Influence of the atomic-wall collision elasticity on the coherent population trapping resonance shape

G A Kazakov¹, A N Litvinov¹, B G Matisov¹, V I Romanenko², L P Yatsenko² and A V Romanenko³

¹ St. Petersburg State Polytechnical University, 29, Polytechnicheskaya st, St. Petersburg 195251, Russia
² Institute of Physics, National Academy of Sciences of Ukraine, 46, Nauky Ave., Kyiv 03028, Ukraine
³ Kyiv National Taras Shevchenko University, 4, Academician Glushkov Ave., Kyiv 03022, Ukraine

E-mail: kazjor@rambler.ru

Received 25 July 2011, in final form 12 September 2011
Published 11 November 2011
Online at stacks.iop.org/JPhysB/44/235401

Abstract
We studied theoretically a coherent population trapping resonance formation in a cylindrical cell without buffer gas irradiated by a narrow laser beam. We take into account non-zero probabilities of elastic (‘specular’) and inelastic (‘sticking’) collisions between the atom and the cell wall. We have developed a theoretical model based on averaging over the random Ramsey pulse sequences of times that the atom spent in and out of the beam. It is shown that the shape of the coherent population trapping resonance line depends on the probability of elastic collision.

1. Introduction
In the simplest case, the coherent population trapping (CPT) effect appears in a three-level Λ-system, see figure 1. The atom is interacting with a two-frequency laser field coupling two metastable states with a short-living state. When the frequency difference between the laser field components is equal to the frequency of the transition between two metastable states (two-photon resonance conditions), the ‘dark state’ that does not interact with the laser field appears. This ‘dark state’ is the coherent superposition of the metastable states. If one scans the frequency difference of two laser components near the two-photon resonance, one observes the CPT resonance, an abrupt decrease of the fluorescence intensity [1–3]. The CPT phenomenon allows us to create a window in the absorption spectrum [4]. This effect is used in the compact frequency standard [5, 6], the stimulated adiabatic passage technique of population transfer between the states of atoms and molecules [7], laser cooling [8] and magnetic field measurements [9, 10].

It is well known [11–19] that the CPT resonance shape is affected by the movement of atoms between the beam zone illuminated by the laser radiation and the dark zone. The theory of the CPT resonance formation in wall-coated cells without buffer gas based on averaging over random Ramsey sequences of times, which the active atom spent in the bright and dark zones, was developed in [12–14]. In these models, it was assumed that in any atomic-wall collision the atom sticks to the cell wall for some time. During this time the atom exchanges its kinetic energy with the cell wall and then it is released back to the cell volume with alternated velocity. At the same time, some parts of atoms can collide elastically with the cell wall.

In this work, we make the generalization of the model [12–14] to the case when there is a nonzero probability (reflection coefficient) of an elastic collision between the atom and the cell wall. Examples of elastic collision are evanescent wave mirrors [22, 23] or magnetic gradient mirrors [24–26] for cold atoms. We assume that an elastic collision does not
change velocity components parallel to the cell wall (normal component changes sign). We study the influence of the change velocity components parallel to the cell wall (normal and \( \Omega \)) of the optical transition. The density matrix equations describing the atomic polarization inside and outside the laser beam are presented in section 2. In section 3, we discuss the influence of elastic and inelastic collisions on the atomic motion in the cell and derive the expression for the excited state population. The procedure of the ensemble averaging is described in section 4. Results of calculations are presented in section 5. Conclusions are made in section 6.

2. Basic equations

Let us consider a gas of three-level atoms with the excited state \( |3\rangle \) and the metastable lower states \( |1\rangle \) and \( |2\rangle \) (one of which can be the ground state). The field component with frequency \( \omega_1 \) couples the states \( |1\rangle \) and \( |3\rangle \), whereas the component with frequency \( \omega_2 \) couples the states \( |2\rangle \) and \( |3\rangle \) (\( \Lambda \)-scheme of the atom–field interaction, see figure 1). The interaction of the field with the atoms is described by Rabi frequencies \( V_1 = \frac{1}{2} d_{13} \bar{E} / \hbar \), \( V_2 = \frac{1}{2} d_{23} \bar{E} / \hbar \), respectively. Here, \( d_{13} \) and \( d_{23} \) are the matrix elements of the atom’s dipole momentum, and \( \bar{E}_1 \) and \( \bar{E}_2 \) are the amplitudes of the laser field components. The one-photon frequency detuning of the first component is \( \Omega_1 = \omega_1 - \omega_{13} \), whereas \( \Omega = \omega_1 - \omega_2 - \omega_{12} \) is the two-photon detuning. Here, \( \omega_{ij} \) is the transition frequency between the \( i \) and \( j \) states. In our case, \( \omega_{ij} \) belongs to the microwave range.

The equation for the density matrix describing the atom in the field reads

\[
\dot{\rho}_{ij} = -\frac{i}{\hbar} \sum_k \left[ H_{ik} \rho_{kj} - \rho_{ik} H_{kj} \right] + \sum_{k,l} \Gamma_{ij,kl} \rho_{kl},
\]

where \( H_{ik} \) are the matrix elements of the Hamiltonian \( \hat{H} = \hat{H}_0 + h \bar{V} (\bar{v}, t) \), \( \hat{H}_0 = \hat{H}_0 + h \bar{V} (\bar{v}, t) \) is the Hamiltonian of the free atom. The operator of dipole interaction of the atom with the laser field \( h \bar{V} (\bar{v}, t) \) depends on time \( t \) and on the component of the atomic velocity \( \bar{v} \) along the field propagation direction; \( \Gamma_{ij,kl} \) are the elements of the relaxation matrix. The nonzero matrix elements are

\[
\begin{aligned}
\Gamma_{12,12} &= \Gamma_{21,21} = -\Gamma, \\
\Gamma_{11,22} &= \Gamma_{22,11} = -\Gamma_{11,11} = -\Gamma_{22,22} = \Gamma, \\
\Gamma_{13,13} &= \Gamma_{31,31} = \Gamma_{23,23} = \Gamma_{32,32} = -\gamma', \\
\Gamma_{11,33} &= \Gamma_{22,33} = \gamma / 2, \\
\Gamma_{33,33} &= -\gamma.
\end{aligned}
\]

