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Electromagnetically induced transparency in cells of finite size with anti-relaxation wall coating

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In this paper, we propose an approach that makes it possible to obtain an analytical expression of the spectral dependence of the resonance of electromagnetically induced transparency detected by bichromatic radiation in a cell of finite size in the longitudinal direction of the laser beam. Specular and diffuse reflections of atoms from walls are considered. It was found that a significant difference between these types of reflections takes place only for cells of small longitudinal dimensions compared to the splitting wavelength of the ground state of atoms. A physical explanation is proposed for the difference between the Stokes and anti-Stokes scattering channels of probe radiation in terms of dressed states.

Keywords: electromagnetically induced transparency, gas cell, antirelaxation coating, diffuse reflection, specular reflection.

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Introduction

It is well known that the presence of two excitation channels in a quantum system can lead to their destructive interference. This behavior is clearly manifested in the interaction of bichromatic laser radiation with resonant atomic media. Under certain conditions, a narrow dip appears in the absorption line, in which external radiation practically ceases to be absorbed. Depending on the conditions for detecting such a resonance, two related effects are distinguished: coherent population trapping (CPT) [1–4] and electromagnetically induced transparency (EIT) [5–7]. Both phenomena are distinguished by an extremely narrow resonance line, which finds application in a wide class of practical applications: magnetometry [8,9], laser generation without inversion [10,11], recording and processing of quantum information [12–14], quantum frequency standards [15–22], etc. etc.

It is convenient to detect narrow dark resonances using a cloud of atoms cooled to low temperatures in special traps. Such experiments in some cases can demonstrate unique physical effects, such as slowing down and stopping light [23–25]. However, the opportunity of observing resonances at room temperatures using gas cells is of great practical interest. In this case, in the theoretical description of such phenomena, it is essential to take into account not only the motion of atoms, but also their potential collisions with each other and with the walls of the cell. Collisions in most cases are a negative factor, since in their process the atomic coherences excited by the field are destroyed, which cause the opportunity of quantum interference.

In sufficiently rarefied atomic media, collisions of atoms with cell walls come to the fore. One way to suppress depolarization on the walls is to use special antirelaxation coatings. There are a large number of both experimental and theoretical papers [25–65] devoted to studying the properties of such coatings and the process of detecting atomic resonances in coated cells. The determining role in the nature of the reflection of atoms from the antirelaxation surface is played by the time of their adsorption. If it is sufficiently small, the atoms do not have time to make a large number of collisions with the structural particles of the walls, and it can be restricted to the mirror (elastic) reflection model. At the comparatively long adsorption, the velocity of the atom after reflection is determined by the wall temperature. In this case it is said about a diffuse type of reflection [66].

A number of the recent papers have been devoted to a theoretical study of the influence of collisions with the end walls of a cell [67–69] on EIT resonances. These collisions play an important role when wide laser beams are used, when the opportunity of an atom flying out of the pumping field is not taken into account. One of the results of these papers was the observation of a significant difference between the two pumping diagrams that implement stokes and anti-stokes scattering of probe radiation.

In this paper, the approach is proposed which allows to obtain an analytical expression for the spectral dependence of the EIT resonance in a hot cell that is limited in the direction of the laser beam. The analytical result expands the opportunities of analyzing the physical behavior of the systems under consideration in comparison with the results of numerical calculations, which researchers often have to resort to when describing such effects [67–72]. Mirror and diffuse models of wall reflection will be considered. It will be shown that a significant difference between the two

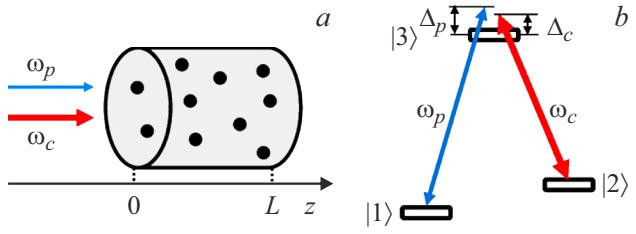

Figure 1. Diagram of mutual orientation of a gas cell and the direction of propagation of external radiation (a) and lambda diagram of excitation of an ensemble of three-level atoms (b).

considered types of reflection takes place only in the case of relatively small longitudinal dimensions of the cell. In addition, based on the analysis of analytical expressions, an alternative physical substantiation of the significant difference between the stokes and anti-stokes scattering channels of probe radiation found in the papers [68,69] based on the Autler-Towns splitting effect will be proposed.

