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Narrowing of the coherent population trapping resonance under zone pumping in cells with different characteristics of the wall coating

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Abstract. **It is shown that when coherent population trapping (CPT) resonance is excited by a narrow laser beam, the presence of elastic collisions with the cell wall significantly affects the line shape of the CPT-resonance. We have constructed a theoretical model, which is based on averaging over the random Ramsey sequences of the atom dwell time in the beam and dark zones and takes into account the probability of elastic bounce of an atom from the wall.**

Keywords: *coherent population trapping, elastic collisions with the wall.*

1. Introduction

Coating the walls of a cell containing vapours of alkali metals with a special antirelaxation composition is one of the methods for increasing the duration of coherent interaction of atoms with the field [1, 2]. Investigation of relaxation of rubidium atoms in a cell with paraffin-coated walls [1] showed that at least some of the atoms in a collision with the wall 'stick' to it for some time during which there occurs an exchange of kinetic energy, and after that the atoms return in a cell volume with a new velocity. At the same time, new wall coating materials and new technologies for coating deposition are being actively developed [3]. On these coatings atoms can, generally speaking, experience elastic collisions during which sticking is absent.

In this paper we show that the presence of elastic collisions with the cell wall largely determines the shape of the line of the coherent population trapping resonance (CPT-resonance) in a cylindrical cell illuminated by a laser beam of small diameter.

It is known (see, for example, [4 – 11] and references therein) that formation of the CPT-resonance in these cells is affected by the process associated with the displacement of active atoms from the zone illuminated by a laser beam in the dark zone and back, during the existence of coherence between the hyperfine sublevels of the ground state of the atom this displacement occurring repeatedly. In paper [6] and, independently, in papers [5, 7] the authors constructed a theoretical model of formation of the CPT-resonance in a cylindrical cell, based on averaging over the random Ramsey sequences of the atom dwell time in the beam and dark zones. The key point of the model was the assumption that atoms stick to the wall in each collision with it, and then return to the cell volume with a new velocity, unrelated to the velocity of an atom before the collision.

In this paper, we generalise the model to the case when the atom can also experience elastic collisions with the cell wall. We assume that in elastic collisions, firstly, all the atomic velocity components parallel to the wall (and the perpendicular component changes the sign) are retained, and secondly, the distribution of the atom dwell time in the beam and dark zones changes. We introduce the reflective coating coefficient α, which is the probability of an elastic collision of an atom with the wall of the cell. We study the influence of this coefficient on the line shape of the CPT-resonance at various laser intensities and provided that the width of the emission spectrum is much smaller than the Doppler width of the optical transition.

2. 2. Theoretical model

2.1. The equations for the density matrix

To describe the internal state of the active atoms interacting with the field, use is made of a three-level model (a lambda-scheme, Fig. 1). The equation for the density matrix ρ describing the internal state of an atom can be written as

$$\dot{\rho}_{ij} = -\frac{\mathrm{i}}{\hbar}\sum_{k}(H_{ik}\rho_{kj} - \rho_{ik}H_{kj}) + \sum_{k,l}\Gamma_{ijkl}\rho_{kl}, \tag{1}$$

where H_{ik} is the matrix element of the Hamiltonian $\hat{H} = \hat{H}_0 + \hbar\hat{V}(v_z, t)$ [$\hat{H}_0$ is the Hamiltonian of a free atom, $\hbar\hat{V}(v_z, t)$ is the operator of the dipole interaction of an atom with the laser field, which depends on the time t and projection v_z of the velocity in the direction of radiation propagation] and Γ_{ijkl} is the element of the relaxation matrix.

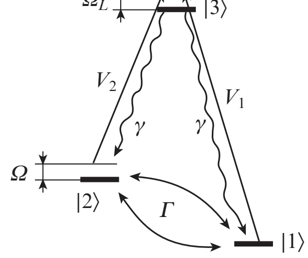

Figure 1. Lambda-scheme of interaction of a three-level atom with the field.