Here, \( \gamma' = (\gamma + \Gamma_L) / 2 \) is the relaxation rate for optical coherences \( \rho_{33}, \rho_{23} \) [27], \( \Gamma_L \) is the spectral width of the laser radiation, \( \gamma \) is the spontaneous relaxation rate for the excited state and \( \Gamma \) is the relaxation rate in the ground state.

The light absorption in the cell is proportional to the excited state population \( \rho_{33} \). We consider the case of weak fields \( V_{1,2} \ll \gamma \). In this case, \( \rho_{33} \) is much smaller than the populations \( \rho_{11} \) and \( \rho_{22} \) and can be expressed via \( \rho_{11}, \rho_{22} \) and \( \rho_{12} \) using a standard adiabatic elimination procedure [28].

Usually this procedure is applied in the case of a large detuning \( \Omega_L \gg V_{1,2} \), but it is also valid in the case of \( \gamma' \gg V_{1,2} \). Let us consider for illustration an open \( \Lambda \)-system, when the atoms due to spontaneous emission decay from the excited state \( |3\rangle \) to the states different from \( |1\rangle \) and \( |2\rangle \). It is well known that the irreversible loss can be incorporated in the Hamiltonian [29]. For the open \( \Lambda \)-system in the rotating frame the Hamiltonian \( \tilde{H} \) is

\[
\tilde{H} = h \begin{pmatrix} 0 & V_1 & 0 \\ V_1 & 0 & -\Omega_L - i\gamma' / 2 \\ 0 & -\Omega & V_2 \end{pmatrix}.
\]

The set of equations for the amplitudes is

\[
\begin{aligned}
i \dot{C}_1 &= V_1 C_3, \\
i \dot{C}_2 &= -\Omega C_2 + V_2 C_3, \\
i \dot{C}_3 &= V_1 C_1 + V_2 C_2 - \left( \Omega_L + \gamma' / 2 \right) C_3.
\end{aligned}
\]

Typical evolution time of \( C_1 \) and \( C_2 \) is about \( 1 / V_{1,2} \). If \( |\Omega_L - i\gamma' / 2| \gg V_{1,2} \), we can put \( C_3 = 0 \) in (4) on the time scales of evolution of \( C_1, C_2 \) and then express \( C_3 \) via \( C_1 \) and \( C_2 \), i.e. to perform the adiabatic elimination of the excited state. In the case of a closed \( \Lambda \) system considered in our paper, the Hamiltonian \( \hat{H} \) is the Hermitian part of \( \tilde{H} \).

Considerations for the density matrix equations similar to ones from the previous paragraph can be performed analogically. Another important point is that in the case considered in this paper (room temperature and laser beam diameter of order of 1 mm or higher), the typical time of flight of the atom through the beam or dark zone is much larger than \( 1 / \gamma' \).

We also neglect the Doppler shift of the frequency of microwave transition or, what is the same, we suppose that the Doppler shift is equal for two optical transitions. This approximation is valid when the relative phase between two optical fields does not change significantly on the length of typical path of the atom (Dicke narrowing [30]). In [31, 32], it was shown that for the longitudinal cell size smaller than \( \lambda_{21} / 4 \) (where \( \lambda_{21} = 2 \pi c / \omega_{21} \) is the wavelength of transition between the states \( |1\rangle \) and \( |2\rangle \)), the shape of the CPT resonance in a coated cell without buffer gas practically does not depend on the cell length. Therefore, we can neglect the Doppler broadening of the microwave transition if the cell size is less
than $\lambda_{23}/4$ that is equal to 1.1 cm for the microwave transition in the $^{87}$Rb atom.

Using the normalization condition

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

and denoting $\hat{\rho} = \{f, R, J\}$, where $f = \rho_{11} - \rho_{22}$, $R = \text{Re}(\rho_{12})$, $J = \text{Im}(\rho_{12})$, we can rewrite the equations for the density matrix for the ensemble of atoms in the laser beam in the form

$$\dot{f} = G(V_f^2 - V_i^2) - (W + \Gamma)f - 4F\, V_i \, V_f, \quad \dot{R} = -\frac{G}{\gamma'} V_i \, V_f - (W + \Gamma)R - (\Omega - \Delta)J,$$

$$\dot{J} = F V_i V_f - (\Omega - \Delta)R - (W + \Gamma)J.$$  \hfill (6)

Here, $G = G(v_z)$ and $F = F(v_z)$ denote the real and the imaginary parts of the expression $\gamma'/(\gamma' - i(\Omega_1 - k v_1))$, respectively, $k = \omega_1/c$ is the wave number of the optical radiation, $\Delta = F(V_f^2 - V_i^2)/\gamma'$ is the light shift and $W = G(V_f^2 + V_i^2)/\gamma'$ denotes the optical pumping rate. The excited state population expressed in terms of $f$ and $R$ is

$$\rho_{33} = \frac{W}{\gamma'} + \frac{G}{\gamma'} \left[ (V_f^2 - V_i^2) \cdot f + 4V_i V_f \cdot R \right].$$  \hfill (7)

To simplify the calculation procedure we use the matrix notation formalism similar to \cite{33}. The set of equations (6) can be symbolically presented as

$$\dot{\hat{\rho}}(v_z, t) = \mathbb{A}(v_z) \hat{\rho}(v_z, t) + \hat{B}(v_z).$$  \hfill (8)

Also, we can write expression (7) in the form

$$\rho_{33}(v_z, t) = \hat{U}^\dagger(v_z) \rho_{33}(v_z, t) + V(v_z).$$  \hfill (9)

Furthermore, for the sake of brevity we will skip the argument $v_z$ in $\mathbb{A}$, $\hat{B}$, $\hat{U}$ and $V$.

