

Rigorous theory of thin-vapor-layer linear optical properties: The case of specular reflection of atoms colliding with the walls

A. V. Ermolaev  and T. A. Vartanyan 

ITMO University, Kronversky Prospekt 49, 197101 St. Petersburg, Russia



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The theory of the thin-vapor-layer linear optical properties is presented for the case of specular reflection of atoms colliding with the walls. The effects of light absorption and the shift in the resonance frequency are taken into account by means of self-consistent calculation of the field and polarization in a gaseous medium. The obtained formulas reproduce the complex dependence of the spectral line profile on the gas layer thickness and allow one to determine numerically the exact values of the “blueshift.” It was shown that, despite the low vapor concentration, the resonance shift is on the same order of magnitude as the width of the spectral lines.

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I. INTRODUCTION

The possibility of observing sub-Doppler resonances in the spectra of light reflection from the interface between a transparent dielectric medium and the atomic vapor makes selective reflection spectroscopy a promising area for theoretical and experimental studies. Doppler-free structures in the line shape of selective reflection were first observed by Cojan [1]. In this paper, the narrowing of the spectral lines in reflection was ascribed to the transient polarization of the atoms departing from the cell wall. In the limit of sufficiently high vapor densities, particle collisions lead to a rapid loss of the oscillation phase. Consequently, polarization at a given spatial location is defined solely by the field in the same location, the condition commonly referred to as local response of the polarization on the field. Contrary to that, in the case of rarefied vapor polarization at a given location is defined by the field at the neighboring location as well. The effects of this nonlocal response, or in other words, spatial dispersion effects become especially important when the mean free path of atoms without loss of polarization becomes greater than the wavelength of the incident light [2].

Recently, it was demonstrated [2,3] that the nonlocal optical response of a resonant gas can be significantly enhanced if the vapor is spatially confined in a cell with a thickness on the order of the incident wavelength λ . The invention of a gas cell with a subwavelength thickness [4] enabled experimenters to examine optical properties of a thin-vapor layer as well as the interaction of atoms with the surface of a dielectric material [5,6]. These studies may become the basis for the creation of miniature atomic standards of frequency and time [7]. From this point of view, it is important to correctly consider factors leading to a shift and broadening of the spectral lines of selective reflection. In this article, we will focus on one of the shifts of a purely electrodynamics nature that arises due to the interference between the contributions of the departing and arriving atoms (hereinafter referred to as the “blueshift”) [8,9].

In Refs. [3,10], the approximate solution for the reflected and transmitted fields was obtained by means of perturbation

expansion of the field with respect to the atomic vapor density for the case of diffuse boundary conditions, which imply that the atom loses polarization upon collision with the wall. In fact, in order to account for the structure of the field inside the vapor exactly, the field and polarization should be calculated in a self-consistent way [11]. In the absence of a rigorous theory, in many works, the experimental results are compared with the calculations performed in the framework of perturbation theory in optical density, which can lead to substantial errors. The aim of this paper is to investigate the linear optical properties of thin-vapor layers beyond the scope of perturbation theory. We will assume throughout this paper that atoms collide with the walls specularly, i.e., the atoms preserve their polarization, and only the normal component of their velocity changes sign after a collision with the wall. Being well aware of experimental studies in which atomic excitation quenching processes predominate in collisions of atoms with dielectric surfaces [12], we would like to emphasize that, to some extent, the situation considered in this paper can be experimentally realized by applying antirelaxation coatings on the windows of the gas cell [4,13,14]. Moreover, we believe that the rigorous solution of the problem of a thin gas layer obtained in our paper for the case of elastic scattering of atoms on the walls is an important step in developing of the comprehensive theoretical model that will include diffuse and quenching collisions as well.

The paper is organized as follows. Section II describes a theoretical model of resonant reflection of light from a thin gas layer based on the Maxwell-Bloch equations. In Sec. III, the rigorous solution of the thin gas layer problem is derived for the case of specular boundary conditions by means of Fourier series expansion of the field. A numerical calculation of the reflectivity of a thin gas layer at various thicknesses and vapor densities, the comparison of the Fourier method with the result of perturbation theory, as well as the dependence of the blueshift on the atomic number density are presented in Sec. IV (the perturbation theory approach is described in the Appendix). Finally, in Sec. V, we discuss some peculiarities connected with the specular reflection of atoms colliding

with the walls and the general significance of the considered problem for the case of diffuse and quenching collisions of atoms with the walls.

II. THEORETICAL MODEL

Consider a vapor layer of thickness l , located between two transparent dielectric media with parallel interfaces. The described system is schematically shown in Fig. 1 where a monochromatic electromagnetic plane wave propagates in the positive direction of the x axis in the first dielectric medium with refractive index n_1 . The wave (at normal incidence) partially reflects from the gas layer and penetrates into the second dielectric medium with refractive index n_2 .