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The absorption of light in the cell is proportional to the population ρ_{33} of the excited state, which can be expressed in terms of populations ρ_{11}, ρ_{22} and coherence ρ_{12} in the ground state by adiabatic elimination [12]. In weak fields, when $V_{1,2} \ll \gamma$ [V_1 and V_2 are the matrix elements of the interaction operator of atoms with a resonant component of the field (Rabi frequency), 2γ is the rate of spontaneous relaxation of the excited state], the population ρ_{33} is much smaller than the populations ρ_{11} and ρ_{22} of the sublevels of the ground state. We also neglect the Doppler frequency shift of the microwave transition. This approximation is valid if the longitudinal dimensions of the cells are small compared with the wavelength λ_{21} of the transition between the states $|1\rangle$ and $|2\rangle$ (Dicke narrowing [13]).

Using the normalisation condition

$$\rho_{11} + \rho_{22} = 1 \tag{2}$$

and introducing the notation $\hat{\rho} = \{f, R, J\}$ ($f = \rho_{11} - \rho_{22}$, $R = \mathrm{Re}\rho_{12}$, $J = \mathrm{Im}\rho_{12}$), we can express the evolution equation for the density matrix of an ensemble of atoms in a laser beam as

$$\begin{aligned} \dot{f} &= G\frac{V_2^2 - V_1^2}{\gamma'} - (W + \Gamma)f - 4F\frac{V_1V_2}{\gamma'}J, \\ \dot{R} &= -G\frac{V_1V_2}{\gamma'} - (W + \Gamma)R - (\Omega - \Delta)J, \\ \dot{J} &= F\frac{V_1V_2}{\gamma'}f + (\Omega - \Delta)R - (W + \Gamma)J. \end{aligned} \tag{3}$$

Here, $G = G(v_z)$ and $F = F(v_z)$ are the real and imaginary parts of the expression $\gamma'/[\gamma' - \mathrm{i}(\Omega_L - kv_z)]$, respectively; $k = \omega/c$ is the wave number of optical radiation; Ω_L and Ω are the optical and two-photon (Raman) detuning; $\Delta = F(V_1^2 - V_2^2)/\gamma'$ is the light shift; $W = G(V_1^2 + V_2^2)/\gamma'$ is the optical pumping rate; $\gamma' = \gamma + \Gamma_L/2$ [14] is the relaxation rate of optical coherences ρ_{13} and ρ_{23}; Γ_L is the spectral width of laser radiation; Γ is the relaxation rate of the ground state. Expressed in terms of f, R and J, the excited state population is

$$\rho_{33} = \frac{W}{2\gamma} + \frac{G}{2\gamma\gamma'}[(V_1^2 - V_2^2)f + 4V_1V_2R]. \tag{4}$$

The linear system of equations (3) can be written symbolically in the matrix form:

$$\dot{\hat{\rho}}(v_z, t) = \hat{\hat{A}}(v_z)\hat{\rho}(v_z, t) + \hat{B}(v_z), \tag{5}$$

and expression (4) – in the form

$$\rho_{33}(v_z, t) = \hat{U}^{\mathrm{T}}(v_z)\hat{\rho}(v_z, t) + V(v_z). \tag{6}$$

Here, the quantities with a hat denote the column vectors, and with two hats – matrices, the superscript 'T' denotes transposition. Below, for the sake of brevity, we will not write the argument v_z in $\hat{\hat{A}}$, $\hat{B}$, $\hat{U}$ and V.

Evolution equations for the density matrix of atoms outside the laser beam can be obtained from equations (3), by setting $V_1 = V_2 = W = \Delta = 0$. Symbolically, this system can be written as

$$\dot{\hat{\rho}}(v_z, t) = \hat{\hat{A}}'\hat{\rho}(v_z, t). \tag{7}$$

Note that $\hat{\hat{A}}'$ is independent of v_z.