The evolution equation for the density matrix of atoms outside the laser beam can be obtained from equations (6) by the substitution $V_i = V_f = W = \Delta = 0$. One can rewrite the resulting set of equations symbolically as

$$\dot{\hat{\rho}}(v_z, t) = \mathbb{A}'(v_z) \hat{\rho}(v_z, t).$$  \hfill (10)

Note that $\mathbb{A}'$ does not depend on $v_z$.

Finally, the density matrix of the atom is described by equation (8) inside the laser beam and by equation (10) outside the beam. The solution of equation (8) can be presented as

$$\hat{\rho}(t) = (\mathbb{I} - e^{\mathbb{A}'(t-t_0)}) \hat{\rho}_{S} + e^{\mathbb{A}'(t-t_0)} \hat{\rho}(t_0),$$

$$\hat{\rho}(t) = e^{\mathbb{A}'(t-t_0)} \hat{\rho}(t),$$

$$\hat{\rho}(t) = e^{\mathbb{A}'(t-t_0)} \hat{\rho}(t_0).$$

Now let us consider a more common case when some of the wall collisions change atomic velocity. After such a collision the atom being initially in the dark regime can either stay in this regime (which does not affect the equations for the density matrix) or switch to the beam passing regime. An atom that was being in the beam passing regime before the collision can either pass into the dark regime or get back to the beam passing regime, but with a different value of the velocity and angle of reflection with the wall of the cell (and therefore with different values of $\tau$ and $\tau'$). For the sake of

![Figure 2](image-url)
definiteness, let us call the time of entry of the atom into the beam zone after an inelastic collision as the beginning of the beam passing regime, and the time of exit from the beam zone as the end, respectively. Obviously, if the atom undergoes \( N \) elastic collisions with the cell wall in the beam passing regime, then it goes \( N \) times through the dark zone and \( N + 1 \) times through the beam zone (see figure 2(c)).

The density matrix \( \hat{\rho} \) describing the atom at the time of exit from the beam passing regime can be obtained similar to expression (13). It reads

\[
\hat{\rho}_e = [I + e^{\hat{v}_z \cdot \tau_d}]^N[I - e^{\hat{v}_z \cdot \tau_d}] \hat{\rho}_b + (I - e^{\hat{v}_z \cdot \tau_d})^N e^{\hat{v}_z \cdot \tau_d} \hat{\rho}_b,
\]

where the angle brackets denote the averaging over the time \( t_A \) of the atom spent in the dark. Similar to (14), one can obtain the expression for the density matrix at the resonance is given by the atoms with small longitudinal velocity whose spectral width is much smaller than the Doppler width we consider the excitation of the CPT resonance by a laser passing regime prior to the observation time \( t_A \).

\[
\hat{\rho}_c = [I + e^{\hat{v}_z \cdot \tau_d}]^N[I - e^{\hat{v}_z \cdot \tau_d}] \hat{\rho}_b + (I - e^{\hat{v}_z \cdot \tau_d})^N e^{\hat{v}_z \cdot \tau_d} \hat{\rho}_b.
\]

Here, \( \hat{\rho}_b \) is the density matrix of the atom at the time of beginning of the beam passing regime, it can be expressed via the density matrix \( \hat{\rho}_{c,-1} \) for the previous time of exit from the beam passing regime (it is indicated by the index ‘\(-1\)’) using expression (12):

\[
\hat{\rho}_b = e^{\hat{v}_z \cdot \tau_d} \hat{\rho}_{c,-1}.
\]

where \( \tau_d \) is the time that the atom spent in the dark. Similar to (14), one can obtain the expression for the density matrix at the observation time \( t_A \):

\[
\hat{\rho}(t_A) = [I - e^{\hat{v}_z \cdot \tau_d}]^N[I - e^{\hat{v}_z \cdot \tau_d}] \hat{\rho}_b + (I - e^{\hat{v}_z \cdot \tau_d})^N e^{\hat{v}_z \cdot \tau_d} \hat{\rho}_b.
\]

The expression for the excited state population \( \rho_{33} \) reads

\[
\rho_{33} = \hat{U}^T [I - e^{\hat{v}_z \cdot \tau_d}]^N[I - e^{\hat{v}_z \cdot \tau_d}] \hat{\rho}_b + (I - e^{\hat{v}_z \cdot \tau_d})^N e^{\hat{v}_z \cdot \tau_d} \hat{\rho}_b + V.
\]

Here, \( n \) is the total number of collisions in the beam passing regime immediately prior to the observation time \( t_A \). Averaging expressions (17), (14) and (15) over all of the atoms in the beam zone, we obtain the average excited state population \( \langle \rho_{33} \rangle \) that determines the absorption of light in the cell.

