Model description of the transit-time problem for fast particles crossing a laser beam

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The model description of the spectral line broadening for atoms crossing the laser beams is presented. The combined effect of the transit-time broadening, natural broadening, and transverse Doppler broadening is considered. This is done by introducing one-parametric adiabatic damping of atom-laser interaction where the particles interact with the laser during a finite time interval. The simplest rectangle distribution of the light intensity inside the laser beam is employed. The dependence of absorption probability on the transit time for different detunings is calculated and compared with earlier results. The limits for using the Lorentz line profile for describing the interaction of fast particles with laser beams are established.

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I. INTRODUCTION

It is well known that the finite interaction time between the particles in atomic (molecular) beams and laser beams leads to the broadening of atomic spectroscopic lines. This broadening is called transit-time broadening and the corresponding width is called transit width. In many experiments, where the atomic resonant spectroscopy is used the transit width becomes larger than the natural (radiative) width. This happens in the case of very fast particles or when the upper or lower resonant atomic (molecular) state is metastable. As a particular example we may refer to the recent experiment on the observation (no observation is yet recorded) of the electron electric dipole moment in the resonant transition in molecule ThO where the lower resonant state is metastable [1]. Another version of such experiment with ThO for the transition with the upper metastable state was proposed recently in Ref. [2], and see also Ref. [3].

There are several books and papers where the transit-time problem was discussed from different points of view. We may refer to the books [4,5] and to Refs. [6–11] and the references therein. In these books and papers various methods (density matrix approach [9,10], relativistic field-theoretical perturbation theory [11]) were applied. As a rule the electromagnetic field created by a laser beam is considered as classical but the interaction of this field with atoms (molecules) is treated in frames of quantum mechanics. Thus, the problem is solved within some model (classical electromagnetic potential) but with the proper choice of this model all the necessary details of the process, i.e., characteristics of the absorption line profile, can be accounted for, including the transit broadening. This broadening (transit width Γ_T) can be introduced in different ways: either simply via the Heisenberg inequalities $\Gamma_T \tau \gg 1$ (here τ is the transit time) [4], or in a more complicated way solving the equations of motion for the particle (atom) moving in the laser field [9,10].

However, as far as we know, there were no attempts to justify the application of the Lorentz line profile theory for description of atoms (molecules) interacting with laser beams during finite time intervals. This becomes especially important when the natural broadening (natural width Γ_N) is negligible, as in the case that the ThO molecule and transit broadening (transit width Γ_T) begin to determine the Lorentz profile of the resonant spectroscopic line, taking the place of Γ_N in the Lorentz denominator. To justify this replacement and to fix the experimental parameters, allowing this replacement, is the main goal of the present paper. In our theory both broadenings, Γ_N and Γ_T, arrive in the energy denominators as a result of evaluation of interaction between atomic electron and classical potential, presenting the laser field.

In the subsequent sections we will consider one by one the transit width Γ_T, natural width Γ_N, and transverse Doppler width Γ_D for atomic beams and their combination.

II. TRANSIT BROADENING

For description of the resonant laser light absorption by the bound electron in an atomic beam we evaluate the amplitude for the elastic photon scattering on the bound atomic electron. The imaginary part of this amplitude will represent the absorption; the square modulus of this part will give the probability of absorption. Dependence of this probability on the frequency detuning and on all types of broadening will represent the absorption resonant spectral line—the main instrument in our paper.

Using standard perturbation theory we begin with an expression for the amplitude, corresponding to the Feynman graph Fig. 1. We should stress that we are working in the Furry picture, which has become standard for relativistic calculations of atoms and molecules. This picture is especially adjusted for calculations of bound particle systems (see Refs. [12–14]). In the case of atoms it employs the complete set of one-electron wave functions which are the solutions of the Dirac equation for the electron in the Coulomb field of the nucleus or the complete set of solutions of the Dirac-Hartree-Fock equations for many-electron atoms. Unlike the

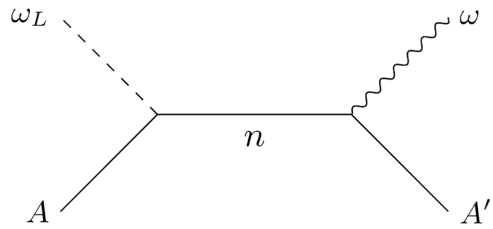

FIG. 1. The first-order Feynman diagram for the resonant interaction between laser light (dashed line) and bound electron (solid line). Here A denotes the initial state of an electron, A' is the final state of the electron, in case of elastic scattering $A' = A$, and n is the resonant state. The wavy line denotes an emitted photon, ω_L is the laser frequency, and ω is the frequency of the emitted photon.