1. Problem statement and approximations

Let us consider a cell filled with pairs of three-level active atoms in the field of a plane two-frequency electromagnetic wave (Fig. 1, a) interacting with the atomic ensemble through the lambda diagram (Fig. 1, b). We write the electric field strength of such a wave in the form:

$$\mathbf{E}(\mathbf{r},t) = \mathbf{E}_p \exp\left[i(k_p z - \omega_p t)\right] + \mathbf{E}_c \exp\left[i(k_c z - \omega_c t)\right] + \text{k.c.}$$

Here $\mathbf{E}_{p,c}, \omega_{p,c}, k_{p,c}$ — complex amplitudes, frequencies, and wave numbers of weak probe and strong binding fields, respectively. Further, we will use the approximation of the medium optical thinness, neglecting the dependences on the coordinates of these amplitudes. Meanwhile, we will not take into account the opportunity of incoherent scattering [73–76].

The quantum state of an atomic ensemble will be described by means of a single-particle Wigner density matrix $\hat{\rho}(\mathbf{r},\mathbf{v},t)$. In doing so, we neglect the collective effects [77–79], i.e., dipole-dipole interaction between atoms, assuming that the medium is sufficiently rarefied. We write the quantum kinetic equation for the density matrix in the Wigner representation in terms of translational degrees of freedom of atoms in the form:

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\boldsymbol{\nabla}\right)\hat{\rho} = -\frac{i}{\hbar}[\hat{H},\hat{\rho}] + \hat{\hat{\Gamma}}\{\hat{\rho}\}, \qquad (1)$$

where $\hat{\hat{\Gamma}}$ — superoperator that phenomenologically takes into account the spontaneous decay of the excited level $|3\rangle$, $\mathbf{v}$ — the rate of translational motion of atoms. Here we do no take into consideration the collisions of atoms with each other, considering the concentration of atoms to be quite small.

Let us expand in (1) the Hamiltonian $\hat{H}$ into the sum of the Hamiltonian of a free atom $\hat{H}_0$ and the interaction operator $\hat{V}$. We write the latter in the dipole approximation in the form:

$$\hat{V} = -\hat{\mathbf{d}}\mathbf{E} = \hbar\Omega_1 \exp\left[-i(\omega_p t - k_p z)\right]|3\rangle\langle 1| - \hbar\Omega_2 \exp\left[-i(\omega_c t - k_c z)\right]|3\rangle\langle 2| + \text{e.c.}, \qquad (2)$$

where $\hat{\mathbf{d}} = \mathbf{e}_d \hat{d}$ — atomic dipole moment vector operator, $\Omega_p = \frac{E_p d_{31}}{\hbar}$, $\Omega_c = \frac{E_c d_{32}}{\hbar}$ — Rabi half frequencies of the probe and binding fields, respectively. This expression is written on the assumption that the vector of the dipole moment is everywhere codirectional with the intensity vectors of both electric fields, and each of them causes transitions from only one of the sublevels of the ground state. The elements of the dipole moment matrix $d_{12} = 0$ due to the fact that the electrodipole transition $|1\rangle \leftrightarrow |2\rangle$ is supposed to be prohibited. Let us note that expression (2) is written taking into account the further use of the rotating wave approximation.