Neglecting nonlinear optical processes, the electric field and polarization inside the vapor can be represented as

$$E(x, t) = \frac{1}{2}E(x) \exp(-i\omega t) + \text{c.c.}, \quad (1a)$$

$$P(x, t) = \frac{1}{2}P(x) \exp(-i\omega t) + \text{c.c.} \quad (1b)$$

From Maxwell's equations, it can be shown that the spatial parts of the electric field and polarization of the medium satisfy the equation,

$$\frac{d^2 E(x)}{dx^2} + k^2 E(x) = -4\pi k^2 P(x), \quad (2)$$

where $k = \omega/c$.

It is assumed that the gas consists of two-level atoms and the frequency of the incident light wave ω is varied in the vicinity of the atomic transition frequency ω_0 . Then, the macroscopic polarization can be expressed in terms of the off-diagonal density-matrix element $\rho(x, v, t) = \rho(x, v) \exp(-i\omega t)$ by averaging the dipole moment operator over the velocity distribution of atoms $f(v)$,

$$P(x) = 2ND \int_{-\infty}^{+\infty} dv f(v) \rho(x, v), \quad (3)$$

where N is the number density of the gas atoms, D is the transition dipole moment, and v is the projection of the atomic velocity on the x axis. Throughout this article, we will assume that the thermal motion of the gas layer atoms is described by

Maxwell's distribution function,

$$f(v) = \frac{1}{\sqrt{\pi} v_T} \exp\left(-\frac{v^2}{v_T^2}\right), \quad (4)$$

where v_T is the most probable thermal velocity.

In order to study the linear optical properties of the described system, we restrict ourselves to the case of small electric-field strengths when the intensity of the light is so low that it cannot saturate the resonant transition. Given that the total time derivative of an off-diagonal element of a two-level atom with a nonzero velocity component v should be defined as $d\rho(x, v, t)/dt = [\partial/\partial t + v(\partial/\partial x)]\rho(x, v) \exp(-i\omega t)$, according to the optical Bloch equations in the resonance region $\omega \gg |\omega_0 - \omega|$, $\rho(x, v)$ satisfies the steady-state equation,

$$v \frac{\partial \rho(x, v)}{\partial x} + [\gamma + i(\omega_0 - \omega)]\rho(x, v) = \frac{i}{2\hbar} DE(x), \quad (5)$$

where γ is the homogenous width of the transition that is the sum of natural and collisional widths of the spectral line. After substituting the expression for polarization [see Eq. (3)] in Eq. (2), we can rewrite the whole set of Eqs. (2)–(5) in the following way:

$$\frac{d^2 E(\xi)}{d\xi^2} + E(\xi) = -2im \int_{-\infty}^{+\infty} \sigma(\xi, v) \exp(-v^2) dv, \quad (6a)$$

$$v \frac{\partial \sigma(\xi, v)}{\partial \xi} + \eta \sigma(\xi, v) = E(\xi). \quad (6b)$$

Here, we have introduced the dimensionless variables $\xi = kx$, $m = 2\sqrt{\pi}ND^2/\hbar k v_T$, $v = v/v_T$, $\sigma(\xi, v) = (2\hbar k v_T/iD)\rho(x, v)$, $\eta = \Gamma - i\Omega$, $\Gamma = \gamma/kv_T$, and $\Omega = (\omega - \omega_0)/kv_T$, where parameter m is proportional to the atomic vapor density.

In accordance with the statement of the problem [see Fig. 1], the electric-field strength of the incident, reflected, and transmitted waves may be written in the following forms [15]:

$$E_0 \exp[in_1 \xi - i\omega t], \quad (7a)$$

$$E_r \exp[-in_1 \xi - i\omega t], \quad (7b)$$

and

$$E_t \exp[in_2(\xi - \phi) - i\omega t], \quad (7c)$$

respectively. From the condition of continuity of the field and its first derivative at the boundaries, we can find

$$E_0 + E_r = E(0), \quad (8a)$$

$$in_1(E_0 - E_r) = E'(0), \quad (8b)$$

$$E_t = E(\phi), \quad (8c)$$

$$in_2 E_t = E'(\phi), \quad (8d)$$

where $\phi = kl$ and $(')$ stands for the derivative with respect to ξ . Introducing the amplitude reflection coefficient $r = E_r/E_0$, we can represent Eqs. (8a) and (8b) as

$$\frac{1-r}{1+r} = \frac{M(0)}{n_1}, \quad (9)$$

where $M(0) = E'(0)/iE(0)$ is the surface admittance of the first boundary of a gas layer. Consequently, reflectivity may