ordinary description where the incident photon interacts with the atomic electron during the time interval from $t = -\infty$ to ∞ we assume that the atom (molecule) interacts with the electromagnetic classical potential created by the laser beam during the transit time τ. This interaction is resonant; the atom becomes excited and then decays. We consider both the cases when an atom decays inside and outside the laser beam, i.e., an arbitrary relation between the natural width Γ_N and transit width Γ_T. In both cases the decay is influenced by the transit time τ. Therefore in both vertices in Fig. 1 the time integration is limited by the τ value. The excitation and the decay should be considered as unique processes. The fast decay will correspond to the case $\Gamma_N \gg \Gamma_T$ and will automatically reproduce the results with $\tau = \infty$.

The S-matrix element for the process, depicted in Fig. 1 within the ordinary description, is

$$S^{(2)} = (-ie)^2 \int dx dx' \overline{\psi}_A(x)\gamma^\mu \widetilde{A}_\mu(x) S(x,x')\gamma^{\mu'} \times A^*_{\mu'}(x')\psi_{A'}(x'), \tag{1}$$

where $x \equiv (\boldsymbol{r}, t)$, $\boldsymbol{r}$ is the space coordinate vector, t is time, γ^μ ($\mu = 0, 1, 2, 3$) are Dirac matrices, e is the electron charge, ψ is the bound electron wave function, $\overline{\psi}$ is the Dirac conjugation of ψ, $\widetilde{A}_\mu(x)$ is the laser harmonic classical potential $\widetilde{A}_\mu(x) = \widetilde{A}_\mu(\boldsymbol{r})e^{i\omega_L t}$, ω_L is the laser frequency, $A^*_\mu(x)$ is the complex conjugated photon wave function $A_\mu(x) = A_\mu(\boldsymbol{r})e^{i\omega t}$, and ω is the emitted after decay photon frequency. $S(x,x')$ is the electron propagator:

$$S(x,x') = \frac{1}{2\pi i}\int d\omega' e^{i\omega'(t-t')} \sum_n \frac{\psi_n(\boldsymbol{r})\overline{\psi}_n(\boldsymbol{r}')}{E_n(1-i0)+\omega'}. \tag{2}$$

Summation in Eq. (2) runs over entire Dirac spectrum for the atomic electron:

$$\psi_n(x) = \psi_n(\boldsymbol{r})e^{-iE_n t}, \tag{3}$$

where E_n are the energies of bound electrons. The resonance condition is $\omega_L = E_n - E_A$, and the resonant state n is a state with positive energy. Throughout the paper we use the relativistic units $\hbar = c = 1$ ($\hbar$ is the Planck constant, and c is the speed of the light) and pseudo-Euclidean metric with metric tensor $g_{\mu\nu} = (-1, -1, -1, +1)$. After substitution of Eqs. (2) and (3) in Eq. (1) and integrating over the time variables we get

$$S^{(2)} = -\frac{e^2}{2\pi i}(2\pi)^2 \int d\boldsymbol{r} d\boldsymbol{r}' \overline{\psi}_A(\boldsymbol{r})\gamma^\mu \widetilde{A}_\mu(\boldsymbol{r}) \times \sum_n \frac{\psi_n(\boldsymbol{r})\overline{\psi}_n(\boldsymbol{r}')}{E_n - E_A - \omega_L}\gamma^{\mu'} A^*_{\mu'}(\boldsymbol{r})\psi_A(\boldsymbol{r}')\delta(\omega_L - \omega). \tag{4}$$

In the resonance approximation only one term in the sum over n in Eq. (4) remains. Usually, in experiments with interaction of atomic and laser beams and, in particular, in experiments of the type of Refs. [1–3], the main quantity of interest is the absorption probability for the photon within an atomic beam. The standard theory for absorption (emission) of the photon by an atomic electron is following Ref. [15]. Instead of absolute probability, the transition rate is introduced by definition

$$\mathcal{W}_{if} = \frac{1}{T}|S_{if}|^2, \tag{5}$$

where T is the observation time. Here i and f denote the initial and final states. The $|S_{if}|^2$ contains squared $\delta(E_i - E_f)$, where E_i and E_f are the initial and final energies of the whole system of particles. This expression is infinite. Then one of the δ functions is presented as the Fourier integral

$$\delta(E_i - E_f) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{i(E_i - E_f)t} dt \tag{6}$$

and replaced by

$$\delta_T(E_i - E_f) = \frac{1}{2\pi}\int_{-T/2}^{T/2} e^{i(E_i - E_f)t} dt, \tag{7}$$

so that

$$\delta(E_i - E_f)\delta_T(E_i - E_f) = \delta(E_i - E_f)\delta_T(0) = \frac{T}{2\pi}\delta(E_i - E_f). \tag{8}$$