Let us write out the matrix elements (13) and (12) of equation (1) in stationary mode, moving to the slow amplitudes of the density matrix and using the rotating wave approximation:

$$v_z \frac{\partial}{\partial z}\rho_{12} = i\Omega_p^*\rho_{32} - i\Omega_c\rho_{13} + \left[i(\Delta_c - \Delta_p + q v_z) - \Gamma_{12}\right]\rho_{12}, \qquad (3)$$

$$v_z \frac{\partial}{\partial z}\rho_{13} = -i\Omega_p^*\rho_{11} - i\Omega_c^*\rho_{12} + i\Omega_p^*\rho_{33} + \left[-i(\Delta_p - k_P v_z) - \Gamma\right]\rho_{13}, \qquad (4)$$

where $\Delta_p = \omega_p - \omega_{13}$, $\Delta_c = \omega_c - \omega_{23}$ — frequency detunings of fields from atomic transitions $|1\rangle \leftrightarrow |3\rangle$ and $|2\rangle \leftrightarrow |3\rangle$, respectively; $q = k_p - k_c$ — difference of wave numbers of incident waves; Γ, Γ_{12} — decay rates of optical and low-frequency coherences, respectively; v_z — velocity vector projection $\mathbf{v}$ onto axis z. Here, considering the incident wave front to be flat and infinite, and the end walls of the cell parallel to it, we moved on to a one-dimensional problem along the z axis.

Now let us use the strong binding field condition: $\Omega_c \gg \Omega_p$. To do this, we discard the terms of the second order of smallness in Ω_p in expressions (3) and (4). These are the terms containing ρ_{33} and ρ_{32}. Since the entire population is approximately concentrated at the level $|1\rangle$, we write its distribution in the equilibrium form normalized to the cell length: $\rho_{11}(z,v_z) = M(v_z)/L$, where$L$ — cell length, $M(v_z) = (\sqrt{\pi}v_T)^{-1}\exp\left[-\frac{v_z^2}{v_T^2}\right]$ — Maxwell distribution, $v_T = \sqrt{\frac{2k_B T}{m}}$ — most probable velocity, m — ensemble atomic mass, T — ensemble temperature, k_B — Boltzmann constant. In addition, as after a collision with the wall, the optical coherence ρ_{13} will come into equilibrium with the field much faster than the low-frequency ρ_{12} (counting $\Gamma \gg \Gamma_{12}$), the dependencies on the coordinate ρ_{13} can be

neglected, looking for solutions at a sufficient distance from the walls. Thus, we get:

$$v_z \frac{\partial}{\partial z}\rho_{12} = -i\Omega_c\rho_{13} - ip_2\Omega_c^*\rho_{14} + ip_1^*\Omega_p^*\rho_{14} - \delta_{12}\rho_{12},$$

$$0 = -i\Omega_p^* \frac{M}{L} - i\Omega_c^*\rho_{12} - \delta_{13}\rho_{13}. \quad (5)$$

The notation is introduced here:

$$\delta_{12}(v_z) = i(\Delta_p - \Delta_c - qv_z) + \Gamma_{12},$$

$$\delta_{13}(v_z) = i(\Delta_p - k_p v_z) + \Gamma.$$

These values can be called complex decay rates of low-frequency and optical coherence, respectively.

Let us express the optical coherence ρ_{13} from (5):

$$\rho_{13} = \frac{-i}{\delta_{13}}\left(\Omega_p^* \frac{M}{L} + \Omega_c^*\rho_{12}\right). \quad (6)$$

Substituting it into (6), we obtain an ordinary first-order differential equation forρ_{12}::

$$v_z \frac{\partial}{\partial z}\rho_{12} = -\left(\delta_{12} + \frac{|\Omega_c|^2}{\delta_{13}}\right)\rho_{12} - \frac{M}{L}\frac{\Omega_p^*\Omega_c}{\delta_{13}}. \quad (7)$$