4. Ensemble averaging

4.1. General scheme of averaging

Let us consider in detail the averaging of (17). In this paper, we consider the excitation of the CPT resonance by a laser whose spectral width is much smaller than the Doppler width of the optical transition. Hence, the main contribution to the resonance is given by the atoms with small longitudinal velocity that are rarely colliding with the ends of the cell. We assume that the collisions of the atoms in the beam passing regime with the side walls of the cell do not change \( v_z \). The average population \( \langle \rho_{33} \rangle \) is

\[
\langle \rho_{33} \rangle = \langle \hat{U}^T [I - e^{\hat{v}_z \cdot \tau_d}] \hat{\rho}_b + (I - e^{\hat{v}_z \cdot \tau_d})^N e^{\hat{v}_z \cdot \tau_d} \hat{\rho}_b + V \rangle
\]

where the angle brackets denote the averaging over the \( z \)-component of the velocity \( v_z \), over the times \( t, \tau, \tau', \tau_d \) and over the numbers \( n \) and \( N \) of elastic collisions in the beam passing regime prior to the observation time \( t_A \) and prior to the exit from the beam passing regime (in the expression for \( \langle \rho_{33} \rangle \)) respectively. The averaged density matrix \( \langle \hat{\rho}_b \rangle \) of the atom entering the beam passing regime can be obtained by averaging of expressions (14) and (15):

\[
\langle \hat{\rho}_b \rangle = \hat{C} + \mathbb{D} \langle \hat{\rho}_b \rangle.
\]

\[
\mathbb{D} = (e^{\hat{v}_z \cdot \tau_d} N e^{\hat{v}_z \cdot \tau_d}).
\]

Let us average over \( n \) and \( N \) first. Assuming that the probability of the elastic scattering \( \alpha \) does not depend on the velocity of an atom and its internal state, we obtain the probability \( P(n) = (1 - \alpha)\alpha^n \) for \( n \) consecutive elastic collisions. Hence, for any matrix \( \mathbb{R} \) we obtain the matrix \( \mathbb{R}^n \) averaged over \( n \) as

\[
\langle \mathbb{R}^n \rangle_n = \sum_{n=0}^{\infty} P(n) \mathbb{R}^n = (1 - \alpha)(1 - \alpha \mathbb{R})^{-1}. \tag{24}
\]

Exactly the same kind of distribution \( P(n) = (1 - \alpha)\alpha^n \) is valid for a total number of elastic collisions \( N \) in the beam passing regime; therefore, the matrix \( \mathbb{R}^N \) averaged over \( N \) is described by the same expression: \( \langle \mathbb{R}^N \rangle_N = \langle \mathbb{R}^n \rangle_n \).

The next steps are the averaging over times \( t, \tau, \tau' \) and \( \tau_d \). It is obvious that the averaging over the time \( \tau_d \) of atomic stay in the dark regime is independent of \( t, \tau, \tau' \), while \( \tau \) and \( \tau' \), in general, are determined by the transversal component of the atomic velocity in the beam passing regime and by the angle between the velocity and the wall of the cell. The time \( \tau \) is also determined by the atom’s position in the beam zone at the observation time \( t_A \). Strictly speaking, one must integrate over four variables \( t, \tau, \tau' \) and \( v_z \). However, we make a simplifying assumption which reduces the number of integration variables to 1.

In the beam passing regime the atom goes \( N \) times through the dark zone and \( N + 1 \) times through the beam zone. Each passage through the dark zone takes time \( \tau' \) and each passage through the beam zone takes time \( \tau \). Obviously, for each passage the times \( \tau' \) and \( \tau \) are not independent. According to our assumption we consider times of different passages of the dark and light zones as independent random variables. We emphasize that this assumption refers only to ensemble averaging. In this case, the averaged values of \( e^{\hat{v}_z \cdot \tau} \), \( e^{\hat{v}_z \cdot \tau'} \) and \( e^{\hat{v}_z \cdot \tau_d} \) in (18)–(23) can be calculated independently. Consider, for example, the averaged expression of \( e^{\hat{v}_z \cdot \tau} \). It is easy to present this expression in the form [12]

\[
\langle e^{\hat{v}_z \cdot \tau} \rangle = \mathbb{X} \begin{pmatrix} \langle e^{\hat{v}_z \cdot \tau} \rangle & 0 & 0 \\ 0 & \langle e^{\hat{v}_z \cdot \tau} \rangle & 0 \\ 0 & 0 & \langle e^{\hat{v}_z \cdot \tau} \rangle \end{pmatrix} \mathbb{X}^{-1}, \tag{25}
\]

where \( \mathbb{X} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix} \).
where $\mathcal{X} = (\mathcal{X}^{(1)}, \mathcal{X}^{(2)}, \mathcal{X}^{(3)})$ is the matrix constructed of the eigenvectors of $A$ corresponding to the eigenvalues $\lambda_1, \lambda_2, \lambda_3$. Therefore, averaging over $\tau$ is reduced to the calculation of integrals

$$
\langle e^{i\lambda \tau} \rangle_{\tau} = g_1(\lambda) = \int_{0}^{\infty} e^{i\lambda \tau} \cdot f_1(\tau) \, d\tau,
$$

that can be performed analytically. Here, $f_1$ is the probability density function of the random variable $\tau$. Averaging over $\tau$, $\tau'$ and $\tau_j$ is analogous to (26) with functions $g_1(\lambda)$, $g_1(\lambda)$ and $g_1(\lambda)$. Note that $\Re(\lambda) < 0$.