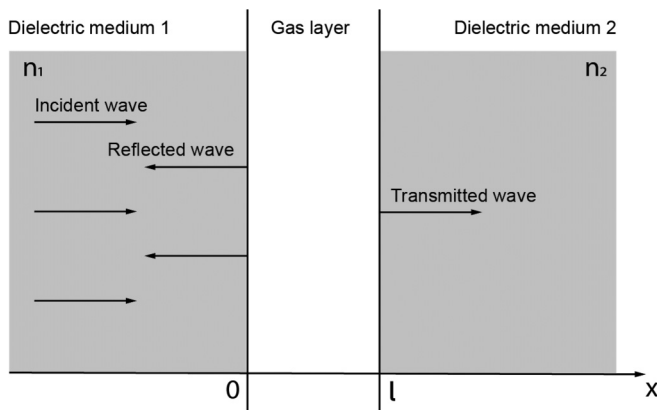


FIG. 1. Schematic of light reflection (at normal incidence) from a thin gas layer.

be expressed in terms of the surface admittance,

$$R = |r|^2 = \left| \frac{n_1 - M(0)}{n_1 + M(0)} \right|^2. \quad (10)$$

According to Eq. (10), in order to study the spectral line profile in reflection, the set of Eqs. (6) needs to be solved with respect to the surface admittance.

III. FOURIER METHOD

In this section, following the method described in Ref. [16], the exact solution of Eqs. (6a) and (6b) is obtained for the case of specular boundary conditions by means of Fourier series expansion of the field. According to the specular boundary conditions, after a collision with the surface of a dielectric material, an atom conserves its polarization and changes the sign of the velocity component normal to a given surface. Thus, we can set

$$\sigma(\xi = 0, \nu) = \sigma(\xi = 0, -\nu), \quad (11a)$$

$$\sigma(\xi = \phi, -\nu) = \sigma(\xi = \phi, \nu) \quad (11b)$$

for the atoms at the front and rear boundaries. Now, in order to determine the field inside the vapor, let us imagine that there are no boundaries at planes $\xi = 0$ and $\xi = \phi$. If we reflect the gas layer with respect to the plane $\xi = 0$ and repeat the resulting layer with the thickness of 2ϕ throughout the space, the field will satisfy the following condition:

$$E(j\phi - \xi) = E(j\phi + \xi), \quad (12)$$

where j is an integer. In this case, the atom reflected from the wall in the layer problem corresponds to an atom freely crossing the interface from the side of the adjacent layer in the infinite space problem.

It is clear from Eqs. (11a), (11b), and (12) that the field and the off-diagonal element of the density matrix can be expanded in the Fourier series as continuous periodic functions with a period of 2ϕ in the form

$$F(\xi) = \sum_{n=-\infty}^{\infty} F(n) \exp(in\pi\xi/\phi), \quad (13)$$

where $F(n) = (2\phi)^{-1} \int_{-\phi}^{\phi} F(\xi) \exp(-in\pi\xi/\phi) d\xi$. After we multiply Eqs. (6a) and (6b) by $(2\phi)^{-1} \exp(-in\pi\xi/\phi)$ and integrate them from $-\phi$ to ϕ , the set of equations for the field and the off-diagonal element of the density matrix is transformed into

$$(q_n^2 - 1)E(n) - \phi^{-1} [e^{in\pi} E'(\phi) - E'(0)] \\ = 2im \int_{-\infty}^{+\infty} \sigma(n, \nu) \exp(-\nu^2) d\nu, \quad (14a)$$

$$(\eta + iq_n\nu)\sigma(n, \nu) = E(n), \quad (14b)$$

where $q_n = n\pi/\phi$. The system of Eqs. (14) can be solved with respect to the Fourier coefficient of the field,

$$E(n) = \phi^{-1} \frac{(-1)^n E'(\phi) - E'(0)}{q_n^2 - 1 - 2imI(n)}, \quad (15)$$

where $I(n) = \int_{-\infty}^{+\infty} [\exp(-\nu^2)/(\eta + iq_n\nu)] d\nu$. From Eq. (15), it is seen that in order to calculate the surface admittance it

is essential to find the connection between field derivatives on the layer boundaries. Substitution of Eq. (15) into Eq. (13) yields

$$E(\xi) = \sum_{n=-\infty}^{\infty} E(n) e^{iq_n\xi} = \sum_{n=-\infty}^{\infty} \frac{(-1)^n E'(\phi) - E'(0)}{q_n^2 - 1 - 2imI(n)} e^{iq_n\xi}. \quad (16)$$

For the field at the boundaries of layers $\xi = 0$ and $\xi = \phi$, we have

$$E(0) = E'(\phi)S^- - E'(0)S^+, \quad (17a)$$

$$E(\phi) = E'(\phi)S^+ - E'(0)S^-. \quad (17b)$$

Here, we have introduced two sums,

$$S^+ = \phi^{-1} \sum_{n=-\infty}^{\infty} \frac{1}{q_n^2 - 1 - 2imI(n)}, \quad (18a)$$

$$S^- = \phi^{-1} \sum_{n=-\infty}^{\infty} \frac{(-1)^n}{q_n^2 - 1 - 2imI(n)}. \quad (18b)$$