We will seek for its general solution in the form:

$$\rho_{12}^+(v,z) = \rho_{12}^{0+}(v)\left\{(W^+(v)+1)\exp\left[-\frac{\lambda^+}{v}z\right] - 1\right\}, \quad (8)$$

$$\rho_{12}^-(v,z) = \rho_{12}^{0-}(v)\left\{(W^-(v)+1)\exp\left[-\frac{\lambda^-}{v}(L-z)\right] - 1\right\}. \quad (9)$$

Here $\rho_{12}^{\pm}(v,z) = \rho_{12}(\pm v, z)$ — partial contributions to the low-frequency coherence from the velocity groups of atoms moving along and against the axis z ($v = |v_z|$);

$$\lambda^{\pm}(v) = \delta_{12}^{\pm}(v) + \frac{|\Omega_c|^2}{\delta_{13}^{\pm}(v)}$$

— the complex rate of establishment of the equilibrium state of a given high-speed group of atoms with a field;

$$\rho_{12}^{0\pm}(v) = \frac{\Omega_p^*\Omega_c}{\lambda^{\pm}(v)\delta_{13}^{\pm}(v)}\frac{M(v)}{L}$$

— distribution of low-frequency coherence over velocities under conditions of equilibrium with the field and the absence of collisions; $W^{\pm}(v)$ — integration constant of equation (7).

Consider the contributions to ρ_{12} of atoms flying off the wall:

$$\rho_{12}^+(v,0) = \rho_{12}^{0+}(v)W^+(v), \quad (10)$$

$$\rho_{12}^-(v,L) = \rho_{12}^{0-}(v)W^-(v). \quad (11)$$

From this it can be seen that the function $W^{\pm}(v)$ has the meaning of a complex indicator of the equilibrium of the low-frequency coherence of a given velocity group of atoms with a field after a collision with a wall. The type of this function is determined by the boundary conditions.

The susceptibility of a unit phase volume of a medium to a probe field is calculated by means of the optical coherence ρ_{13} [80] as

$$\chi = \frac{n_a d_{13}}{\hbar\Omega_p}\rho_{31},$$

where n_a — the concentration of active atoms.

Here is a general formula of susceptibility obtained by means of expressions (6), (8) and (9):

$$\chi^{\pm} = \chi^{0\pm}\left\{1 + \frac{|\Omega_c|^2}{\delta_{13}^{\pm}\delta_{12}^{\pm}}(W^{\pm}+1)\exp\left[\mp\frac{\lambda^{\pm}}{v}\left(z - \frac{L \mp L}{2}\right)\right]\right\}^*. \quad (12)$$

Here

$$\chi^{0\pm} = \frac{in_a d_{13}}{\hbar}\frac{M}{L}\left(\frac{\delta_{12}^{\pm}}{\delta_{12}^{\pm}\delta_{13}^{\pm} + |\Omega_c|^2}\right)^* \quad (13)$$

— the susceptibility of a medium in equilibrium with the field of moving atoms in the absence of collisions with walls, normalized by the length of the cell. The dependence of the imaginary part of expression (12) on the tuning of the test field determines the absorption spectrum of a unit of the phase volume of the medium.

2. Results and discussion

2.1. Infinite medium model

The susceptibility of a medium boundless in all directions is described by the well-known [81] expression (13). The necessity for additional discussion of this case follows from the remark made in the papers [68,69] about the essential difference between the Stokes and anti-Stokes channels of scattering of probe radiation. Let us try to explain this difference using the example of Fig. 2, which shows the absorption spectra of probe radiation by a given velocity group of atoms for both cases: $q < 0$ (a) and $q > 0$ (b). For convenience, the figure also shows the Maxwell distributions over the frequency Doppler shifts of the probe radiation. Calculations were carried out for low temperature $T = 3$ K for reasons of clarity.

Figure 2 shows the shifts of the absorption and transparency resonances as the atomic velocity increases. It can be seen that the shift of the dark resonance qv turns out to be either codirectional with the shift of the absorption resonance $k_p v$, or opposite, depending on the sign q. Meanwhile, an additional peak appears on the left wing of the dark resonance in both cases, which decreases with increasing velocity. The appearance of this peak can be explained in terms of dressed states.