The transition rate $\mathcal{W}_{if}$ defined by Eq. (5) becomes now finite. However, with introduction of transit time τ this procedure becomes uncertain. The existence of two different time intervals T and τ makes the definition of transition rate nonunique. Below we will demonstrate how it is possible to avoid it using the adiabatic damping of interaction between the photons and electrons.

Unlike the nonrelativistic quantum mechanics, where transit time can be introduced via dependence of absorption on gauge invariant electric field, we have to deal with electric-field potentials [see Eq. (1)] and the gauge invariance problem should be discussed from the beginning. The classical laser potential in Eq. (4) is a pure vector potential. The scalar potential is absent due to the absence of electric charges in the laser beam. The vector potential $\boldsymbol{A}(\boldsymbol{r}, t)$ we can choose, for example, as

$$\boldsymbol{A}(\boldsymbol{r}, t) = \boldsymbol{e}_L e^{i\omega_L t} e^{-i\omega_L \boldsymbol{v}_L \boldsymbol{r}}, \tag{9}$$

where $\boldsymbol{v}_L$ and $\boldsymbol{e}_L$ are the laser beam direction and polarization vectors, respectively, and ω_L is frequency. In the case when the scalar potential is absent, the gauge invariance

condition is

$$\mathrm{div}\boldsymbol{A} = 0. \tag{10}$$

The potential Eq. (9) satisfies this condition, since vectors $\boldsymbol{v}_L$ and $\boldsymbol{e}_L$ are orthogonal:

$$\mathrm{div}\boldsymbol{A} = -i(\boldsymbol{v}_L\boldsymbol{e}_L)\omega_L = 0. \tag{11}$$

Equation (9) corresponds to the rectangular distribution of the light intensity within the laser beam. Moreover, the real laser is not pure monochromatic as in Eq. (9). The generalization is straightforward but we remain with these simplifications because they do not influence our main goal—investigation of the transit-time problem.

The standard definition of the amplitude of any process is

$$S_{if} = -2\pi i\mathcal{U}_{if}\delta(E_i - E_f). \tag{12}$$

Below we will use another definition:

$$S_{if} = -\mathcal{U}_{if}. \tag{13}$$

The absolute probability of the process we define as

$$W_{if} = |\mathcal{U}_{if}|^2. \tag{14}$$

According to the discussion above an expression for the amplitude of the process of the laser light scattering on an atom can be written down in the form [see Eq. (4)]

$$\mathcal{U} = e^2(2\pi)^2\sum_n \frac{\langle A|\gamma_\mu\widetilde{A}_\mu|n\rangle\langle n|\gamma_{\mu'}A^*_{\mu'}|A\rangle}{E_n - E_A - \omega_L}\delta(\omega - \omega_L), \tag{15}$$

where

$$\langle i|\gamma_\mu A_\mu|f\rangle = \int d\boldsymbol{r}\overline{\psi}_A(\boldsymbol{r})\gamma_\mu A_\mu(\boldsymbol{r})|f\rangle. \tag{16}$$

In the resonance approximation the amplitude is proportional to the resonant energy denominator:

$$\mathcal{U} = (2\pi)^2 AI(\omega,\omega_L)\delta(\omega-\omega_L) = (2\pi)^2 A\frac{1}{\Delta\omega}\delta(\omega-\omega_L), \tag{17}$$

where

$$A = e^2\langle A|\gamma_\mu\widetilde{A}_\mu|n\rangle\langle n|\gamma_{\mu'}A^*_{\mu'}|A\rangle, \tag{18}$$

and $\Delta\omega = E_n - E_A - \omega_L$ is the detuning. In principle, A depends also on ω_L via dependence of the potential Eq. (9). However, this dependence is nonresonant (NR) and we can set $\omega_L = E_n - E_A$ everywhere but in the resonant energy denominator. This assumption is common for the resonance spectroscopy studies. In Eq. (12) the most important is the factor $I(\omega,\omega_L)$ which determines the dependence of the absorption (emission) intensity on the frequency (detuning) and on the various types of broadening. This factor, which may be called "reduced amplitude," will be the main subject of our further studies. In the case of the absence of any broadening the reduced amplitude is $I(\omega,\omega_L) = \frac{1}{\Delta\omega}$.