Thus, the density matrix describing the internal state of the atom obeys equation (5) when the atom is in the beam zone, and equation (7) when the atom is in the dark zone. The solution to equation (5) can be written in the form

$$\hat{\rho}(t) = \{\hat{\hat{I}} - \exp[\hat{\hat{A}}(t - t_0)]\}\hat{\rho}_{\mathrm{s}} + \exp[\hat{\hat{A}}(t - t_0)]\hat{\rho}(t_0), \tag{8}$$

where $\hat{\hat{I}}$ is the identity matrix; $\hat{\rho}_{\mathrm{s}} = -\hat{\hat{A}}^{-1}\hat{B}$ is the stationary solution to equation (5). Similarly, the solution to equation (7) can be written as

$$\hat{\rho}(t) = \exp[\hat{\hat{A}}'(t - t_0)]\hat{\rho}(t_0). \tag{9}$$

2.2. Motion of an atom in the cell

Consider a cylindrical cell, irradiated by a cylindrical laser beam propagating along the cylinder axis. Let us suppose first that the atom moving in the cell experiences only elastic collisions with the walls. Then, during each pass through the cell it either crosses the beam zone, as shown in Fig. 2a, or is outside it (Fig. 2b). In the first case, the atom is in the beam passing regime, and in the second case, – in the dark regime. Obviously, only atoms in the beam passing regime will contribute to the formation of the resonance. Let the atom be in the beam zone during the time t' at the moment of observation t_{a}. Before entering the beam zone the atom during the time τ' was in the dark zone. Before this the atom during the time τ was in the

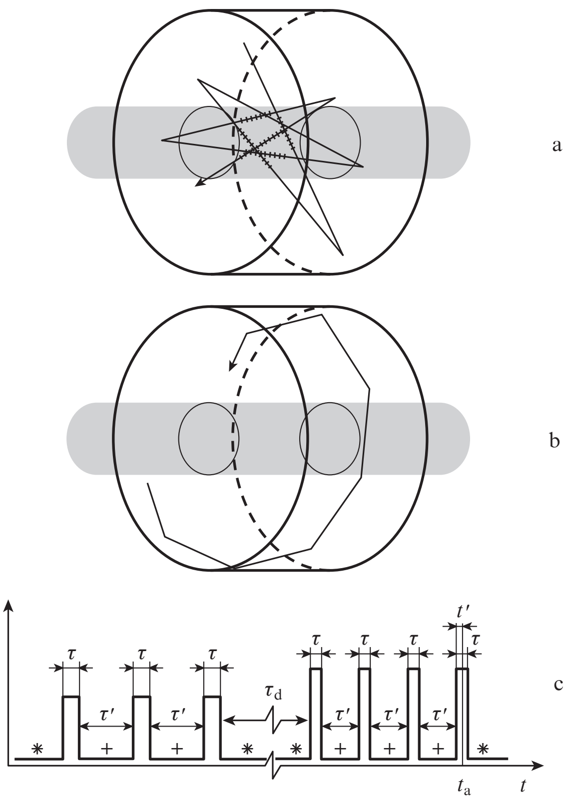

Figure 2. Trajectory of a single atom in a cell in the beam passing regime (a) and in the dark regime (b), as well as time dependence of the optical pumping rate W (depending on the atomic velocity due to the Doppler shift of laser radiation) (c). Crosses show the moments of elastic collisions of the atom with the wall of the cell, and asterisks – inelastic collisions.

beam zone, and again before that during the time τ' it was in the dark zone, etc. Using equations (8) and (9) we can easily obtain the expression

$$\hat{\rho}(t_a) = [\hat{\hat{I}} - \exp(\hat{\hat{A}}t')]\hat{\rho}_s + \exp(\hat{\hat{A}}t')\exp(\hat{\hat{A}}'\tau')$$
$$\times\{[\hat{\hat{I}} - \exp(\hat{\hat{A}}\tau)]\hat{\rho}_s + \exp(\hat{\hat{A}}\tau)\exp(\hat{\hat{A}}'\tau')$$
$$\times\{[\hat{\hat{I}} - \exp(\hat{\hat{A}}\tau)]\hat{\rho}_s + \exp(\hat{\hat{A}}\tau)\exp(\hat{\hat{A}}'\tau')[...]\}\}$$
$$= \{[\hat{\hat{I}} - \exp(\hat{\hat{A}}t')] + \exp(\hat{\hat{A}}t')\exp(\hat{\hat{A}}'\tau')$$
$$\times[\hat{\hat{I}} - \exp(\hat{\hat{A}}\tau)\exp(\hat{\hat{A}}'\tau')]^{-1}[\hat{\hat{I}} - \exp(\hat{\hat{A}}\tau)]\}\hat{\rho}_s. \quad (10)$$