As is well known [82,83], the effect of resonance radiation on a quantum transition leads to the Autler-Towns splitting of the excited state. The frequencies of the obtained quasi-energetic (dressed) states relative to the unsplit excited level

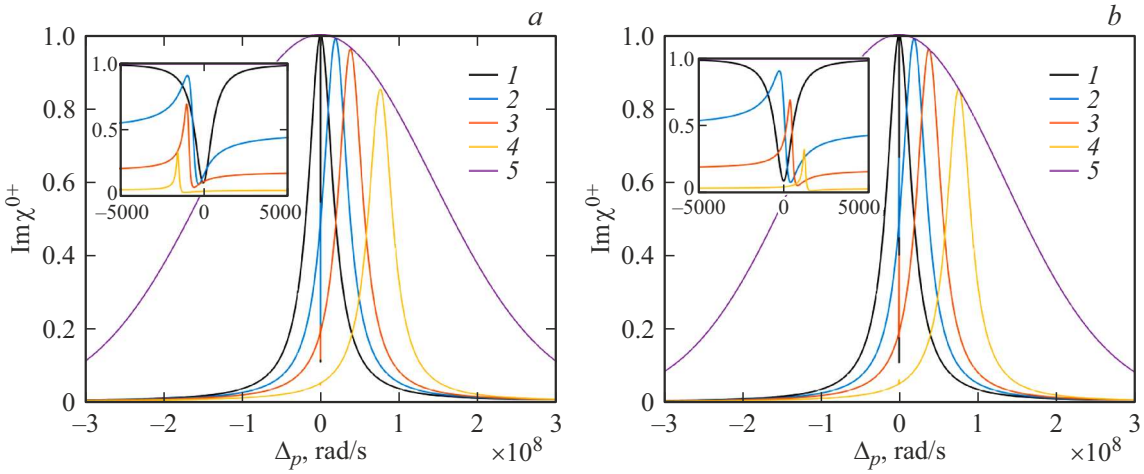

Figure 2. Absorption spectra (curves *1*–*4*) of probe radiation by different velocity groups of atoms against the background of the Maxwell distribution (curve *5*) in relative units for the cases $q < 0$ (*a*) and $q > 0$ (*b*). *1* — $v_z = 0$; *2* — $v_z = 0.1v_T$; *3* — $v_z = 0.2v_T$; *4* — $v_z = 0.4v_T$; *5* — $M(\Delta_p/k_P)/M(0)$. The small inserts on the left side of the figures depict the narrow central pit on a larger scale along the horizontal axis. Other parameters of the calculation: $\Delta_c = 0$, $\Omega_c = 10^5$ rad/s, $\Gamma = 6$ MHz, $\Gamma_{12} = 10$ Hz, $k_p = 79033.27\,\text{cm}^{-1}$, $\lambda_{12} = 4.5$ cm, $|q| = 2\pi/\lambda_{12}$, $T = 3$ K.

are determined by the expression:

$$\omega_\pm = \Delta/2 \pm \sqrt{\Omega^2 + (\Delta/2)^2},$$

where Ω, Δ — half the Rabi frequency and the detuning of the external radiation, respectively.

In our case, the splitting occurs under the action of the binding field, and the detuning at exact resonance is the Doppler shift $-k_c v$. Thus, the Doppler-shifted probe radiation will be in resonance with the states dressed by the binding field at detunings

$$\Delta_p = k_p v - k_c v/2 \pm \sqrt{\Omega_c^2 + (k_c v/2)^2}$$

$$\overset{k_c v \gg \Omega_c}{\approx} \begin{cases} k_p v + \Omega_c^2/k_c v \\ q v - \Omega_c^2/k_c v \end{cases}.$$

This expression determines the positions of the strongly detuned absorption maximum and the narrow peak mentioned above near the dark resonance in Fig. 2. Thus, we can conclude that this peak appears as a result of the detection by probe radiation of one of the states dressed by the binding field.

