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# Influence of collisions on the effect of electromagnetic-induced transparency in cells of finite sizes with anti-relaxation coating of walls

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Based on the semiclassical theory of the interaction of two-frequency laser radiation with a resonant atomic medium, an analytical expression is obtained that describes the form of resonances of electromagnetically induced transparency detected in a cell with a finite direction in the longitudinal laser beam direction with an anti-relaxation coating of walls, taking into account the motion of atoms and collisions with walls. The following types of reflection from walls are considered: diffuse, specular, and specular-incoherent. It is found that in a number of cases the diffuse type of reflection exhibits the properties of a mirror-incoherent one. It is also shown that the nondegeneracy of the excited state in a number of cases significantly changes the shape of the transparency resonance.

**Keywords:** Electromagnetically induced transparency, Gas cell, anti-relaxation coating, diffuse reflection, specular reflection, specular-incoherent reflection.

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## Introduction

As is known, the interaction of two-frequency laser radiation with a resonant medium under certain conditions can lead to the appearance of a narrow frequency window of transparency [1-4]. In the case when the scanning (test) field is weak compared to the pumping one, they talk about detecting the resonance of electromagnetic-induced transparency (EIT) [5-7]. The widths of such resonances can be made many orders of magnitude smaller than the width of the natural absorption line, which causes interest in such phenomena from numerous practical applications, such as magnetometry [8,9], laser generation without inversion [10,11], recording and processing of quantum information [12-14], quantum frequency standards [15-22], etc. In addition, these phenomena in some cases lead to amazing physical effects, such as extreme light deceleration [23-25], leading to the possibility of controlling light pulses [26,27].

The reason of EIT and other related phenomena, such as coherent population captivity (CPC), is the excitation of quantum coherence in atoms, which leads to destructive interference of two excitation channels. In the case of detecting a transparency resonance in a gas cell, atomic coherence can be destroyed by collisions with walls, which negatively affects the quality of resonances. One of the effective ways to deal with this is to apply a special antirelaxation coating on the walls, which leads to an increase in the time of coherent interaction of the atom with the field and contributes to achieving better resonance quality.

At present, the properties of such coatings are well studied, and the process of detecting atomic resonances

in coated cells has been studied both experimentally and theoretically [25,28–65]. However, some features of the processes of atomic excitation often remain without due attention. In particular, a simple three-level lambda excitation scheme is often used in the theoretical analysis of transparency effects. At the same time, it is well known that taking into account the real atomic structure leads to qualitative features that do not allow description by means of a simple three-level model [68]. So, in the case of CPC, it turns out [69–71] that taking into account already one additional excited state in the theoretical description leads to significant differences from the predictions obtained when describing such phenomena in the framework of a threelevel lambda model.

Several recent papers [72–74] are devoted to the study of the influence of the finite longitudinal dimensions of the cell, taking into account the different patterns of atoms reflection from the walls. Collisions with end walls play an essential role in the detection of resonance by wide laser beams, when the atom does not leave the field of action when moving in the transverse direction of the laser beam [75]. The behavior of atoms in collisions with walls is largely determined by the time of adsorption of atoms on the coating, and the phase shift accompanied concurrently. At the same time, there are several limiting cases that theoretically idealize these processes. Among them, the 3 are considered in this paper:

1. Diffuse reflection, when atoms collide with a surface and are adsorbed on it for a relatively long time, but do not lose their internal state. This type of reflection is typical for paraffin coatings. 2. Mirror reflection, in which the atoms completely retain their internal state, elastically reflecting off the surface. This type is observed in coatings with a short adsorption time.

3. Mirror-incoherent reflection, in which collisions are not accompanied by prolonged adsorption, but undergo a large phase shift. This kind of reflection takes place presumably for some types of silicone surfaces [76].

Due to the complexity of the course of the physical processes under consideration, researchers are often forced to resort to the use of numerical modeling in the theoretical description. In this paper, based on the semi-classical theory of the interaction of light and matter, an approach is proposed that allows us to obtain analytical expressions for the EIT spectra, considering the finite longitudinal dimensions of the cell and the presence of ideal coatings of these types. The obtained expressions were generalized taking into account the spatial limitations of the environment and the presence of an additional energy level. The latter circumstance, as will be shown, in some cases leads to significant changes in the shape of the transparency resonance. The analytical result makes it possible to better understand the physical features of the excitation process of the media under consideration and the behavior of atoms when reflected from the walls. Thus, analyzing this result, it will be shown, that under certain conditions diffuse reflection exhibits the properties of a mirror-incoherent one.

# 1. Problem statement and approximations

Consider a cell filled with a vapor of active atoms in the field of a plane electromagnetic wave having two carrier frequencies  $\omega_p$  and  $\omega_c$  (Fig. 1, *a*). The intensity of the electrical component of such a field will be written as:

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

Using the approximation of the optical fineness of the medium, we neglect the dependence of the complex amplitudes  $\mathbf{E}_p$  and  $\mathbf{E}_c$  on coordinates and time. The interaction of the atomic ensemble with the field will be described semiclassically using the lambda excitation scheme (Fig. 1, *b*). In this case, both the ground and excited states are assumed to be non-degenerate. The task of this paper is to calculate the susceptibility of such an environment to the test field  $\mathbf{E}_p$  under the action of a strong binding field  $\mathbf{E}_c$ .

The state of the atomic ensemble, neglecting the collective effects of [77–79], will be described by the method of a single-particle density matrix in the Wigner representation by translational degrees of freedom of atoms  $\hat{\rho}(\upsilon, \mathbf{r}, t)$ , which satisfies the following quantum kinetic equation:

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

Here  $\hat{\Gamma}$  — is a superoperator that phenomenologically takes into account the spontaneous decay of an excited

state. Collisions of atoms with each other are neglected, considering the concentration of atoms to be quite small.