Finally, solving Eqs. (8c), (8d), and (17) for the ratio $E(0)/E'(\phi)$, we can find an exact expression for the surface admittance in terms of sums S^+ and S^- ,

$$M(0) = \frac{1 - in_2 S^+}{n_2(S^- - S^+)(S^- + S^+) - iS^+}. \quad (19)$$

In order to test our theory, let us first assume that there is no vapor in the layer. For this instance, we can set parameter m equal to 0 in Eq. (19). Therefore, we get rid of the integral in Eqs. (18a) and (18b), and sums S^+ and S^- converge to explicit analytic functions,

$$S^+ = \phi^{-1} \sum_{n=-\infty}^{\infty} \frac{1}{(n\pi/\phi)^2 - 1} = -\cot \phi, \quad (20a)$$

$$S^- = \phi^{-1} \sum_{n=-\infty}^{\infty} \frac{(-1)^n}{(n\pi/\phi)^2 - 1} = -\csc \phi. \quad (20b)$$

Substitution of Eqs. (20a) and (20b) into Eq. (19) leads to the well-known result of the wave interference in the empty Fabry-Pérot resonator,

$$r = \frac{i(n_1 - n_2) + (n_1 n_2 - 1) \tan \phi}{i(n_1 + n_2) + (n_1 n_2 + 1) \tan \phi}, \quad (21)$$

where ϕ denotes the thickness of the gap between dielectric media l divided by reduced wavelength $\lambda = \lambda/2\pi$.

IV. NUMERICAL CALCULATION

In the following section, the reflectivity is calculated numerically for different thicknesses of the gas layer via the Fourier method described above [Eqs. (10), (18), and (19) were used to calculate the reflectivity]. The results of calculation are displayed in Fig. 2 where the reflectivity is plotted as a function of dimensionless detuning Ω .

From the comparison of dashed and solid curves in Fig. 2, it can be seen that with an increase in the layer thickness by the wavelength of the incident light, the spectral line contour of the reflection coefficient almost periodically repeats itself.

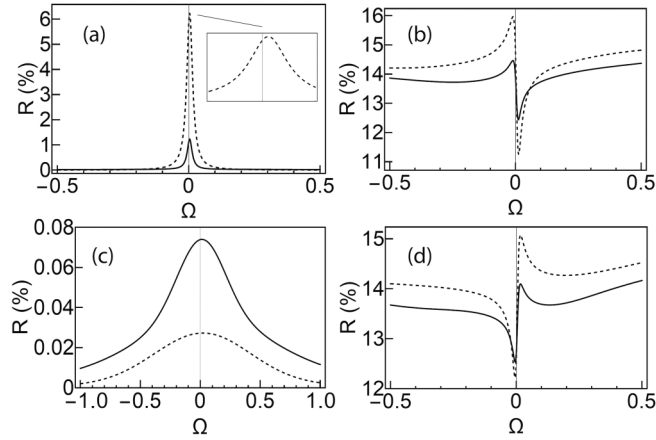


FIG. 2. Reflectivity of a thin-vapor layer against dimensionless detuning calculated by the Fourier method for eight different layer thicknesses. Dashed curves: (a) $l = \lambda/2$ (the inset: spectral behavior of reflectivity near resonance: $|\Omega| \leq 0.03$), (b) $l = 3\lambda/4$, (c) $l = \lambda$, and (d) $l = 5\lambda/4$; solid curves: (a) $l = 3\lambda/2$, (b) $l = 7\lambda/4$, (c) $l = 2\lambda$, and (d) $l = 9\lambda/4$. The parameters Γ , n_1 , n_2 , and m were taken equal to 0.01, 1.5, 1.5, and 0.001, respectively.