Now we suppose that the atom can experience not only specular collisions with the wall, but also collisions, changing its velocity. After such a collision the atom, initially in the dark regime, can either dwell in it (which does not affect the equations for the density matrix), or switch to the beam passing regime. The atom, which before the collision was in the beam passing regime can either pass to the dark regime, or again to the beam passing regime, but with different values of the projection of the velocity vector and the angle of collision with the cell wall (and, consequently, with different values of τ and τ'). For definiteness, the time of entry of the atom into the beam zone after an inelastic collision with the wall we agree to call the onset of the beam passing regime, and the time of its exit – the end of the regime. Thus, if in the beam passing regime the atom undergoes N elastic collisions with the wall, it passes N times through the dark zone and $N + 1$ times through the beam zone (Fig. 2c).

The density matrix of the atom $\hat{\rho}_e$ at the time of exit from the beam passing regime can be obtained as expression (10), with the only difference being that the number of terms is now finite:

$$\hat{\rho}_e = \{\hat{\hat{I}} + \exp(\hat{\hat{A}}\tau)\exp(\hat{\hat{A}}'\tau') + ... + [\exp(\hat{\hat{A}}\tau)\exp(\hat{\hat{A}}'\tau')]^N\}$$
$$\times[\hat{\hat{I}} - \exp(\hat{\hat{A}}\tau)]\hat{\rho}_s + [\exp(\hat{\hat{A}}\tau)\exp(\hat{\hat{A}}'\tau')]^N\exp(\hat{\hat{A}}\tau)\hat{\rho}_b$$
$$= [\hat{\hat{I}} - \exp(\hat{\hat{A}}\tau)\exp(\hat{\hat{A}}'\tau')]^{-1}\{\hat{\hat{I}} - [\exp(\hat{\hat{A}}\tau)\exp(\hat{\hat{A}}'\tau')]^{N+1}\}$$
$$\times[\hat{\hat{I}} - \exp(\hat{\hat{A}}\tau)]\hat{\rho}_s + [\exp(\hat{\hat{A}}\tau)\exp(\hat{\hat{A}}'\tau')]^N\exp(\hat{\hat{A}}\tau)\hat{\rho}_b. \quad (11)$$

Here, $\hat{\rho}_b$ is the density matrix of the atom at the time of its entry into the beam. This matrix, in turn, can be related to the density matrix of the atom $\hat{\rho}_{e(-1)}$ at the time of the previous (as indicated by the superscript '–1') exit from the beam passing regime with the help of expression (9):

$$\hat{\rho}_b = \exp(\hat{\hat{A}}'\tau_d)\hat{\rho}_{e(-1)}, \quad (12)$$

where τ_d is the dwelling time of the atom in the dark regime. Similarly to (11), we can obtain an expression for the density matrix of the atom at the moment of observation t_a:

$$\hat{\rho}(t_a) = \exp(\hat{\hat{A}}t')\exp(\hat{\hat{A}}'\tau')[\hat{\hat{I}} - \exp(\hat{\hat{A}}\tau)\exp(\hat{\hat{A}}'\tau')]^{-1}$$
$$\times\{\hat{\hat{I}} - [\exp(\hat{\hat{A}}\tau)\exp(\hat{\hat{A}}'\tau')]^n\}[\hat{\hat{I}} - \exp(\hat{\hat{A}}\tau)]\hat{\rho}_s$$
$$+ [\hat{\hat{I}} - \exp(\hat{\hat{A}}t')]\hat{\rho}_s + \exp(\hat{\hat{A}}t')[\exp(\hat{\hat{A}}'\tau')\exp(\hat{\hat{A}}\tau)]^n\hat{\rho}_b, \quad (13)$$

and the expression for the population ρ_{33} of the excited state:

$$\rho_{33} = \hat{U}^{T}\{\exp(\hat{\hat{A}}t')\exp(\hat{\hat{A}}'\tau')[\hat{\hat{I}} - \exp(\hat{\hat{A}}\tau)\exp(\hat{\hat{A}}'\tau')]^{-1}$$
$$\times\{\hat{\hat{I}} - [\exp(\hat{\hat{A}}\tau)\exp(\hat{\hat{A}}'\tau')]^n\}[\hat{\hat{I}} - \exp(\hat{\hat{A}}\tau)]\hat{\rho}_s$$
$$+ [\hat{\hat{I}} - \exp(\hat{\hat{A}}t')]\hat{\rho}_s + \exp(\hat{\hat{A}}t')[\exp(\hat{\hat{A}}'\tau')\exp(\hat{\hat{A}}\tau)]^n\hat{\rho}_b\} + V, \quad (14)$$

where n is the number of collisions with the walls which the atom experiences during its continuous dwelling in the beam passing regime up to the moment of observation t_a. Averaging (14), (11) and (12) in atoms in the beam zone, we obtain the average population $\langle\rho_{33}\rangle$ of the excited state, which determines the absorption of radiation in the cell.

3. Results of calculations

We have performed calculations of the line shape and parameters of the dark resonance in vapours of three-level lambda-atoms in a cylindrical cell of radius $R = 0.5$ cm. The mass of the atoms m was assumed equal to the mass m_{Rb} of the ^{87}Rb isotope, the temperature was $T = 20\,°C$, the relaxation rate of the ground state was $\Gamma = 300$ s^{-1}, and the relaxation rate of optical coherences was $\gamma' = 1.8\times10^7$ s^{-1}. The Rabi frequencies V_1 and V_2 were considered equal ($V_1 = V_2 = \bar{V}$), and the optical detuning was $\Omega_L = 0$. As a main quantitative characteristic of the radiation intensity, use was made of the optical pumping rate $\bar{W}$, averaged over v_z and the volume of the cell:

$$\bar{W} = \frac{2\bar{V}^2 r^2}{\gamma' R^2}\bar{G}, \quad \bar{G} = \int_{-\infty}^{+\infty} M_1(v_z)G(v_z)\mathrm{d}v_z. \quad (15)$$

Here, $M_1(v_z) = (\pi v_T)^{-1/2}\exp(-v_z^2/v_T^2)$ is the Maxwell distribution function for the velocity v_z; r is the beam radius; $v_T = (2k_B T/m_{Rb})^{1/2}$ is the most probable velocity of atoms. For the parameters used in the calculation, $\bar{G} \approx 0.0168$. The value of $\bar{W}$ depends on the ratio of the radiation power to the cross-sectional area of the cell [6].

The spectral structure of the CPT-resonance with the probability of an elastic collision $\alpha = 0$ (a broad pedestal of width of several tens of kilohertz and a narrow central peak) was discussed in [4–7]. The broad pedestal is due to atoms that have passed through the beam zone only once, whereas the central peak is formed due to multiple passages. At nonzero values of α in a wide range of values of r and $\bar{W}$ there appears another 'intermediate' peak, whose width is greater than that of a narrow central peak, but lower than that of the broad pedestal (Fig. 3).This peak is due to atoms that have repeatedly passed through the beam zone, elastically bouncing off the walls of the cell. It is important to note that their longitudinal velocity v_z remains constant, in contrast to atoms that have once crossed the beam zone in the beam passing regime. The 'intermediate' peak appears when the characteristic dwelling time of the atom in this regime is sufficient for pumping in the dark state. Figure 4 shows the calculated CPT-resonance in a weak field ($\bar{W} = 10$ s^{-1}). The peak appears when the probabilities of elastic collision are equal to $\alpha = 0.75$ and 1, whereas for smaller values of α it is absent, because the atoms have too little time to dwell in the beam passing regime.