The Hamiltonian of the system  $\hat{H}$  is represented as the sum of  $\hat{H} = \hat{H}_0 + \hat{V}$ , where  $\hat{H}_0$  — the Hamiltonian ensemble in the absence of an external field,  $\hat{V}$  — the interaction operator with the field. Let us write  $\hat{V}$  in the dipole approximation as follows:

$$\begin{split} \hat{V} &= -\hat{\mathbf{d}}\mathbf{E} = \hbar\Omega_1 \exp[-i(\omega_p t - k_p z)]|3\rangle\langle\langle 1| \\ &- \hbar\Omega_2 \exp[-i(\omega_c t) - k_c z]|3\rangle\langle 2| \\ &- \hbar p_1\Omega_1 \exp[-i(\omega_p t) - k_p z]|4\rangle\langle 1| \\ &- \hbar p_2\Omega_2 \exp[-i(\omega_c t) - k_c z]4\rangle\langle 2| + \text{e.s.}, \end{split}$$

where  $\hat{\mathbf{d}} = \mathbf{e}_d \hat{d}$  — operator of the dipole moment vector of atoms,  $\Omega_p = \frac{E_p d_{31}}{\hbar}$ ,  $\Omega_c = \frac{E_c d_{32}}{\hbar}$  — half the frequencies of Rabi incident fields,  $p_j = \frac{d_{4j}}{d_{3j}}$  — the ratio of matrix elements of the dipole moment operator (j = 1, 2). Here it is assumed that the polarizations of the incident waves are co-directed with the dipole moment vector of the atoms  $(\mathbf{e}_d \cdot \mathbf{e}_{pc} = 1, \mathbf{e}_{pc} = \mathbf{E}_{pc}/E_{pc})$ , and each of the fields 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 prohibited.

Let us write out the matrix elements (13), (14) and (12) of equation (1) in stationary mode, moving to the slow amplitudes of the density matrix and using the rotating wave approximation [69]:

$$\upsilon_{z} \frac{\partial}{\partial z} \rho_{12} = i \Omega_{p}^{*} \rho_{32} - i \Omega_{c} \rho_{13} + i p_{1}^{*} \Omega_{p}^{*} \rho_{42}$$
$$- i p_{2} \Omega_{c} \rho_{14} + \left[ i (\Delta_{c} - \Delta_{p} + q \upsilon_{z}) - \Gamma_{12} \right] \rho_{12}, \qquad (2)$$
$$\upsilon_{z} \frac{\partial}{\partial z} \rho_{13} = -i \Omega_{p}^{*} \rho_{11} - i \Omega_{c}^{*} \rho_{12}$$

$$+ i\Omega_p^* \rho_{33} + \left[ -i(\Delta_p - k_p \upsilon_z) - \Gamma \right] \rho_{13}, \qquad (3)$$

$$\upsilon_{z} \frac{\partial}{\partial z} \rho_{14} = -ip_{1}^{*} \Omega_{p}^{*} \rho_{11} - ip_{2}^{*} \Omega_{c}^{*} \rho_{12} + ip_{1}^{*} \Omega_{p}^{*} \rho_{44}$$
$$+ \left[ -i(\Delta_{p} - \omega_{34} - k_{p}\upsilon) - \Gamma \right] \rho_{14}, \tag{4}$$

where  $\Delta_p = \omega_p - \omega_{13}$ ,  $\Delta_c = \omega_c - \omega_{23}$  — frequency detuning of fields from atomic transitions  $|1\rangle \leftrightarrow |3\rangle$  and  $|2\rangle \leftrightarrow |3\rangle$ respectively;  $\omega_{34}$  — the frequency of splitting of the excited state,  $q = k_p - k_c$  — difference of the wave numbers of incident waves;  $\Gamma$ ,  $\Gamma_{12}$  — the decay rates of optical and low-frequency coherence, respectively;  $\upsilon_z$  — projection of the velocity vector **v** on the 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.

Further, we will use the approximation of the strong binding field  $\Omega_c \gg \Omega_p$ , which approximately allows us to consider the entire population concentrated at the level of  $|1\rangle$ . Due to this, in equations (2)–(4), second-order terms of  $\Omega_p$  can be neglected, i.e. proportional to  $\rho_{33}$ ,  $\rho_{44}$ ,  $\rho_{23}$  and  $\rho_{24}$ , and the element  $\rho_{11}$  is considered constant in coordinate and thermalized in velocity:  $\rho_{11}(\upsilon_z, z) = M(\upsilon_z)/L$ , where  $M(\upsilon_z) = (\sqrt{\pi}\upsilon_T)^{-1} \exp\left[-\frac{\upsilon_z^2}{\upsilon_T^2}\right]$  — Maxwell distribution, L — cell length. Since after a collision with the wall, the optical coherence  $\rho_{1e}$  will come into equilibrium with the field much faster than the low-frequency  $\rho_{12}$  (counting  $\Gamma \gg \Gamma_{12}$ ), one can also neglect the dependencies on the coordinate  $\rho_{1e}$ , e = 3, 4, looking for solutions at a sufficient distance from the walls. Thus, we get

$$\upsilon_{z}\frac{\partial}{\partial z}\rho_{12} = -i\Omega_{c}\rho_{13} - ip_{2}\Omega_{c}^{*}\rho_{14} + ip_{1}^{*}\Omega_{p}^{*}\rho_{14} - \delta_{12}\rho_{12},$$
(5)

$$0 = -i\Omega_{p}^{*}\frac{M}{L} - i\Omega_{c}^{*}\rho_{12} - \delta_{13}\rho_{13}, \qquad (6)$$

$$0 = -ip_1^* \Omega_p^* \frac{M}{L} - ip_2^* \Omega_c^* \rho_{12} - \delta_{14} \rho_{14}.$$
(7)