This λ -periodic dependence arises from spatial oscillations of the transient component of the polarization. In addition, the presence of the Fabry-Pérot resonances determines the $\lambda/2$ -periodic dependence of the nonresonant contribution to the total reflection on the layer thickness. This effect is manifested in a significant increase in the total reflection level at layer thicknesses $l = (n - 1/2)\lambda/2$, where $n = 1, 2, 3, \dots$ [see Figs. 2(b) and 2(d)]. Despite the fact that, according to Eq. (21), the condition $l = (2n - 1)\lambda/2$ corresponds to the destructive interference of waves reflected from the layer boundaries, in this case, we obtain a narrow Doppler-free even spectral contour (with the Lorentzian line shape), which is related to the resonant contribution of gas to the total reflection. Contrary to that, whenever the layer thickness coincides with an integer number of wavelengths $l = n\lambda$, the sub-Doppler contributions coming from adjacent half-wavelength-thick slices cancel each other and the sub-Doppler structure in the reflection spectrum disappears. At the same time, partial absorption of the transmitted wave causes a decrease in the effect of destructive interference of nonresonant contributions from waves reflected from the boundaries of the layer. This explains the appearance of a wide Doppler spectral contour in the reflection as well as an increase in its amplitude at a layer thickness $l = 2\lambda$ [see Fig. 2(c)]. Finally, the situation $l = (n - 1/2)\lambda/2$ corresponds to the presence of the sub-Doppler feature with approximately an odd spectral contour with respect to the resonant transition of the gas atoms. In this case, the slope of the odd spectral contour is determined by the interference of resonant contributions to the total reflection. For all other layer thicknesses, reflectivity is determined by a superposition of even and odd contributions. It is also worth noting that, in the limit of large thicknesses of the gas layer, the second boundary ceases to contribute to total reflection due to the attenuation of the transmitted wave. Consequently, numerical calculation gives the well-known logarithmic spectral line profile of the selective reflection of

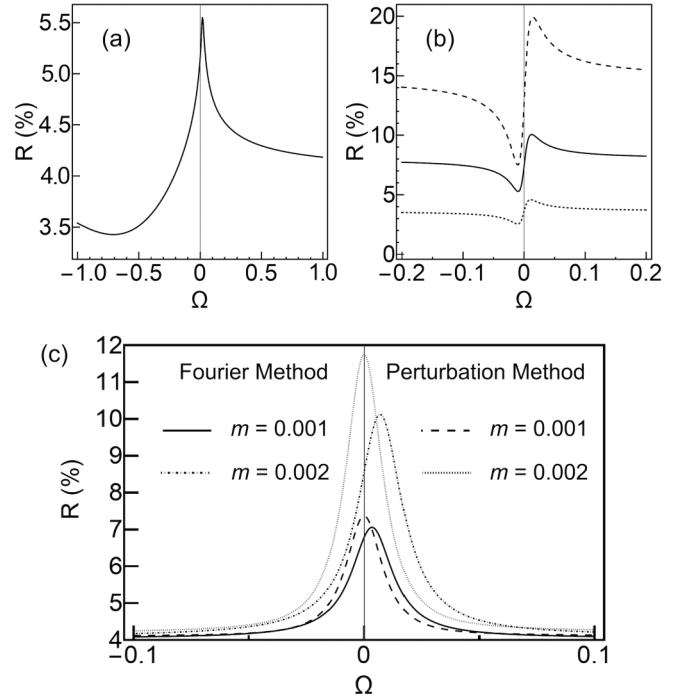


FIG. 3. (a) Reflectivity of a thick gas layer in the resonance region versus the dimensionless detuning calculated by the Fourier method for $m = 0.005$, $\Gamma = 0.01$, $l = 500\lambda$, $n_1 = 1.5$, and $n_2 = 1.5$. (b) Reflectivity of the sub- λ -thick vapor layers calculated by the Fourier method for $m = 0.001$, $\Gamma = 0.01$, $n_1 = 1.5$, and $n_2 = 1.5$: dashed curve: $l = \lambda/4$; solid curve: $l = \lambda/8$; and dotted curve: $l = \lambda/13$. (c) Comparison of the spectral line profiles of the reflectivity calculated by the Fourier method (solid and dot-dashed curves) with the perturbation theory solution (dashed and dotted curves) at various atomic vapor densities: solid and dashed curves: $m = 0.001$; dotted and dot-dashed curves: $m = 0.002$ for $\Gamma = 0.01$, $n_1 = 1.5$, and $n_2 = 1$.

light from a thick gas layer $R \propto -\ln(\Omega^2 + \Gamma^2)$ in the region $|\Omega| \ll 1$ [3,17], which is shifted towards the short-wave part of the spectrum [this result is presented in Fig. 3(a)].

Taking into account recent experiments in which the resonance reflection from and transmission through the gas cells with a thickness less than a wavelength are studied [2,6], it seems of value to examine the situation $l \ll \lambda$ as well. However, the simulation of this case requires accurate consideration of the van der Waals interactions, which is beyond the scope of this paper. In Fig. 3(b), we provide the results of numerical calculation of the reflectivity of sub- λ -thick vapor layers in order to demonstrate that, in the absence of van der Waals interactions, the reflectivity contains the approximately odd spectral line profile in the case of specular reflection of atoms colliding with the walls. It can be seen that a decrease in the layer thickness (from $l = \lambda/4$ to $l = \lambda/13$) leads to a decrease in both the total reflection (i.e., the efficiency of the constructive interference of the nonresonant contributions of waves reflected from the boundaries of the layer) and the resonant contribution of gas.