### 4.2. Averaging over $\tau$ and $t$

First of all, we find the probability density functions $f_1$ and $f_2$ of the random variables $\tau$ and $t$. The time $\tau$ that the atom spent in the beam zone is

$$
\tau = \frac{2\sqrt{R^2 - R^2 \sin^2 \varphi}}{v_{\perp}},
$$

where $\varphi$ is the angle between the atomic velocity projection on the plane orthogonal to the cell axis and the normal vector to the cell surface, $R$ is the radius of the cell, $r$ is the radius of the beam and $v_{\perp}$ is the component of the velocity orthogonal to $z$. The probability density function of the transversal velocity $v_{\perp}$ is the two-dimensional Maxwell distribution $M_2(v_{\perp}) = 2\exp(-v_{\perp}^2/v^2_{\perp}) v_{\perp}/v^2_{\perp}$ where $v_{\perp} = \sqrt{2k_BT/m}$ is the most probable atomic velocity, $k_B$ is the Boltzmann constant, $T$ is the temperature and $m$ is the mass of the atom. The probability density $\Phi(\varphi)$ of the random variable $\varphi$ is proportional to $\cos \varphi$ [14, 34, 35]. Taking into account that in the beam passing regime $\varphi$ takes values between 0 and $\arcsin(r/R)$, we obtain

$$
\Phi(\varphi) = R \cos \varphi/r. \quad \text{Then, the cumulative distribution function} \quad F_1(\tau) \quad \text{of the random variable} \quad \tau \quad \text{is}
$$

$$
F_1(\tau) = \int_{0}^{\arcsin(r/R)} \Phi(\varphi) \int_{\sqrt{R^2 - R^2 \sin^2 \varphi}}^{\infty} M_2(v_{\perp}) \, dv_{\perp} \, d\varphi
$$

$$
= \frac{\tau v_{\perp} \sqrt{\pi}}{4r} \cdot \exp \left( \frac{2i\tau}{v_{\perp}} \right),
$$

and the probability density function is

$$
f_1(\tau) = \frac{dF_1(\tau)}{d\tau} = -\frac{1}{\tau} + \frac{v_{\perp} \sqrt{\pi}}{4r} \cdot \exp \left( \frac{2i\tau}{v_{\perp}} \right) \left[ 1 + \frac{8\tau^2}{\pi^2 \tau^2} \right].
$$

Note that the average time that the atom moves through the beam zone is

$$
\bar{\tau} = \int_{0}^{\infty} f(\tau) \, d\tau = \int_{0}^{\infty} (1 - F(\tau)) \, d\tau = \frac{\pi^{3/2}}{2} \frac{r}{v_{\perp}}.
$$

Let us find the probability distribution function $f_1$ of time $t$ that the atom stays continuously in the beam zone before the observation time $t_\Delta$. The averaging is carried out over all atoms in the beam zone. Let $dn/dt$ atoms arrive at the beam zone per unit of time. Then, the total number of atoms in the beam zone is

$$
N_{\text{tot}} = \frac{dn}{dt} \int_{0}^{\infty} (1 - F_1(\tau')) \, d\tau'.
$$

Here, we took into account that $1 - F_1(\tau')$ is the atom’s probability of remaining in the beam zone after time $\tau'$ from the entrance to the beam. Therefore, the number of atoms that have already spent time $\tau$ or more in the beam zone is

$$
N_{\tau} = \frac{dn}{dt} \int_{\tau}^{\infty} (1 - F_1(\tau')) \, d\tau'.
$$

Hence, the cumulative distribution function $F_2(t)$ of the random variable $t$ reads

$$
F_2(t) = 1 - \int_{0}^{\infty} (1 - F_1(\tau')) \, d\tau' = \frac{1 - F_1(t)}{\bar{\tau}}.
$$

and the corresponding probability density function

$$
f_2(t) = \frac{dv_{\perp}}{\pi^{3/2} v_{\perp}} \left[ 1 - \frac{v_{\perp} \sqrt{\pi}}{4r} \cdot \exp \left( \frac{2i\tau}{v_{\perp}} \right) \right].
$$

Using (34) it is easy to express $g_1(\lambda)$ in terms of $g_2(\lambda)$:

$$
g_1(\lambda) = 1 + \bar{\tau} g_2(\lambda).
$$

Note that the function $g_2(\lambda) = \int_{0}^{\infty} e^{i\lambda \tau} f_2(\tau) \, d\tau$ cannot be calculated in terms of elementary functions. We can find an approximate but close to accurate result by replacing the precise expression (35) by some approximation similar to (36)

$$
f_2(t) \approx \frac{v_{\perp}}{\bar{\tau}} \cdot f_2 \left( \frac{v_{\perp} t}{\bar{\tau}} \right) = \frac{v_{\perp}}{\bar{\tau}} \cdot f_2 \left( \frac{v_{\perp} t}{\bar{\tau}} \right).
$$

We choose the values of coefficients $A = 0.708 39$, $a = 0.49$, $b = 2.286$ and $c = 1.272$. These coefficients ensure (1) the same asymptotical behaviour of $f_2(t)$ and $f_2(v_{\perp} t/r) v_{\perp} / r$ at infinity, (2) coincidence of $f_2(\lambda)$ and $f_2(v_{\perp} t/r) v_{\perp} / r$ near the zero to the second order with respect to $\tau$ and (3) correct normalization $\int_{0}^{\infty} f_2(x) \, dx = 1$. The plots of the functions $r \cdot f_2(x \cdot r/v_{\perp}) / v_{\perp}$ and $f_2(x)$ are presented in figure 3.

The approximation (37) makes it possible to find the analytical expression for $g_2(\lambda)$:

$$
g_2(\lambda) = \tilde{g}(\lambda r/v_{\perp}).
$$
\[ \tilde{g}_x(\lambda) = \frac{2}{\pi^{3/2}} \frac{1}{b - \lambda} + \frac{A}{(c - \lambda)^2} \]
\[ + \frac{16}{3\pi^{3/2}} \left[ p(-\Lambda) - 3p(a - \Lambda) + 3p(2a - \Lambda) - p(3a - \Lambda) \right], \]  
where we denoted \[ p(\beta) = \beta \ln \beta. \]  
It is easy to find \( g_x(\lambda) \) from (38) and (36).