The notation is introduced here:  $\delta_{12}(\upsilon_z) = i(\Delta_p - \Delta_c - q\upsilon_z) + \Gamma_{12}$ ,  $\delta_{13}(\upsilon_z) = i(\Delta_p - k_p\upsilon_z) + \Gamma$ ,  $\delta_{14}(\upsilon_z) = i(\Delta_p - \omega_{34} - k_p\upsilon_z) + \Gamma$ . These values can be called complex decay rates of low-frequency and optical coherence, respectively. We express the optical coherence of (6) and (7):

$$\rho_{13} = \frac{-i}{\delta_{13}} \left( \Omega_p^* \frac{M}{L} + \Omega_c^* \rho_{12} \right), \tag{8}$$

$$\rho_{14} = \frac{-i}{\delta_{14}} \left( p_1^* \Omega_p^* \frac{M}{L} + p_2^* \Omega_c^* \rho_{12} \right).$$
 (9)

Substituting them into (5), we obtain an ordinary first-order differential equation for  $\rho_{12}$ :

$$\upsilon_{z} \frac{\partial}{\partial z} \rho_{12} = -\left(\delta_{12} + \frac{|\Omega_{c}|^{2}}{\delta_{13}} + \frac{|p_{2}\Omega_{c}|^{2}}{\delta_{14}}\right)\rho_{12}$$
$$-\frac{M}{L}\Omega_{p}^{*}\Omega_{c}\left(\frac{1}{\delta_{13}} + \frac{p_{1}^{*}p_{2}}{\delta_{14}}\right).$$
(10)

We will seek for its general solution in the form

$$\rho_{12}^{+}(\upsilon, z) = \rho_{12}^{0+}(\upsilon) \left\{ (W^{+}(\upsilon) + 1) \exp\left[-\frac{\lambda^{+}}{\upsilon}z\right] - 1 \right\},$$
(11)  
$$\rho_{12}^{-}(\upsilon, z) = \rho_{12}^{0-}(\upsilon) \times \left\{ (W^{-}(\upsilon) + 1) \exp\left[-\frac{\lambda^{-}}{\upsilon}(L - z)\right] - 1 \right\}.$$
(12)

Here  $\rho_{12}^{\pm}(\upsilon, z)$  — partial contributions to lowfrequency coherence from high-velocity groups of atoms moving along and against the axis  $z(\upsilon = |\upsilon_z|)$ ;  $\lambda^{\pm}(\upsilon) = \delta_{12}^{\pm}(\upsilon) + \frac{|\Omega_c|^2}{\delta_{13}^{\pm}(\upsilon)} + \frac{|p_2\Omega_c|^2}{\delta_{14}^{\pm}(\upsilon)}$  — the complex rate of establishing the equilibrium state of a given velocity group of atoms with a field;  $\rho_{12}^{0\pm}(\upsilon) = \frac{\Omega_p^*\Omega_c}{\lambda^{\pm}(\upsilon)\delta_{134}^{\pm}(\upsilon)} \frac{M(\upsilon)}{L}$  —the distribution of low-frequency coherence over velocities under conditions of equilibrium with the field and absence of collisions;  $\delta_{134}^{\pm} = \left(\frac{1}{\delta_{13}^{\pm}} + \frac{p_1^* p_2}{\delta_{14}^{\pm}}\right)^{-1}$ — the complex rate of coherence decay between the level of  $|1\rangle$  and a non-degenerate excited state;  $W^{\pm}(\upsilon)$ — equation integration constant (10).

Consider the contributions to  $\rho_{12}$  of atoms flying off the wall:

$$\rho_{12}^{+}(\upsilon, 0) = \rho_{12}^{0+}(\upsilon)W^{+}(\upsilon), \qquad (13)$$

$$\rho_{12}^+(\upsilon, L) = \rho_{12}^{0-}(\upsilon)W^-(\upsilon). \tag{14}$$

From this it can be seen that the function 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 difference from the unit of this function indicates a loss of equilibrium of low-frequency coherence after a collision. The type of this function is determined by the boundary conditions.

The susceptibility of a unit of the phase volume of the medium to the test field, exclusive of the possible incoherent scattering [80-83], is calculated by means of optical coherence (8) and (9) (see, for instance, [71]) as

$$\chi = \frac{n_a d_{13}}{\hbar \Omega_p} \left( \rho_{13} + \rho_1^* \rho_{41} \right),$$

where  $n_a$  is the concentration of active atoms.

Here is a general formula of susceptibility obtained by means of expressions (8), (9), (11) and (12), for the case  $p_1 = p_2$ :

$$\chi^{\pm} = \chi^{0\pm} \left\{ 1 + \frac{|\Omega_c|^a}{\delta_{134}^{\pm} \delta_{12}^{\pm}} (W^{\pm} + 1) \right.$$
$$\times \exp\left[ \mp \frac{\lambda^{\pm}}{\upsilon} \left( z - \frac{L \mp L}{2} \right) \right] \right\}^*. \tag{15}$$

Here

$$\chi^{0\pm}=rac{in_ad_{13}}{\hbar}rac{M}{L}\left(rac{\delta^{\pm}_{12}}{\delta^{\pm}_{12}\delta^{\pm}_{134}+|\Omega_c|^2}
ight)^{rac{1}{2}}$$

— 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 (15) on the tuning of the test field determines the absorption spectrum of a unit of the phase volume of the medium.

### 2. Results and their discussion

To establish the explicit form of the function  $W^{\pm}$ , it is necessary to set boundary conditions for the expressions (11) and (12). The behavior of active atoms at the cell boundary is determined by the properties of its coating. Further, let us consider some well-known [76] limiting cases of such behaviors.



**Figure 1.** Scheme of mutual orientation of a gas cell and the direction of propagation of external radiation (a) and lambda scheme of excitation of an ensemble of four-level atoms (b).