In the limit of a highly rarefied gas, an approximate solution of Eqs. (6a) and (6b) is to be found via the perturbation theory method described in detail in Ref. [3]. In order to

compare the rigorous solution of the thin gas layer problem for the case of specular boundary conditions with the result of perturbation theory, we presented the procedure for finding the first-order correction of the reflected field in the framework of perturbation theory in the Appendix. In this paper, we are mostly interested in the resonant part of the reflectivity. In fact, the nonresonant contribution to the total reflection can be eliminated by the proper selection of the parameters of the antireflection coatings [18]. Although Eqs. (A5)–(A7) in the perturbation method are written for the case of the absence of reflection from the rear boundary, to exclude the influence of the second wall in the Fourier method, we can put formally parameter $n_2 = 1$ in Eqs. (10), (18), and (19). A comparison of the reflection spectra of resonant radiation from a thin gas layer calculated by two methods is displayed in Fig. 3(c).

Given that the calculation according to perturbation theory is performed in the first order in m , we can see from Fig. 3(c) that high-order terms lead to a minor deformation of the spectral line profile and to a shift of its maximum. In addition, the peaks in the reflection spectra calculated through the perturbation theory have larger amplitude since the perturbation method does not take into account the absorption of the light. In the far wings of the resonance, the spectral line profiles practically coincide.

Equations (10), (18), and (19) allow us to accurately calculate the values of the blueshift in a wide range of atomic number densities. With an increase in the concentration of atomic vapors, the spectral line contour of the reflection coefficient broadens, the amplitude of the maximum increases, and its position shifts to the short-wavelength part of the spectrum [see Fig. 3(c)]. In the case of a thick layer, a universal proportionality constant may be found in the framework of the perturbation theory [9]. Contrary to that, in the case of a thin gas layer, the meaning of the shift is to be elaborated because the line shape depends on the layer thickness. Because of the inequality $m < \Gamma$ that holds due to the self-broadening effect [19] in the domain of the strong spatial dispersion $\Gamma < 1$, the shift of the resonance frequency $\Delta\Omega$ is well described by a linear dependence on m . As an example, we quote here the relation $\Delta\Omega = 3.54m$ obtained numerically from Eqs. (10),

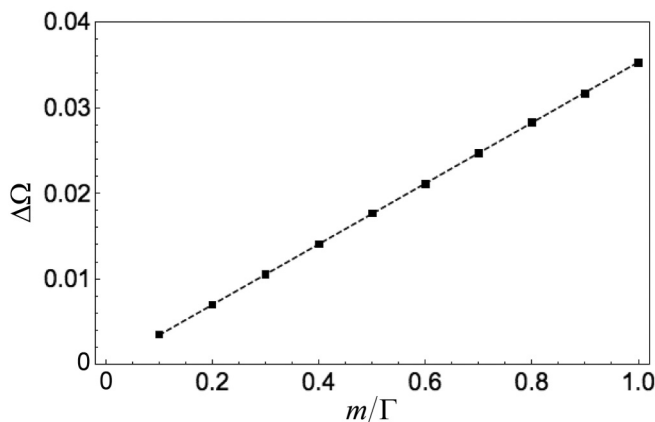


FIG. 4. The blueshift normalized to the Doppler width as a function of m for $m \leq \Gamma = 0.01$ and $l = \lambda/2$. The dashed line represents the linear fit $\Delta\Omega = 3.54m$.

(18), and (19) by simulation of the even spectral line contour corresponding to $l = \lambda/2$ and $\Gamma = 0.01$. In Fig. 4, the shift of the resonance frequency normalized to the Doppler width $k\nu_T$ is plotted as a function of m for the aforementioned case. The deviation from the linear dependence resulting from the influence of high-order terms in m is only 2×10^{-4} at the maximum possible $m = \Gamma = 0.01$. We emphasize that this linear dependence of the shift on m manifests the nonlinear dependence of the spectra on the vapor density.

V. DISCUSSION AND CONCLUSION

The special role of the cavities of half-wavelength thickness in the Doppler width cancellation was first pointed out in the microwave region. The main idea of Ref. [20] was that, in a pillbox-shaped cell, the slowly moving atoms contribute most to the absorption as well as to the fluorescence simply because they spend more time in their free flights between the walls. With the highly reflective walls, the field inside the cell is very close to a standing wave. Then, in the case of a half-wavelength-thick cell, all atoms experience the field oscillations of the same phase. Nevertheless, after the work on selective reflection from a gaseous half-space [1], it becomes clear that the main reason for the Doppler width cancellation is the transient polarization of the atoms that was not accounted for in Ref. [20]. Indeed, in the case of specular reflection, considered above, polarization of the atom, which has just changed the direction of its movement, does not correspond to its newly acquired detuning. Transient polarization of an atom departing from the surface with velocity v is depicted in Fig. 5 and contrasted with the steady-state polarizations of an atom with the same velocity and an atom arriving at the surface with velocity $-v$.