### 4.3. Averaging over \( \tau' \)

Now let us calculate the function \( g_x(\lambda) \). The time \( \tau' \) that the atom being in the beam passing regime spent in the dark zone continuously is

\[ \tau' = \frac{2}{v_x} (R \cos \varphi - \sqrt{r^2 - R^2 \sin^2 \varphi}). \]  

If the beam diameter is much smaller than the cell diameter, the expression in brackets can be approximately replaced by the constant

\[ \ell_x = 2 \int_0^{\arcsin(r/R)} \left( R \cos \varphi - \sqrt{r^2 - R^2 \sin^2 \varphi} \right) \Phi(\varphi) \, d\varphi \]
\[ = R \left[ 1 - \frac{r^2}{2} - \frac{\pi r}{2} + \frac{R^2}{r} \arcsin \frac{r}{R} \right] \]
that equals the average transversal path length in the dark zone. This approximation allows us to readily obtain the cumulative distribution function \( F' \) of the random variable \( \tau' \):

\[ F'(\tau') = e^{-\frac{\tau'}{\tau_x}}, \]  
and

\[ \tau' = \frac{\ell_x}{v_x} \sqrt{\pi}. \]  
An attempt to directly calculate the function \( g_x(\lambda) = \lambda \int_0^\infty (1 - F'(\varphi)) e^{\lambda \varphi} \, d\varphi \) does not yield the result that can be expressed through elementary functions. As in the previous case, we can find an approximate but close to accurate result by replacing the precise expression (42) by

\[ F'(\tau') \rightarrow F' \left( \tau' \frac{v_x}{\ell_x} \right), \]

\[ \tilde{F}'(x) \]
\[ = \begin{cases} 
1 - \frac{(1-e^{-x-x_0})^2}{(x-x_0)^2} - e^{-b(x-x_0)}(1 + b(x-x_0)), & x > x_0 \\
0, & x \leq x_0.
\end{cases} \]  

The values of the parameters \( a = 0.89, b = 2.56 \) and \( x_0 = 0.3864 \) ensure smoothness of the function \( \tilde{F}'(x) \) anywhere, good approximation of \( F'(x) \ell_x/v_T \) in the region where it is appreciably different from zero (see figure 4) and obtain the correct value of time \( \tau' \). Using the approximation function (44), we find

\[ g_x(\lambda) \approx \tilde{g}_x(\lambda \ell_x/v_T), \]
\[ \tilde{g}'(\Lambda) = A \Lambda^0 \left[ \frac{2b - \Lambda}{(b - \Lambda)^2} + p(4a - \Lambda) - 4p(3a - \Lambda) - 4p(a - \Lambda) + 6p(2a - \Lambda) + p(-\Lambda) + \frac{1}{\Lambda} \right]. \]  

### 4.4. Averaging over \( \tau_d \)

The last function we should find for calculating the fluorescence of the atom in the cell is \( g_x(\lambda) \). The exact calculation of the probability density function \( f_d(\tau_d) \) of the random variable \( \tau_d \) is a nontrivial problem because in the dark regime the trajectory of the atom consists of a random number of segments of random length; along each of them the atom moves with a random velocity. Nevertheless, it is clear that the probability density exponentially tends to zero when \( \tau_d \rightarrow 0 \). The minimal distance that an atom can travel in the dark regime in the transverse direction is \( 2(R - r) \). The transverse velocity \( v_x \) of the atom with probability greater than 98% is less than \( 2v_T \), so we assume that \( f_d(\tau_d) = 0 \) when \( \tau_d < \tau_0 = (R - r)/v_T \). If the beam diameter is much smaller than the diameter of the cell, the atom usually returns from the dark regime into the beam passing regime after many wall collisions. The probability for an atom to return to the beam passing regime after a certain collision does not depend on the number of previous collisions. Such processes are usually described by an exponential probability distribution function, so for the function \( f_d(\tau_d) \) of the random variable \( \tau_d \) we take

\[ f_d(\tau_d) = \begin{cases} 
he^{-h(\tau_d - \tau_0)}, & \tau_d > \tau_0 \\
0, & \tau_d < \tau_0.
\end{cases} \]  

where \( h = 1/(\ell_x - \tau_0) \). \( \tau_d \) is the average time of the atom being in the dark zone. From (26) and (46) it is easy to obtain \( g_x(\lambda) = e^{hN}/(1 - \lambda/h) \).

The value of \( \tau_d \) can be found from the ‘time balance’ conditions. It means that the total time that an atom spends in the beam or in the dark zone is proportional to the volume of this zone. Consider the cycle in which an atom being in the beam passing regime undergoes \( N \) elastic collisions with the wall, and then moves into the dark regime. The history of any atom can be represented as a sequence of such cycles. The total time \( \ell_b \) spent by an atom in the beam zone over the cycle is \( \ell_b = (N + 1) \tau_b \). In turn, the time \( \ell_d \) spent by the atom in a dark zone during the cycle is \( \ell_d = \tau_d = N \tau' \). The ratio of averaged times \( \ell_b \) and \( \ell_d \) over the different cycles must be equal to the ratio of volumes of the bright and dark zones. Hence, we find

\[ \tilde{\tau}_d = \left( \frac{R^2}{r^2} - 1 \right) (\ell_b + \tilde{\tau}' \ell_d), \]
Figure 5. The shape of CPT resonance for different $\alpha, V$ and $r$. Parameters: $r = 1.5$ mm, $V = 7.7 \times 10^5$ s$^{-1}$ (a, b); $r = 0.5$ mm, $V = 7.3 \times 10^3$ s$^{-1}$ (d); $r = 5$ mm, $V = 3.3 \times 10^5$ s$^{-1}$ (c); $r = 5$ mm, $V = 1.46 \times 10^6$ s$^{-1}$ (f).

