ground state sublevels to their equilibrium values due to collisions of the atom with the cell walls occurs in a characteristic time \( \tau_g \). In antirelaxation coated cells it significantly exceeds not only the flight times, but also the optical pumping time of the whole ensemble of atoms over all velocities: \( \tau_g \gg T \sim \tau_v \omega_D/\gamma \gg \tau_v \). Now we focus on the case when \( \tau_{\text{sc}} \sim T \) and the retardation effect plays an important role. As soon as \( \tau_{\text{sc}} \) is much smaller than \( \tau_g \), the process of collisional relaxation of total populations can be neglected, and population of the state \( |g\rangle \) is described by expression \( n^g_g(t) = \frac{1}{2} e^{\left(-\int_{-\infty}^{t} \frac{dt}{T(t)}\right)} \), which contains explicitly the retardation effect.

We integrate using the approximate relation \( 1/ (\psi^2 + \gamma^2/4) = (2\pi/\gamma) \delta(\psi) \) and arrive at:

\[
n^g_g(\omega_L) = \frac{1}{2} e^{\frac{\pi \tau_{\text{sc}}}{4 \omega_D \tau_v} \left[ (1 - e^{-N_3}) \left( 1 \pm \Phi \left( \frac{\Delta_1}{\omega_D} \right) \right) + (1 - e^{-N_4}) \left( 1 \pm \Phi \left( \frac{\Delta_4}{\omega_D} \right) \right) \right]}, \quad (4)
\]

where \( N_{3,4} = 4\tau_0 \gamma_{3,4} \Omega^2_{3,4}/\gamma^2, \Delta_{3,4} = \omega_L - \omega_0 \pm \Delta/2, \Phi \) is error function: \( \Phi(x) = \frac{2}{\sqrt{\pi}} \int_{-\infty}^{x} e^{-u^2} du \), signs \( \pm \) correspond to the two different frequency scanning directions and responsible for the hysteresis phenomenon.

By summarizing the absorption coefficients for the cyclic \( k_3 = (\gamma \Omega^2_{3,4}/(\Delta^2_3(\omega_L, v) + \gamma^2/4)) n^g_g(\omega_L) \) and open transitions \( k_{3,4} = (\gamma/\tau_0 \gamma_{3,4})(1 - e^{\tau_0 \Gamma_{3,4}(\omega_L, v)}) n^g_g(\omega_L) \), we obtain the final expression for the total absorption coefficient:

\[
k^\pm(\omega_L) = \frac{\sqrt{\tau_v \gamma^2}}{2\tau_0 \omega_D} \left[ (1 - e^{-N_3}) e^{-\Delta_1/\omega_D} + (1 - e^{-N_4}) e^{-\Delta_4/\omega_D} + 4\tau_0 \Omega^2_{3,4} e^{-\Delta_5/\omega_D} \right] n^g_g(\omega_L). \quad (5)
\]

Here \( \Omega_5 = \delta_5 \Delta_{\text{L}}/2\hbar \) is the Rabi frequency for the transition \( |g\rangle \Rightarrow |5\rangle \), \( \Delta_5 = \omega_L - \omega_0 - \Delta_0 \). Influence of the antirelaxation coating, including the hysteresis effect, is described by the factor \( n^g_g(\omega_L) \).

If optical pumping is small, i.e. \( N_{3,4} \ll 1 \), the factors \( 1 - e^{-N_{3,4}} \) standing in the exponent in Eq. (4) are equal to \( N_{3,4} \) in the lowest order approximation and thereby are proportional to the field intensity. The latter appears as a common multiplier for the both frequency scanning directions and does not change the position of the crossing point of the absorption contours \( k^\pm(\omega_L) \). Dependence of the crossing point position on laser radiation power appears in the higher orders in terms of the field intensity.

Crossing point frequency is determined by the condition:

\[
e^{-N_3} \Phi \left( \frac{\Delta/2 + \omega_L - \omega_0}{\omega_D} \right) = e^{-N_4} \Phi \left( \frac{\Delta/2 - \omega_L + \omega_0}{\omega_D} \right). \quad (6)
\]
It is obvious that in the case $N_3 = N_4$ the condition (6) is satisfied by $\omega_L = \omega_0$, that is, the crossing occurs when the laser frequency is equal to the half-sum of frequency intervals between levels $|g\rangle$, $|3\rangle$ and $|g\rangle$, $|4\rangle$. For $^{133}$Cs atoms we have $d_4 > d_3$. Therefore Eq. (6) is satisfied if $\omega_L < \omega_0$, so that the crossing point shifts toward the level $|3\rangle$ when the intensity increases, as it has been observed in the experiment, see Fig. 1.

Thereby, the theory, generalized to the realistic model with one cyclic and two open transitions, as it takes place in $D_2$-lines of alkali metal atoms, adequately describes the hysteresis effect and dependence of the crossing point frequency for two opposite scanning directions on laser radiation power, that are observed in experiment with $^{133}$Cs atoms.

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