#### 2.1. Mirror-incoherent reflection

In this case, it is assumed that when colliding with a wall, all atomic populations are preserved, and the coherence is completely destroyed. Boundary conditions of this type have the form

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

As it is easy to see from (13), (14), for the function  $W^{\pm}$  in the case of boundary conditions of mirror-incoherent reflection, we get:  $W_{snq}^{\pm} = 0$ . It is worth noting that this case differs from the boundary conditions of complete quenching considered in [73,74], since due to the approximations made in the previous section, this approach does not allow for mixing between sublevels of the ground state.

#### 2.2. Mirror reflection

With a mirror reflection from the wall, 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 (11), (12), we obtain an explicit form of the function  $W^{\pm}$  for the case of mirror reflection:

$$W_{spec}^{\pm} = \frac{1 - \exp\left(-\frac{\lambda^{\pm}}{v}L\right)}{1 - \exp\left(-\frac{\lambda^{\pm} + \lambda^{-}}{v}L\right)} \left(1 - \frac{\lambda^{\pm} \delta_{134}^{\pm}}{\lambda^{\mp} \delta_{134}^{\mp}}\right) - 1. \quad (16)$$

Let us compare the spectral dependences of the contrast of the transparency resonances calculated using (16) and (15) with the corresponding numerical calculation performed in [71]. The resonance contrast is defined as  $C(\Delta_p) = 1 - \chi''(\Delta_p)/\chi''_0$ , where  $\chi''(\Delta_p)$  and  $\chi''_0$  absorption coefficients at a given detuning and in the absence of transparency resonance, respectively.

Fig. 2 shows a good convergence of the obtained analytical result with the corresponding numerical calculation. Similar comparisons for some of the other cases discussed below, although they have been carried out, will be omitted in this paper.



**Figure 2.** Comparison of numerical (dots) and analytical (solid curve) calculations of the dependences of the contrast of the resonances of transparency on the tuning of the test field. Calculation parameters:  $\Delta_c = 0$ ,  $\Omega_c = 10^5$  rad/s,  $\Gamma = 6$  MHz,  $\Gamma_{12} = 10$  Hz,  $\omega_{34} = 300$  MHz,  $k_p = 79035.27$  cm<sup>-1</sup>,  $\lambda_{12} = 4.5$  cm,  $q = -2\pi/\lambda_{12}$ ,  $L = \lambda_{12}$ ,  $T = 50^{\circ}$  C.



**Figure 3.** Function module dependency  $W_{spec}$  from the tuning of the test field  $\Delta_p$  with the cell length  $L = \lambda_{12}$  on a semilogarithmic scale along the vertical axis for different speeds:  $I - \upsilon_z = \Omega_c^2/q\Gamma$ ;  $2 - \upsilon_z = -\Omega_c^2/q\Gamma$ ;  $3 - \upsilon_z = -\omega_{34}/k_c$ . Hereafter denote  $W(\pm \upsilon) = W^{\pm}(\upsilon)$ . The other parameters are the same as in the caption to Fig. 2.

Fig. 3 shows the dependence of the absolute value of the function  $W_{\text{spec}}$  on the tuning of the test field at different speeds. It is clear that with further averaging of the expression (15) over velocities, the greatest contribution to the main resonance of transparency (near zero detuning) will be made by atoms moving at such speeds that the Doppler shift of the dark resonance does not greatly exceed its width:  $|qv_z| \leq \Omega_c^2/\Gamma$ . Furthermore, as noted in [73], the main contribution to the formation of additional resonances located on the detuning  $\Delta_{pn} = \pi n/k_p L$  is made by the



**Figure 4.** The dependence of the contrast of the transparency resonances on the tuning of the test field for the specular nature of the atoms reflection from the walls at different frequencies of the Rabi binding field:  $I - \Omega_c = 10^6 \text{ rad/s}$ ;  $2 - \Omega_c = 5 \cdot 10^5 \text{ rad/s}$ ;  $3 - \Omega_c = 10^5 \text{ rad/s}$ . All parameters are the same as in the caption to Fig. 2.

velocities at which the Doppler shift of the frequency of the binding field is close to the frequency of splitting of the excited state:  $-k_c v_z = \omega_{34}$ . These two factors determine the choice of speeds in Fig. 3. As can be seen from Fig. 3, near the main and secondary resonances of transparency, the absolute value of the function  $W_{spec}$ becomes significantly different from unity for the velocity groups of atoms that make the greatest contribution to the corresponding resonances. This means that for these constructions, atomic coherence loses its equilibrium in collisions with walls due to the high role of transients. This explains the difference in the shape of resonances in cells with mirror coatings from the corresponding resonances in an infinite medium. Let us present the spectral dependences of the contrast of the transparency resonances at different frequencies of the Rabi binding field (Fig. 4).

It can be seen from Fig. 4 that as the amplitude of the binding field increases, the widths and amplitudes of all resonances increase. At the same time, for sufficiently large resonances begin to overlap, which leads to a significant distortion of their shape. Thus, the presence of an additional excited level not only leads to the appearance of additional peaks, but also affects the shape of the main resonance, moreover, the greater the amplitude of the binding field. Let us analyze the influence of the fourth level on the spatial distribution of low-frequency coherence (Fig. 5).

Figure 5 demonstrates how strongly the presence of the fourth level affects the heterogeneous EIT structure for large values of  $\Omega_c$ . It can be seen here, that in addition to the larger amplitude of spatial oscillations of low-frequency coherence, the four-level atoms also leads to an asymmetric spatial picture of the effect relative to the center of the cell at  $\Delta_p = 0$ . This is explained by the fact that near the wall, the



**Figure 5.** Dependence of the absolute value of the velocityaveraged low-frequency coherence  $\rho_{12}$  on the coordinate z inside the cell in units of the wavelength of the splitting of the ground state  $\lambda_{12}$  for the mirror nature of the atoms reflection from the walls. The solid and dashed curves are calculated on the basis of four- $(p_1 = p_2 = 1)$  and three-  $(p_1 = p_2 = 0)$  level models of the atom, respectively. The calculation was carried out for tuning  $\Delta_p = 0$ , the cell length  $L = 10\lambda_{12}$ , and the frequency of the Rabi binding field  $\Omega_c = 10^6$  rad/s. The other parameters are the same as in the caption to Fig. 2.

contributions of flying and reflected atoms to low-frequency coherence are different, inasmuch as during reflection a transition process to a new equilibrium state occurs. The incoming and reflected atoms experience Doppler shifts in the frequencies of external radiation  $k_{c,p}v_z$ , which have different signs on the two end walls of the cell, which means that they either capture or do not capture the fourth level, depending on the wall.