The difference between the transient and the steady-state polarizations of the departing atom underlies narrow sub-Doppler features in reflection spectra. The same is true in the case of polarization quenching at the surface considered earlier [3]. It is interesting to note that the major role of the polarization transients was overlooked in the first demonstration of the Doppler width cancellation [1]. Interference of two transient contributions that start their oscillations from the opposite walls leads to the periodic dependence of the line shapes on the cell thickness and further enhancement of the effect, whereas the details of the scatterings at the walls are of minor importance. Because of that, the results of the rigorous solution of the problem formulated for the specular reflection of the atoms have the general significance and may be used as a clue in the case of diffuse and quenching collisions for which the rigorous solutions are still absent. Below, we specify some peculiarities connected with the specular reflection of atoms colliding with the walls.

According to our calculations [see Fig. 2(a)], the amplitude of selective reflection of light from a thin layer of resonant gas in the case of specular reflection of atoms colliding with the walls is larger when in the case of complete quenching considered in Ref. [3]. As shown in Ref. [17], at large layer thicknesses $l \gg \lambda$, the spectral line profile of the selective reflection signal has a logarithmic form, and its amplitude for the case of specular reflection is approximately two times larger than the corresponding amplitude in the case of diffuse

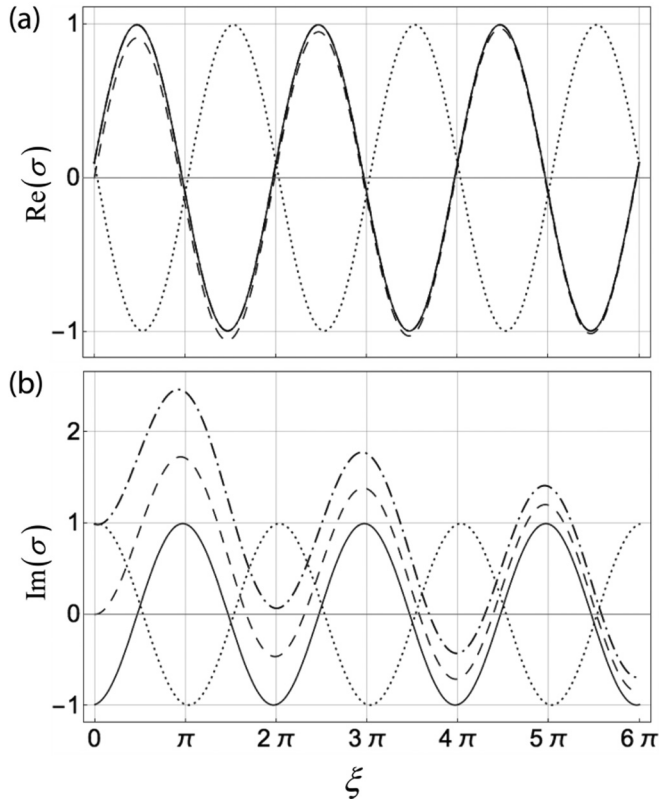


FIG. 5. The origin of the sub-Doppler features illustrated on the example of atomic polarization near an isolated boundary situated at $\xi = 0$. The difference between transient and steady-state polarizations of an atom departing from the surface is given for the particular absolute value of atomic velocity $|v| = 1$ and the following set of parameters: $\Omega = 0$, $\Gamma = 0.1$, and $E_0 = 1$. All complications connected with the self-consistent solution of Eqs. (6) are omitted. The real and imaginary parts of σ are presented in subfigures (a) and (b), respectively. Dotted curve: steady-state polarization of an atom arriving at the surface with velocity $v = -1$; solid curve: steady-state polarization of an atom departing from the surface with velocity $v = 1$; dashed curve: transient polarization of the departing atom after quenching collision with the surface for $v = 1$ [$\sigma(\xi = 0, v > 0) = 0$]; dot-dashed curve: transient polarization of the departing atom after specular collision with the surface for $v = 1$ (the real part of this solution coincides with the steady-state polarization of the departing atoms represented by the solid curve). Although the steady-state polarization implied in the conventional dispersion theory leads to the Doppler broadened spectral lines, transient polarizations initiated by specular as well as by quenching collisions lead to sub-Doppler linewidths.

boundary conditions. However, if the thickness of the vapor layer is comparable to wavelength λ , the differences in the amplitude and line shape of the resonance for models of diffuse and specular reflection are much more considerable. In the case of specular reflection, collisions with the walls allow a large part of the atoms to oscillate at their own frequency for a long time. The interference of contributions from individual atoms leads to the formation of a narrow and intense peak with a Lorentzian spectral profile near the resonance. In the crude approximation, the most slowly moving atoms make the major contribution to the reflectivity spectrum. With an

increase in the layer thickness, atoms have enough time to adapt to the external field before they collide with the wall. As a result, the amplitude of the Lorentz spectral contour decreases significantly, and the spectral contour of selective reflection has a mostly logarithmic singularity [see Fig. 3(a)].