As can be seen from Fig. 2, in case of $q > 0$ such a peak at velocities close to $\sqrt{\Omega_c^2/k_c q}$ is superimposed on the main dark resonance, which, when averaged over velocities, significantly weakens the transparency effect, in contrast to the case of $q < 0$. Thus, the enhancement of resonant absorption in the case of $q > 0$ occurs due to the interaction of the probe field with one of the dressed states of atoms moving at velocities close to $\sqrt{\Omega_c^2/k_c q}$. Further in the paper, we restrict ourselves to consideration of the pumping diagrams with $q < 0$.

2.2. Mirror and diffuse reflection

To establish the explicit form of the function $W^\pm$, it is essential to set boundary conditions for the expressions (8) and (9). The behavior of active atoms at the cell boundary is determined by the properties of its coating. Next, we consider two well-known [66] limiting cases of such behaviors: mirror and diffuse reflections.

In case of mirror reflection from the wall, the adsorption time of atoms on the surface is relatively short. In this case, we believe that the internal state of the atom does not change, and its velocity in the one-dimensional case changes sign:

$$\rho_{12}^+|_{z=0,L} = \rho_{12}^-|_{z=0,L}.$$

Using expressions (10), (11), we obtain an explicit form of the function $W^\pm$ for the case of mirror reflection:

$$W_{\text{spec}}^\pm = \frac{1 - \exp(-\frac{\lambda^\pm}{v}L)}{1 - \exp(-\frac{\lambda^+ + \lambda^-}{v}L)}\left(1 - \frac{\lambda^\pm \delta_{13}^\pm}{\lambda^\mp \delta_{13}^\mp}\right) - 1.$$

The diffuse type of reflection occurs at relatively long adsorption times of atoms on the surface. In this case, we believe that after a collision with the wall, the atom flies out, perhaps at a different speed, determined by the temperature of the wall, but in the same internal state. Thus, the ensemble state after the collision with the wall is assumed to be thermalized:

$$\rho_{12}^+(v, 0) = \tilde{\rho}_{12}^+(0) M(v),$$

$$\rho_{12}^-(v, L) = \tilde{\rho}_{12}^-(L) M(v), \qquad (14)$$

where

$$\tilde{\rho}_{12}^\pm(z) = 2\int_0^\infty \rho_{12}^\pm(v, z)\, dv,$$

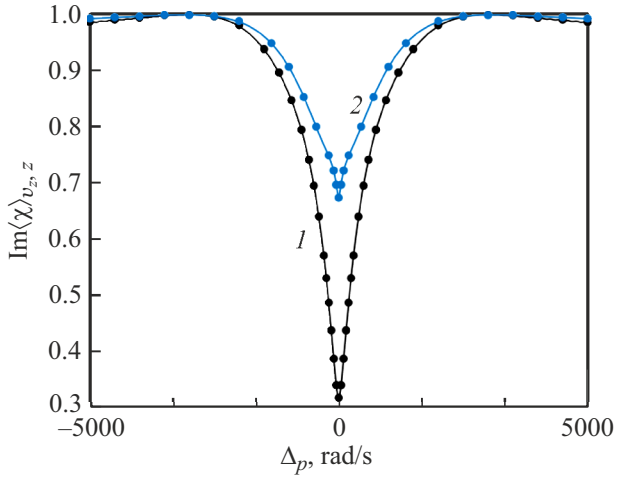

Figure 3. Comparison of numerical (points) and analytical (solid curves) calculations of the spectral dependence of the absorption coefficient in relative units for the case of mirror (curve *1*) and diffuse (curve *2*) reflection of atoms from walls. The calculation was carried out for $L = 0.1\lambda_{12}$ and $T = 50^{\circ}\mathrm{C}$. The other parameters are the same as indicated in the caption to Fig. 2.