#### 2.3. Diffuse reflection

This 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:

$$\rho_{12}^{+}(\upsilon, 0) = \tilde{\rho}_{12}^{+}(0)M(\upsilon),$$
  
$$\rho_{12}^{-}(\upsilon, 0) = \tilde{\rho}_{12}^{-}(0)M(\upsilon), \qquad (17)$$

where  $\tilde{\rho}_{12}^{\pm}(z) = 2 \int_0^{\infty} \rho_{12}^{\pm}(v, z) dv$ , and as boundary conditions, the equality of the coherence flows flying into the wall and flying away from, is taken:

$$j_{12}^{+}|_{z=0,L} = j_{12}^{-}|_{z=0,L},$$
(18)

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



**Figure 6.** Dependence of the modulus of the function  $W_{\text{diff}}$  on the length of the cell *L* in units of the wavelength of the splitting of the ground state  $\lambda_{12}$  at a fixed  $\Delta_p = 0$  (*a*), and also from tuning the test field  $\Delta_p$  at a fixed  $L = \lambda_{12}$  on a semi-logarithmic scale along the vertical axis (*b*) for various velocity projections  $v_z$  at a temperature of  $T = 50^{\circ}$ C. The other parameters are the same as in the caption to Fig. 2.



**Figure 7.** The dependence of the contrast of the transparency resonances on the tuning of the test field for a cell of length  $L = 10\lambda_{12}$  and the diffuse nature of the reflection of atoms from the walls at different frequencies of the Rabi binding field:  $I - \Omega_c = 10^6$  rad/s;  $2 - \Omega_c = 5 \cdot 10^5$  rad/s;  $3 - \Omega_c = 10^5$  rad/s. The rest parameters are the same as in the caption to Fig. 2.

Using (11), (12), in light of (17) and (18), we obtain the expression of the function  $W^{\pm}$  for the boundary conditions of diffuse type:

$$W_{\text{diff}}^{\pm} = \lambda^{\pm} \delta_{134}^{\pm} \frac{V^{\pm} U^{\mp} + V^{\mp}}{1 - U^{+} U^{-}},$$

where

$$U^{\pm} = rac{2\sqrt{\pi}}{\upsilon_T} \int\limits_0^\infty M(\upsilon) \exp\left(-rac{\lambda^{\pm}}{\upsilon}L
ight) \upsilon d\upsilon,$$

$$V^{\pm} = \frac{2\sqrt{\pi}}{\upsilon_T} \int_0^{\infty} \frac{1}{\lambda^{\pm}(\upsilon)\delta_{134}^{\pm}(\upsilon)} \times \left[ \exp\left(-\frac{\lambda^{\pm}}{\upsilon}L\right) - 1 \right] M(\upsilon)\upsilon d\upsilon.$$

We present the dependences of the absolute value of this function on the length of the cell and the tuning of the test field for different speeds. The choice of speed groups is made similarly to the previous point.

Figure 6, *a* demonstrates an mixed result. For certain cell lengths close to the integer number  $\lambda_{12}$ , the function  $W_{\text{diff}}$  has minima, becoming much less than one. This, in turn, means that under such conditions, low-frequency coherence is almost completely destroyed when it collides with the walls. Thus, in the cells of these lengths, atoms exhibit the properties of mirror-incoherent reflection, and the shape of the transparency resonance can be described with good accuracy using the result of Section 2.1. From Fig. 6, *b* it can be seen that the above is also true for the side resonance that is on the detuning  $\Delta_p = -\omega_{34}q/k_c$ , where the minimum of the function  $W_{\text{diffalsotakesplace}}$ , which is also preserved for large cells due to the decreasing with *L* nature of the function  $W_{\text{diff}}$ .

We note that the minimum observed for the mirror reflection in Fig. 2 does not lead to similar consequences, since its width is much smaller than the width of the corresponding resonance.

We present in Fig. 7 a dependence similar to that shown in Fig. 4 for a diffuse reflection type with a large cell length  $L = 10\lambda_{12}$ , at which a side resonance becomes noticeable.

Since the main and secondary resonances in this case are separated by a larger amount than in the case of mirror reflection, the effect of overlapping resonances observed in Fig. 4 has a weaker effect. However, the asymmetry of the main resonance increases markedly with the growth of  $\Omega_c$ , which is a consequence of the presence of a side resonance, which means the presence of the fourth level.

# Conclusion

In this paper, we constructed a theory describing the interaction of bichromatic laser radiation with an optically thin and limited in the direction of wave propagation medium of atoms, under conditions of excitation by means of a lambda scheme. At the same time, the non-degeneracy of both the ground and excited states was taken into account. Based on this theory, an analytical expression was obtained for the susceptibility of the medium to weak probe radiation, considering various types of reflection of atoms from the cell walls. The cases of mirror-incoherent, mirror and diffuse reflection patterns were analyzed. It was found that in a number of practically significant cases, diffuse reflection exhibits the properties of mirror-incoherent. It was shown that the presence of an additional fourth level leads not only to the appearance of additional resonances, but also to a distortion of the shape of the main one, the greater the higher the amplitude of the binding field.

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#### **Conflict of interest**

The author declares that he has no conflict of interest.