Finally, we note the emergence of the 'intermediate' peak when the diameter of the laser beam coincides with the diameter of the cell (Fig. 5). This is due to the fact that the atoms, whose longitudinal velocity v_z is small compared with the heat velocity, mainly contribute to the formation of the CPT-resonance. Thus, in elastic collisions with the wall the atom for a long time is in a 'resonant' velocity group of atoms inter-

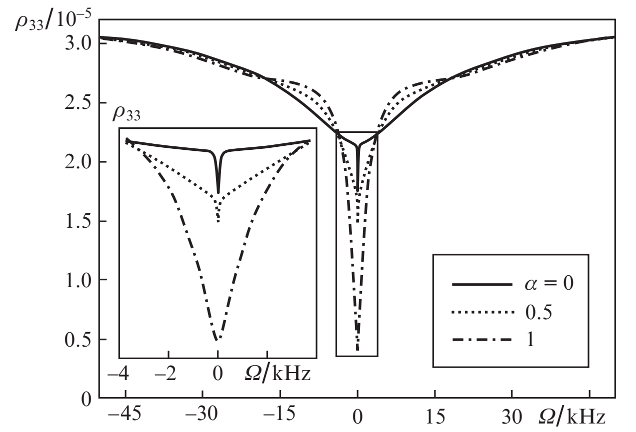

Figure 3. Shape of the CPT-resonance line at different α for $r = 1.5$ mm and $\overline{W} = 100$ s^{-1}. The inset shows the central part of the resonance.

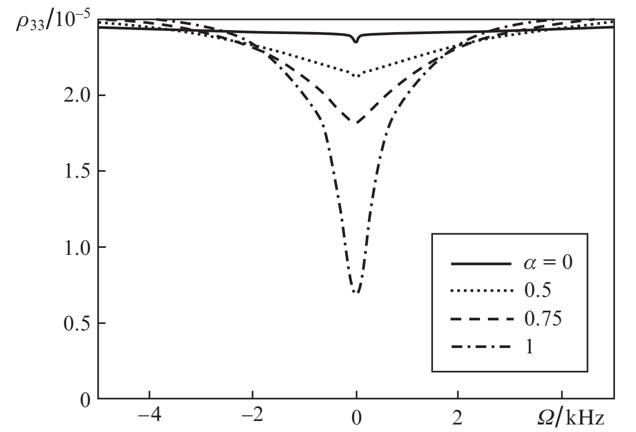

Figure 4. Shape of the CPT-resonance line (central part) at different α for $r = 0.5$ mm and $\overline{W} = 10$ s^{-1}.

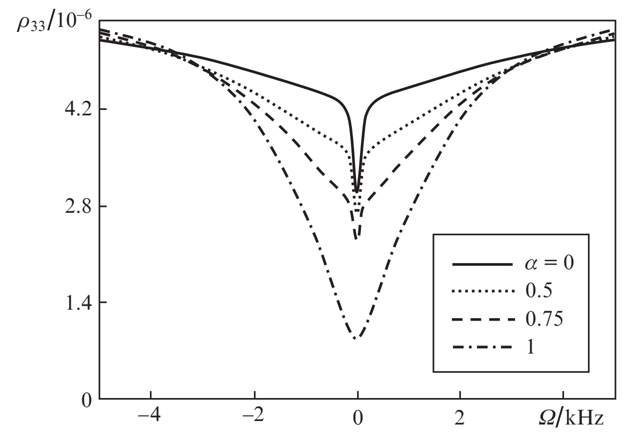

Figure 5. Shape of the CPT-resonance line at different α for $r = 5$ mm and $\overline{W} = 200$ s^{-1}.

acting with the field (and hence contributing to the formation of the CPT-resonance), whereas in inelastic collisions it leaves this group. The 'intermediate' peak, in this case, is due to atoms in a 'resonant' velocity group.

4. Conclusions

We have constructed a theory of the formation of CPT-resonances under conditions of zone pumping in a cylindrical cell with the antirelaxation wall coating on the assumption that the atoms can experience, with some probability, elastic collisions with the wall. We have shown that the presence of these collisions leads to a distortion of the line shape of the CPT-resonance, namely, to the appearance of an additional peak that is narrower than the 'pedestal', formed by a single passage of the atoms through the beam zone, and is wider than the narrow central peak.

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