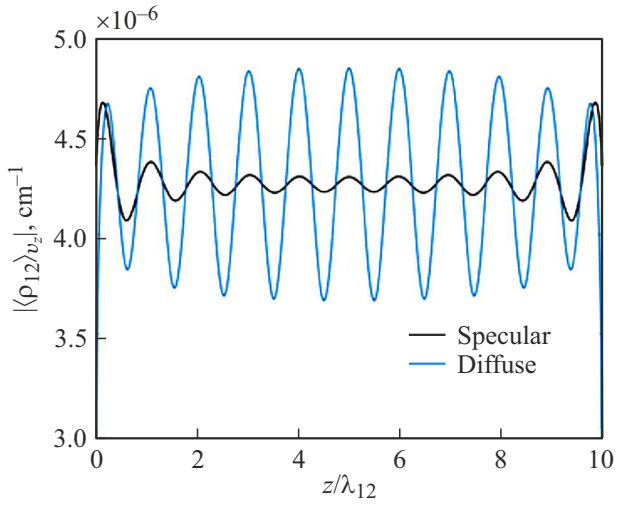

Figure 4. Dependence of the absolute value of the velocity-averaged low-frequency coherence ρ_{12} on the coordinate z inside the cell in units of the wavelength of the splitting of the ground state λ_{12} for the mirror and diffuse nature of the atoms reflection from the walls. The calculation was carried out for temperature $T = 50^{\circ}\mathrm{C}$, detuning $\Delta_p = 0$ and cell length $L = 10\lambda_{12}$. The other parameters are the same as indicated in the caption to Fig. 2.

and as boundary conditions, the equality of the coherence flows flying into the wall and flying away from, is taken:

$$j_{12}^{+}|_{z=0,L} = j_{12}^{-}|_{z=0,L}, \qquad (15)$$

where

$$j_{12}^{\pm} = \int_0^{\infty} \rho_{12}^{\pm}(v, z) v dv.$$

Using (8), (9), in light of (14) and (15), we obtain the expression of the function $W^{\pm}$ for the boundary conditions of diffuse type:

$$W_{\mathrm{diff}}^{\pm} = \lambda^{\pm}\delta_{13}^{\pm}\frac{V^{\pm}U^{\mp} + V^{\mp}}{1 - U^{+}U^{-}},$$

where

$$U^{\pm} = \frac{2\sqrt{\pi}}{v_T}\int_0^{\infty} M(v)\exp\left(-\frac{\lambda^{\pm}}{v}L\right)v dv,$$

$$V^{\pm} = \frac{2\sqrt{\pi}}{v_T}\int_0^{\infty}\frac{1}{\lambda^{\pm}(v)\delta_{13}^{\pm}(v)}\left[\exp\left(-\frac{\lambda^{\pm}}{v}L\right) - 1\right]M(v)v dv.$$

Let us compare the obtained analytical results with the numerical calculation done in [69].

Figure 3 demonstrates good agreement between the obtained analytical result and the numerical solution of the complete system of equations (1). The calculation error is less than 0.1%. Next, we compare the analytical results obtained for the mirror and diffuse reflections of atoms from walls.

Figure 4 demonstrates the quasi-periodic dependence of the absolute value of the low-frequency coherence on the coordinate inside the cell with a period equal to λ_{12}. The resulting oscillations do not decay even at a large distance compared to λ_{12} due to the long lifetime of ρ_{12}. Thus, the transparency resonance will undergo various heterogeneous distortions within the cell. It can be seen that in the case of mirror reflection, this heterogeneity is weaker than for diffuse reflection. Note that the symmetry inherent in Fig. 4 takes place only at zero value Δ_p.