#### References

- G. Alzetta, A. Gozzini, L. Moi, G. Orriols. Nuovo Cimento. B, 36 (1), 5 (1976). DOI: 10.1007/BF02749417
- [2] E. Arimondo, G. Orriols. Lett. Nuovo Cimento, 17 (10), 333 (1976). DOI: 10.1007/BF02746514
- [3] H.R. Gray, R.M. Whitley, C.R. Stroud. J. Opt. Lett., 3 (6), 218 (1978). DOI: 10.1364/OL.3.000218
- B.D. Agapyev, M.B. Gorny, B.G. Matisov, Yu.V. Rozhdestvenskiy. UFN, 163 (9), 1 (1993). (in Russian). DOI: 10.3367/UFNr.0163.199309a.0001
- [5] S.E. Harris, J.E. Field, A. Imamoglu. Phys. Rev. Lett., 64 (10), 1107 (1990). DOI: 10.1103/PhysRevLett.64.1107
- [6] K.H. Hahn, D.A. King, S.E. Harris. Phys. Rev. Lett., 65 (22), 2777 (1990). DOI: 10.1103/PhysRevLett.65.2777
- [7] K.J. Boiler, A. Imagoglu, S.E. Harris. Phys. Rev. Lett., 66 (20), 2593 (1991). DOI: 10.1103/PhysRevLett.66.2593
- [8] M. Stähler, R. Wynands, S. Knappe, J. Kitching, L. Hollberg,
   A. Taichenachev, V. Yudin. Opt. Lett., 27 (23), 1472 (2002).
   DOI: 10.1364/OL.27.002130

- [9] A. Akulshin, A. Celikov, V. Velichansky. Opt. Commun., 84 (3–4), 139 (1991). DOI: 10.1016/0030-4018(91)90216-Z
- [10] S. Harris. Phys. Rev. Lett., 62 (9), 1033 (1989).
   DOI: 10.1103/PhysRevLett.62.1033
- [11] O.A. Kocharovskaya, Ya.I. Khanin. Pis?ma v ZhETF, 48 (11), 581 (1988). (in Russian).
- [12] M. D. Lukin. Rev. Mod. Phys., 75 (2), 457 (2003).
   DOI: 10.1103/RevModPhys.75.457
- [13] M. Fleischhauer, A. Imamoglu, J.P. Marangos. Rev. Mod. Phys., 77 (2), 633 (2005).
   DOI: 10.1103/RevModPhys.77.633
- [14] R. Zhang, X.B. Wang. Phys. Rev. A, 94 (6), 063856 (2016).
   DOI: 10.1103/PhysRevA.94.063856
- [15] J. Vanier. Appl. Phys. B, 81, 421 (2005).
   DOI: 10.1007/s00340-005-1905-3
- [16] S.A. Zibrov, V.L. Velichansky, A.S. Zibrov, A.V. Taichenachev,
   V.I. Yudin. JETP Lett., 82 (8), 477 (2005). DOI: 10.1134/1.2150865.
- [17] G. Kazakov, B. Matisov, A. Litvinov, I. Mazets. J. Phys. B: At. Mol. Opt. Phys., 40 (19), 3851 (2007). DOI: 10.1088/0953-4075/40/19/006
- [18] S.A. Zibrov, I. Novikova, I. Phillips, D. F. Walsworth, R.L. Zibrov, A. S. V.L. Velichansky, A.V. Taichenachev, V.I. Yudin. Phys. Rev. A, 81 (1), 013833 (2010). DOI: 10.1103/PhysRevA.81.013833
- [19] K.A. Barantsev, E.N. Popov, A.N. Litvinov, V.M. Petrov. Radiotekhnika, 12, 164 (2016). (in Russian).
- [20] S. Khripunov, D. Radnatarov, S. Kobtsev. Proc. SPIE, 9378, 93780A (2015). DOI: 10.1117/12.2080165
- [21] J. Kitching. Appl. Phys. Rev., 5 (3), 031302 (2018).DOI: 10.1063/1.5026238
- S. Kobtsev, S. Donchenko, S. Khripunov, D. Radnatarov, I. Blinov, V. Palchikov. Opt. Laser Technol., 119, 105634 (2019). DOI: 10.1016/j.optlastec.2019.105634
- [23] L.V. Han, S.E. Harris, Z. Dutton, C.H. Behroozi. Nature, 397, 594 (1999). DOI: 10.1038/17561
- [24] N.A. Vasiliev, A.S. Troshin. ZhETF, 125 (6), 1276 (2004). (in Russian).
- [25] I. Novikova, R.L. Walsworth, Y. Xiao. Laser Photonics Rev.,
   6 (3), 333 (2012). DOI: 10.1002/lpor.201100021
- [26] N.A. Vasiliev, A.S. Troshin. Izvestiya RAN. Seriya fizicheskaya, 69 (8), 1096 (2005). (in Russian).
- [27] A.S. Losev, A.S. Troshin. Opt. i spektr., 110 (1), 76 (2011).
   (in Russian). DOI: 10.1134/S0030400X11010127
- [28] D. Budker, V. Yashchuk, M. Zolotorev. Phys. Rev. Lett., 81 (26), 5788 (1998). DOI: 10.1103/PhysRevLett.81.5788
- [29] D. Budker, L. Hollberg, D.F. Kimball, J. Kitching, S. Pustelny, V.V. Yashchuk. Phys. Rev. A, 71 (1), 012903 (2005).
- [30] M.T. Graf, D.F. Kimball, S.M. Rochester, K. Kerner, C. Wong,
   D. Budker, E.B. Alexandrov, M.V. Balabas, V.V. Yashchuk.
   Phys. Rev. A, **72** (2), 023401 (2005).
   DOI: 10.1103/PhysRevA.72.023401
- [31] D. Budker, M. Romalis. Nature Phys., 3, 227 (2007). DOI: 10.1038/nphys566
- [32] E.B. Alexandrov, A.K. Vershovsky. UFN, **179**, 605 (2009). (in Russian). DOI: 10.3367/UFNe.0179.200906f.0605
- [33] M.V. Balabas, T. Karaulanov, M.P. Ledbetter, D. Budker. Phys. Rev. Lett., **105** (7), 070801 (2010).
   DOI: 10.1103/PhysRevLett.105.070801
- [34] M.V. Balabas, K. Jensen, W. Wasilewski, H. Krauter, L.S. Madsen, J.H. Muller, T. Fernholz, E.S. Polzik. Opt. Express, 18 (6), 5825 (2010). DOI: 10.1364/OE.18.005825