(This figure is in colour only in the electronic version)

where $\bar{N} = \alpha/(1 - \alpha)$ is the average number of successive elastic collisions with the cell wall.

In the conclusion of this section we write down the expression for the averaged excited state population:

$$\langle \rho_{33} \rangle = \int_{-\infty}^{\infty} M_1(v_z) G_t(1 - \alpha)(I - \alpha G_t' G_t')^{-1} dv_z \cdot \langle \hat{\rho}_b \rangle \quad \text{(48)}$$

where expressions (22) and (23) for $\hat{C}$ and $\hat{D}$ can be rewritten as

$$\hat{C} = \int_{-\infty}^{\infty} M_1(v_z) \left( I - \alpha G_t G_t' \right)^{-1} \left( I - (1 - \alpha)G_t G_t' \right) \hat{\rho}_b dv_z, \quad \text{(53)}$$

and

$$\langle \hat{D} \rangle = \int_{-\infty}^{\infty} M_1(v_z) G_t(1 - \alpha) \left( I - \alpha G_t G_t' \right)^{-1} dv_z \quad \text{(54)}$$

where

$$G_t' = X' \begin{pmatrix} g_{r} (\lambda_1) & 0 & 0 \\ 0 & g_{r} (\lambda_2) & 0 \\ 0 & 0 & g_{r} (\lambda_3) \end{pmatrix} X^{-1} \quad \text{(49)}$$

and

$$G_t = X \begin{pmatrix} g_{r} (\lambda_1) & 0 & 0 \\ 0 & g_{r} (\lambda_2) & 0 \\ 0 & 0 & g_{r} (\lambda_3) \end{pmatrix} X^{-1} \quad \text{(50)}$$
5. Results of calculations

An absorption of the laser power in the cell filled by the gas of three-level Λ-atoms is proportional to the average excited state population described by expression (18). Typical parameters of the 87Rb atom in a vacuum cell were taken as the parameters of the Λ-atoms in our calculations. The atomic mass m was assumed to be equal to the mass of the 87Rb isotope, the temperature \(T = 20\ ^\circ C\), the ground state relaxation rate \(\Gamma = 300\ \text{s}^{-1}\) and the optical coherence relaxation rate \(\gamma' = 1.8 \times 10^7\ \text{s}^{-1}\). We assume \(V_1 = V_2 = V\) and we set the optical detuning \(\Omega_0 = 0\). Calculations were performed for the cell radius \(R = 0.5\ \text{cm}\) and for three values of the laser beam radius \(r (r = 0.5, 1.5\ \text{and}\ 5\ \text{mm})\) and for five values of the probability \(\alpha\) of the atomic-wall elastic collision (\(\alpha = 0, 0.25, 0.5, 0.75\) and 1).

The structure of the CPT resonance for \(\alpha = 0\) was discussed in [11–14]. Resonance consists of a broad pedestal whose width is about several tens of kilohertz and of a narrow central peak (see figure 5(a)). A broad background is formed by the atoms that have passed through the beam zone only once, whereas the central peak is formed by multiple passes. If the elastic collision probability \(\alpha\) is nonzero, the additional ‘intermediate’ structure in CPT resonance appears. This is a peak whose width is greater than the width of the narrow central peak, but less than the width of the broad background. This peak is formed by atoms that pass the beam zone many times in the beam passing regime. It should be noted that in the elastic collision act the atomic longitudinal velocity \(v_z\) remains constant, in contrast with the case of inelastic scattering when atoms pass the beam zone just once in the beam passing regime. This fact explains the occurrence of ‘intermediate’ structure depending on \(\alpha\) when the cell radius coincides with the radius of the beam (see figures 5(e) and (f)). Indeed, in the considered case of the laser field with narrow spectral linewidth only the atoms with small \(|v_z|\) contribute to the CPT resonance. The ‘intermediate’ structure is formed by the atoms that remain in this velocity group during several passages through the cell volume, whereas the narrow structure is formed by the atoms that change their velocities after one wall collision.

In figures 5(a)–(f), we see that the higher elastic collision probability \(\alpha\) corresponds to a lower excited state population at \(\Omega = 0\). To explain it we should note that for higher \(\alpha\) atoms spent a longer time in the beam passing regime; therefore, they have more time to enter the dark state.

It should be noted that if \(\alpha = 1\), the narrow peak disappears. It can be explained by the fact that all of the atoms contributing to the CPT resonance formation are in the beam passing regime permanently. These atoms spent a relatively longer time in the beam zone than all of the atoms in the case of \(\alpha = 0\). Therefore, the narrow peak is formed by atoms which passed the beam, then spent quite a long time in the dark regime and then returned back to the beam. Intermediate structure is formed by atoms which passed the beam several times repeatedly in the beam passing regime. The time between two subsequent beam passages happens to be smaller than in the case of in elastic collision. Therefore, the resonance happens to be broader. The broad pedestal is formed by atoms that pass the beam zone once during the coherence time.