Finally, the shift of the resonant frequency is reasonably well described by a linear dependence on the number density of gas atoms. This result is consistent with the magnitude of the blueshift obtained numerically in Ref. [17] for the case of a thick gas layer and specular wall collisions. Although, it was shown numerically in Ref. [11] and analytically in Ref. [9] that the Lorentz-Lorenz local-field correction partially reduces the value of the blueshift, this shift of purely electrodynamic nature is to be accounted for in any attempts of measuring van der Waals atom-surface interaction in selective reflection from and transmission through thin-vapor layers.

APPENDIX

In the framework of perturbation theory with respect to the vapor density, one starts with the solution of Eq. (6a) without the right-hand side,

$$E(\xi) = E_0 \exp(i\xi), \quad (\text{A1})$$

that corresponds to a plane electromagnetic wave of a constant amplitude inside the vapor layer. When this solution is substituted to the right-hand side of Eq. (6b), its particular solution reads as

$$\sigma(\xi, v) = \frac{E_0 \exp(i\xi)}{\eta + iv} + c \exp(-\eta\xi/v), \quad (\text{A2})$$

where c is determined by the boundary conditions (11a) for atoms with positive velocities and by (11b) for atoms moving in the opposite direction. The first term on the right-hand side of Eq. (A2) stands for a steady-state polarization value, which exists in the case of the absence of the walls and can be obtained by eliminating the derivative in Eq. (6b). The imposition of boundary conditions (11) on Eq. (A2) yields

$$\frac{\sigma(\xi, v > 0)}{E_0} = \frac{e^{i\xi}}{\eta + iv} + \frac{1 - e^{i\phi - \eta\phi/v}}{1 - e^{-2\eta\phi/v}} \frac{2iv e^{-\eta\xi/v}}{\eta^2 + v^2} \quad (\text{A3})$$

for atoms with a positive velocity projection $v > 0$ and

$$\begin{aligned} \frac{\sigma(\xi, v < 0)}{E_0} &= \frac{e^{i\xi}}{\eta + iv} + (e^{i\phi + \eta\phi/v} - e^{4\eta\phi/v}) \\ &\times (1 - \coth \eta\phi/v) \frac{ive^{-\eta\xi/v}}{\eta^2 + v^2} \end{aligned} \quad (\text{A4})$$

for atoms with a negative velocity projection $v < 0$. After we determine the nondiagonal element of the density matrix, we can obtain the approximate solution of Eq. (6a) in terms of the Green's function. For the case of the absence of reflection from the rear boundary for the reflected wave, we have

$$E_r = E_0 \frac{n_1 - 1}{n_1 + 1} - m \int_0^\phi e^{i\xi} d\xi \int_{-\infty}^{+\infty} \sigma(\xi, v) e^{-v^2} dv. \quad (\text{A5})$$

The first term in Eq. (A5) corresponds to the nonresonant reflection of light from the first boundary, whereas the second term determines the contribution of selective reflection.

Finally, after the integration, we can obtain an approximate expression for the reflectivity to the first order with respect to m in the following way:

$$R = \left(\frac{n_1 - 1}{n_1 + 1} \right)^2 + 2 \frac{n_1 - 1}{n_1 + 1} \text{Re}(r_s), \quad (\text{A6})$$

where

$$r_s = m \left\{ \frac{e^{2i\phi} - 1}{2} \int_{-\infty}^{+\infty} \frac{e^{-v^2} dv}{v - i\eta} + e^{2i\phi} \int_{-\infty}^{+\infty} \frac{v^2 e^{-v^2} [-1 + \coth \phi\eta/v] dv}{(v - i\eta)(v + i\eta)^2} + \int_0^{+\infty} \frac{v^2 e^{-v^2} [1 + \coth \phi\eta/v] dv}{(v - i\eta)(v + i\eta)^2} - 2e^{i\phi} \int_0^{+\infty} \frac{v^2 e^{-v^2} \text{csch } \phi\eta/v dv}{(v - i\eta)(v + i\eta)^2} + \int_{-\infty}^0 [-e^{i\phi}(1 + e^{2\phi\eta/v}) + e^{3\phi\eta/v} v] \frac{v^2 e^{-v^2} \text{csch } \phi\eta/v dv}{(v - i\eta)(v + i\eta)^2} \right\}. \quad (\text{A7})$$

Equations (A6) and (A7) allow us to calculate the spectral line profile of the reflection coefficient by means of numerical integration.

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