- [35] E. Breschi, G. Kazakov, C. Schori, G. Di Domenico, G. Mileti,
   A. Litvinov, B. Matisov. Phys. Rev. A, 82 (6), 063810 (2010).
   DOI: 10.1103/PhysRevA.82.063810
- [36] K. Nasyrov, S. Gozzini, A. Lucchesini, C. Marinelli, S. Gateva,
   S. Cartaleva, L. Marmugi. Phys. Rev. A, 92 (4), 043803 (2015). DOI: 10.1103/PhysRevA.92.043803
- [37] M.A. Hafiz, V. Maurice, R. Chutani1, N. Passilly, C. Gorecki,
   S. Guerande, E. de Clercq, R. Boudot. J. Appl. Phys., 117 (18), 184901 (2015). DOI: 10.1063/1.4919841
- [38] H. Chi, W. Quan, J. Zhang, L. Zhao, J. Fang. Appl. Surf. Sci., 501, 143897 (2020). DOI: 10.1016/j.apsusc.2019.143897
- [39] S.J. Seltzera, M.V. Romalis. J. Appl. Phys., 106 (11), 114905 (2009). DOI: 10.1063/1.3236649
- [40] K.A. Barantsev, S.V. Bozhokin, A.S. Kuraptsev, A.N. Litvinov,
   I.M. Sokolov. JOSA B, 38 (5), 1613 (2021).
   DOI: 0.1364/JOSAB.412513
- [41] A. Krasteva, R.K. Nasyrov, N. Petrov, S. Gateva, S. Cartaleva, K.A. Nasyrov. Optoelectron. Instrument. Proc., 54 (3), 307 (2018).
- [42] W. Li, M. Balabas, X. Peng, S. Pustelny, A. Wickenbrock,
   H. Guo, D. Budker. J. Appl. Phys., **121** (6), 063104 (2017).
   DOI: 10.1063/1.4976017
- [43] G.A. Kazakov, A.N. Litvinov, B.G. Matisov, V.I. Romanenko,
   L.P. Yatsenko, A.V. Romanenko. J. Phys. B, 44 (23), 235401 (2011). DOI: 10.1088/0953-4075/44/23/235401
- [44] M. Klein, M. Hohensee, D.F. Phillips, R.L. Walsworth. Phys. Rev. A, 83 (1), 013826 (2011).
  - DOI: 10.1103/PhysRevA.83.013826
- [45] A. Litvinov, G. Kazakov, B. Matisov, I. Mazets. J. Phys. B: At. Mol. Opt. Phys., 41 (12), 125401 (2008).
   DOI: 10.1088/0953-4075/41/12/125401
- [46] S. Knappe, H.G. Robinson. New J. Phys., 12 (6), 1–9 (2010).
   DOI: 10.1088/1367-2630/12/6/065021
- [47] E.N. Pestov, A.N. Besedina, D.E. Pestov, V.V. Semenov. Appl.
   Magn. Reson., 51, 195 (2020). DOI: 10.1007/s00723-019-01186-w
- [48] S.J. Seltzer, M.V. Romalis. J. Appl. Phys., 106 (11), 114905 (2009). DOI: 10.1063/1.3236649
- [49] S.N. Atutov, A. I. Plekhanov, V.A. Sorokin, S.N. Bagayev, M.N. Skvortsov, A.V. Taichenachev. Eur. Phys. J. D, 72, 155 (2018). DOI: 10.1140/epjd/e2018-90127-6
- [50] S.N. Atutov, V.A. Sorokin, S.N. Bagayev, M.N. Skvortsov,
   A.V. Taichenachev. Eur. Phys. J. D, 73, 240 (2019).
   DOI: 10.1140/epjd/e2019-100206-5
- [51] M. Bhattarai, V. Bharti, V. Natarajan, A. Sargsyan, D. Sarkisyan. Phys. Lett. A, **383** (1), 91 (2019).
   DOI: 10.1016/j.physleta.2018.09.036
- [52] S. Kobtsev, D. Radnatarov, S. Khripunov, I. Popkov,
   V. Andryushkov, T. Steshchenko. J. Opt. Soc. Am. B, 36 (10),
   2700 (2019). DOI: 10.1364/JOSAB.36.002700
- [53] A. Krasteva, E. Mariotti, Y. Dancheva, C. Marinelli, L. Marmugi, L. Stiaccini, S. Gozzini, S. Gateva, S. Cartaleva. J. Contemporary Physics (Armenian Academy of Sciences), 55 (4), 383 (2020).
  - DOI: 10.3103/S1068337220040209
- [54] H. Chi, W. Quan, J. Zhang, L. Zhao, J. Fang. Appl. Surf. Sci., 501 (31), 143897 (2020).
   DOI: 10.1016/j.apsusc.2019.143897
- [55] M. Bhattarai, V. Bharti, V. Natarajan. Scientific Reports, 8 (1), 7525 (2018). DOI: 10.1038/s41598-018-25832-8