6. Conclusions

We developed a theory of the coherent population trapping resonance in a cylindrical cell filled with antirelaxation coated walls. The laser beam is assumed to have constant intensity in the cylindrical region. The atom–wall interaction is described by the probability of elastic collisions. It is shown that the nonzero probability of elastic collision leads to distortion of the CPT resonance lines that consists in appearance of the additional peak formed by the atoms, repeatedly passing through the beam zone in the beam passing regime. The general scheme of calculation can be applied not only to CPT formation in wall-coated cells but also for the cell with small buffer gas pressure (much lower than that necessary for the diffusion approximation applicability). Another possible application can be connected with the high-precision clock based on an ensemble of cold atoms or ions, e.g. ions of 229Th [37]. The energy splitting of the 229Th ground-state doublet is about 7.6 eV [38] corresponding to vacuum ultraviolet and long, about an hour, expected half-life of the excited state of the nuclei [39] makes this nuclei a possible promising object for nuclear frequency standard. The study of one-photon resonances in the two-dimensional trap with zonal pumping may be applicable to the spectroscopy of this nucleus.

Acknowledgments

This work was supported by the grant of the Russian Federation President for young candidates of science MK-5318.2010.2, the Federal Program ‘Scientific and scientific-pedagogical personnel of innovative Russia in 2009-2013’, the grant of Russian Foundation for Basic Research 11-02-90426_Ukr_f_a, NASU projects V136, VC139, State Foundation of Fundamental Researches of Ukraine (project F40.2/039) and by the grant of the St. Petersburg Government for young candidates of science. We thank E Breschi and M Hohensee for helpful discussions.

References

[1] Alzetta G, Gozzini A, Moi L and Orriols G 1976 Nuovo Cimento B 36 5–20
[2] Arimondo E and Orriols G 1976 Lett. Nuovo Cimento 17 333–8
[3] Gray H R, Whitley R W and Stroud C R Jr 1978 Opt. Lett. 3 218–20
[4] Harris S E 1997 Phys. Today 50 36–42
[5] Knappe S, Wynands R, Kitching J, Robinson H G and Hollberg L 2001 J. Opt. Soc. Am. B 18 1545–53
[6] Vanier J 2005 Appl. Phys. B 81 421–2
[7] Bergmann K, Theur H and Shore B W 1998 Rev. Mod. Phys. 70 1003–25
[8] Aspect A, Arimondo E, Kaiser R, Vansteenkiste N and Cohen-Tannoudji C 1988 Phys. Rev. Lett. 61 826–9
[9] Nagel A, Graf L, Naumov A, Mariotti E, Biancalana V, Meschede D and Wynands R 1998 Europhys. Lett. 44 31–6
[10] Schwindt P D D, Knappe S, Shah V, Hollberg L, Kitching J, Liew L-A and Moreland J 2004 Appl. Phys. Lett. 85 6409–11
[11] Breschi E, Kazakov G, Schori C, Di Domenico G, Mileti G, Litvinov A and Matisov B 2010 Phys. Rev. A 82 063810
[12] Kazakov G A, Matisov B G and Litvinov A N 2010 Nauchno-Technicheskii Vedomostii SPbGPU 4 11–20 (in Russian)
[13] Klein M, Hohensee M, Phillips D F and Walsworth R L 2011 Phys. Rev. A 83 013826
[14] Hohensee M 2009 Testing fundamental Lorentz symmetries of light PhD Thesis Harvard University
[15] Ye C Y and Zibrov A S 2002 Phys. Rev. A 65 023806
[16] Klein M, Novikova I, Phillips D F and Walsworth R L 2006 J. Mod. Opt. 53 2583–91
[17] Xiao Y 2009 Mod. Phys. Lett. B 23 661–80
[18] Xiao Y, Novikova I, Phillips D F and Walsworth R L 2008 Opt. Express 16 14128–41
[19] Romanenko V I, Romanenko A V and Yatsenko L P 2010 Ukr. J. Phys. 55 393–402
[20] Vanier J and Audoin C 1989 The Quantum Physics of Atomic Frequency Standards (Bristol: Adam Higler) pp 1567
[21] Bouchiat M A and Brossel J 1966 Phys. Rev. 147 41–54
[22] Balykin V I, Letokhov V S, Ovchinnikov Yu B and Sidorov A I 1988 Phys. Rev. Lett. 60 2137–40
[23] Kasevich M A, Weiss D S and Chu S 1990 Opt. Lett. 15 607–9
[24] Vladmirskii V V 1961 Sov. Phys.—JETP 12 740–6
[25] Opat G I, Wark S and Cimmino A 1992 Appl. Phys. B 54 396–402
[26] Hayward T J et al 2010 J. Appl. Phys. 108 043906
[27] Mazets I E and Matisov B G 1992 Sov. Phys.—JETP 74 13–7
[28] Stenholm S 2005 Foundations of Laser Spectroscopy (New York: Dover) pp 268
[29] Shore B W 1990 The Theory of Coherent Atomic Excitation vol 1 (New York: Wiley) pp 774
[30] Dicke R H 1953 Phys. Rev. 89 472–3
[31] Kazakov G, Matisov B, Litvinov A and Mazets I 2007 J. Phys. B: At. Mol. Opt. Phys. 40 3851–60
[32] Litvinov A N, Kazakov G A and Matisov B G 2009 J. Phys. B: At. Mol. Opt. Phys. 42 165402
[33] Jaynes E T 1955 Phys. Rev. 98 1099–105
[34] Frueholz R P and Volk C H 1985 J. Phys. B: At. Mol. Phys. 18 4055–67
[35] Bhaskar N D, Kahla C M and Martin L R 1990 Carbon 28 71–8
[36] Mazets I E and Shifrin L B 2000 Opt. Commun. 175 227–31
[37] Peik E and Tamm Chr 2003 Europhys. Lett. 61 181–6
[38] Beck B R, Becker J A, Beiersdorfer P, Brown G V, Moody K J, Wilhelm J B, Porter F S, Kilbourne C A and Kelly R L 2007 Phys. Rev. Lett. 98 142501
[39] Tkalya E V, Zherebkin A N and Zhudov V I 2000 Phys. Rev. C 61 064380