It can be seen from Fig. 5 that, as expected, the width and amplitude of the EIT resonance grow together with the amplitude of the binding field. The maxima appearing on the wings of the transparency resonances are consequences of the contributions to the absorption coefficient from dressed-state resonances, which were discussed in Sec. 2.1. For cells with a length of the order of λ_{12} (Fig. 5, *a*), the forms of resonances for mirror and diffuse reflections are slightly different. This is explained by the fact that the oscillations observed in Fig. 4 are averaged over the cell, and only the average value ρ_{12} contributes to the total absorption coefficient, which is the same for both types of reflection. In cells that are small compared to λ_{12} (Fig. 4, *b*), the difference between diffuse and mirror reflection is much stronger, since the low-frequency coherence does not have time to complete a full period of oscillations during the time of flight of an atom between the walls and cell averaging results in different averages ρ_{12}. In this case, the transparency resonance exhibits a double structure, experiencing additional narrowing in the central region due to the Dicke effect.

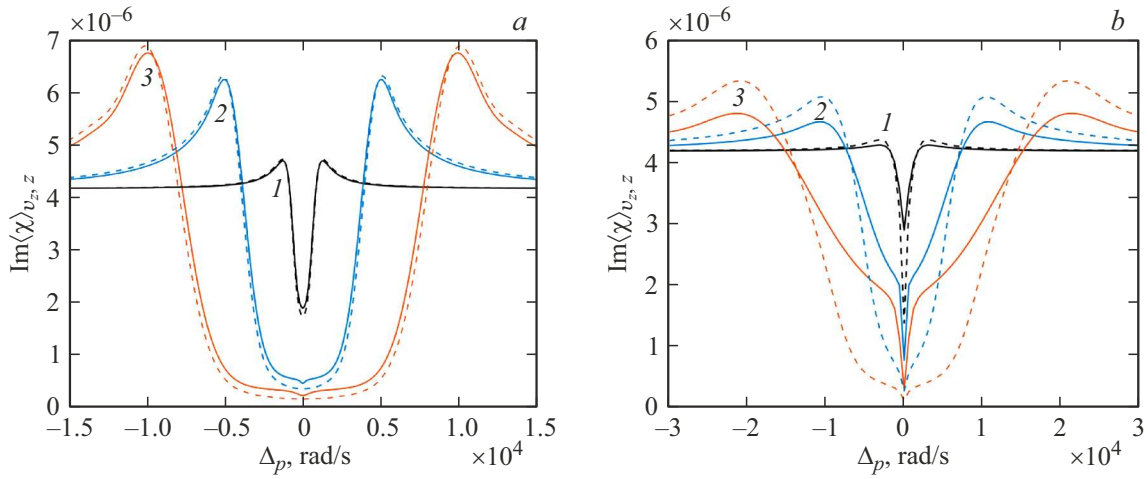

Figure 5. Absorption spectra of probe radiation for the cases of mirror (solid curves) and diffuse (dashed curves) reflections of atoms from walls in arbitrary units for various Ω_c: *1* — 10^5, *2* — $5 \cdot 10^5$, *3* — 10^6 rad/s. The calculation was carried out for temperature $T = 50^\circ$C. Cell lengths: $L = \lambda_{12}$ (*a*) and $L = 0.1\lambda_{12}$ (*b*). The other parameters are the same as indicated in the caption to Fig. 2.

3. Conclusion

In this paper, based on the semiclassical theory of the interaction of light and matter, an approach was proposed that allows to obtain an analytical expression for the shape of the EIT resonance detected in a cell with coatings that are finite in the longitudinal direction of the laser beam and demonstrate the mirror and diffuse nature of atomic reflection. When describing the interaction of two-frequency laser radiation with an atomic medium, a three-level lambda model was used.

It is shown that the obtained analytical expressions agree with the corresponding numerical calculation. Based on the analysis of these expressions, a physical explanation different from that given in the paper [69] was proposed for the difference between the stokes and anti-stokes scattering channels of probe radiation during EIT detection in terms of dressed states. Transparency effects detected in cells with mirror and diffuse reflections of atoms from walls were compared with each other in terms of the form of resonances and in terms of the spatial distribution of low-frequency coherence. It is shown that a significant difference in the form of resonances is observed only for cells with small longitudinal dimensions compared to λ_{12}.

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Conflict of interest

The author declares that he has no conflict of interest.

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