- [56] S. Kobtsev, D. Radnatarov, S. Khripunov, I. Popkov, V. Andryushkov, T. Steschenko. Proc. SPIE 10548 (SPIE OPTO, 2015), 93780A (2018). DOI: 10.1117/12.2080165
- [57] Y. Ji, J. Shang, Q. Gan, L. Wu. 2017 IEEE 67th Electronic Components and Technology Conference (ECTC, 2017), p. 2116. DOI: 10.1109/ECTC.2017.136
- [58] Sekiguchi N., Hatakeyama A. Appl. Phys. B: Lasers and Optics, **122** (4), 81 (2016). DOI: 10.1007/s00340-016-6352-9
- [59] O. Yu. Tretiak, J.W. Blanchard, D. Budker, P.K. Olshin, S.N. Smirnov, M. Balabas. J. Chem. Phys., **144** (9), 094707 (2016). DOI: 10.1063/1.4943123
- [60] M. Pellaton, C. Affolderbach, G. Mileti, R. Straessle, Y. Petremand, D. Briand, N.F. De Rooij. European Frequency and Time Forum (EFTF, 2014), p. 554. DOI: 10.1109/EFTF.2014.7331561
- [61] G. Zhang, L. Wei, M. Wang, K. Zhao. J. Appl. Phys., 117 (4), 043106 (2015). DOI: 10.1063/1.4906851
- [62] R. Straessle, M. Pellaton, C. Affolderbach, Y. Pétremand,
   D. Briand, G. Mileti, N.F. De Rooij. Appl. Phys. Lett., 105 (4), 043502 (2014). DOI: 10.1063/1.4891248
- [63] Z. Chowdhuri, M. Fertl, M. Horras, K. Kirch, J. Krempel, B. Lauss, A. Mtchedlishvili, D. Rebreyend, S. Roccia, P. Schmidt-Wellenburg, G. Zsigmond. Appl. Phys. B: Lasers and Optics, **115** (2), 257 (2014). DOI: 10.1007/s00340-013-5598-8
- [64] R. Straessle1, M. Pellaton, C. Affolderbach, Y. Pétremand1,
   D. Briand, G. Mileti, N. F. de Rooij. J. Appl. Phys., 113 (6),
   064501 (2013). DOI: 10.1063/1.4789942
- [65] T. Bandi, C. Affolderbach, G. Mileti. J. Appl. Phys., 111 (12), 124906 (2012). DOI: 10.1063/1.4789942
- [66] M. Hasegawa, P. Dziuban, L. Nieradko, A. Douahi, C. Gorecki, V. Giordano. IEEE/LEOS International Conference on Optical MEMs and Nanophotonics (Freiburg, Germany, 2008), p. 162. DOI: 10.1109/OMEMS.2008.4607879
- [67] K.A. Nasyrov. Avtometriya, 52 (1), 85 (2016). (in Russian).
- [68] D. V. Kupriyanov, I. M. Sokolov, M. D. Havey. Optics Commun., 243, 165 (2004).
- [69] G.V. Voloshin, K.A. Barantsev, E.N. Popov, A.N. Litvinov. ZhETF, 156 (1), 5 (2019). (in Russian).
   DOI: 10.1134/S0044451019070010
- [70] G.V. Voloshin, K.A. Barantsev, A.N. Litvinov. Kvant. elektron., 50 (11), 1023 (2020). (in Russian). DOI: 10.1070/QEL17064
- [71] G.V. Voloshin, K.A. Barantsev, A.N. Litvinov. Kvant. elektron.,52 (2), 108 (2022). (in Russian). DOI: 10.1070/QEL17976
- [72] A.N. Litvinov, I.M. Sokolov. Pis?ma v ZhETF, 113 (12), 791 (2021). (in Russian). DOI: 10.31857/S1234567821120041
- [73] G.V. Voloshin, Hui Meng, A.S. Kuraptsev, I.M. Sokolov. ZhETF, 162 (3), 313 (2022). (in Russian).
   DOI: 10.31857/S0044451022090036
- [74] Ya.A.Fofanov, I.M. Sokolov. ZhETF, 162 (3), 297 (2022).(in Russian). DOI: 10.31857/S0044451022090012
- [75] S. N. Nikolic, A. J. Krmpot, N. M. Lu, B. V. Zlatkovi, M. Radonji, B. M. Jelenkovic. Phys. Scr., 157, 014019 (2013).
- [76] B. D. Agapyev, M. B. Gorny, B. G. Matissov. ZhTF 58, 12 (1988). (in Russian).
- [77] Ya.A. Fofanov, A.S. Kuraptsev, I.M. Sokolov, M.D. Havey.
   Phys. Rev. A, 84 (5), 053811 (2011).
   DOI: 10.1103/PhysRevA.84.053811
- [78] A.S. Kuraptsev, I.M. Sokolov. Phys. Rev. A, 90 (1), 012511 (2014). DOI: 10.1103/PhysRevA.90.012511

- [79] I.M. Sokolov, D.V. Kupriyanov, M.D. Hevy. ZhTF, 139 (2), 288 (2011). (in Russian). DOI: 10.1134/S106377611101016X
- [80] D.V. Kupriyanov, I.M. Sokolov, N.V. Larionov, P. Kulatunga,
   C.I. Sukenik, S. Balik, M.D. Havey. Phys. Rev. A, 69 (3),
   033801 (2004). DOI: 10.1103/PhysRevA.69.033801
- [81] V. M. Datsyuk, I.M. Sokolov, D.V. Kupriyanov, M.D. Havey. Phys. Rev. A, 74 (4), 043812 (2006). DOI: 10.1103/PhysRevA.74.043812
- [82] V.M. Datsyuk, I.M. Sokolov, D.V. Kupriyanov, M.D. Havey. Phys. Rev. A, 77 (3), 033823 (2008).
   DOI: 10.1103/PhysRevA.77.033823
- [83] A.S. Kuraptsev, I.M. Sokolov, M.D. Havey. Phys. Rev. A, 96 (2), 023830 (2017). DOI: 10.1103/PhysRevA.96.023830