To introduce the transit time τ we will apply the adiabatic S-matrix approach employed earlier for the evaluation of the energy levels for bound electrons in relativistic theory of atoms [16–18]. This approach consists of multiplying the operator $\hat{H}_{\mathrm{int}}(t)$ (interaction of electromagnetic and electron-positron fields in the interaction representation) which enters the S-matrix element and corresponds to each interaction vertex in the related Feynman graph, by adiabatic exponent $e^{-\lambda|t|}$. At the end of evaluation $\lambda \to 0$, which makes the process of introducing the interaction "adiabatic." We introduce the adiabatic exponent in both vertices in the Feynman graph Fig. 1, describing the interaction of an atomic electron with the classical laser potential not to make it adiabatic but to make it finite in time. For this purpose we do not set $\lambda \to 0$, but set it equal to $\lambda = \frac{1}{\tau}$, where τ is transit time. The τ value remains as a parameter in the theory which depends on the particular experimental conditions. It is natural to define τ via particle velocity $\boldsymbol{v}$ and the laser beam diameter d. Presenting $\boldsymbol{v}$ as the sum of two components $\boldsymbol{v} = \boldsymbol{v}_{||} + \boldsymbol{v}_\perp$ where indices $||$ and $\perp$ correspond to the parallel and orthogonal directions with respect to the direction of the atomic beam, and assuming that the atomic and laser beams are orthogonal, we may set $\tau = \frac{d}{v_{||}}$, where $v_{||} = |\boldsymbol{v}_{||}|$. The component $\boldsymbol{v}_\perp$ will define the transverse Doppler broadening (see Sec. IV). Below it will be shown that with introduction of the adiabatic exponent both expressions for the amplitude Eq. (17) and for the probability Eq. (14) will become finite (dependent on transit time τ). It should be noted that the introduction of the time-dependent damping factor violates the Lorentz invariance of the expression for the absorption probability. However, Lorentz invariance is of minor importance for the process with bound electrons since in this case there always exists one preferable frame of reference connected with the atomic nucleus for atoms or with center of mass for molecules.

A question about the choice of damping factor also might arise. The answer is that there is no reason for the dependence of the results on this choice, at least in the case when the damping effect is not too strong. What "too strong" means for the problem of interest we will discuss at the end of this section. Regardless, the adiabatic exponential form of the damping factor is, to our mind, most plausible.

For our purposes it will be enough to consider further only time- and frequency-dependent parts of the amplitude, i.e., to set

$$\mathcal{U}_\lambda = AJ_\lambda(\omega,\omega_L), \tag{19}$$

where

$$J_\lambda(\omega,\omega_L) = \frac{1}{2\pi i}\int dtdt'd\omega'\frac{1}{E_n(1-i0)+\omega'} \times e^{iE_nt}e^{-i\omega_Lt}e^{-\lambda|t|}e^{i\omega'(t-t')}e^{-iE_At'}e^{i\omega t'}e^{-\lambda|t'|}. \tag{20}$$

For performing the integration over time variable t in Eq. (20) we employ the following formula (known as the Poisson kernel):

$$\int_{-\infty}^{+\infty} dte^{-\lambda|t|+iat} = \frac{i}{2\pi}\left[\frac{1}{a+i\lambda} + \frac{1}{-a+i\lambda}\right] = \frac{1}{\pi}\frac{\lambda}{\lambda^2+a^2} \equiv \delta_\lambda(a), \tag{21}$$

which approaches $\delta(a)$ as $\lambda \to 0$. Then for the integral Eq. (20) we will have

$$J_\lambda = \frac{1}{2\pi i}\frac{1}{\pi^2}\int_{-\infty}^{\infty} d\omega' \frac{1}{E_n - i\delta + \omega'}\frac{\lambda}{(E_A+\omega_L+\omega')^2+\lambda^2} \times \frac{\lambda}{(E_A+\omega+\omega')^2+\lambda^2}, \tag{22}$$

where $\delta > 0$ is the infinitesimal shift indicating that the resonant state is the positive-energy one. To connect Eq. (19) with Eq. (12) which after introduction of the adiabatic exponent should look like

$$\mathcal{U}_\lambda = AI_\lambda(\omega, \omega_L)\delta_\lambda(\omega - \omega_L) \tag{23}$$

we need to separate out from $J_\lambda(\omega, \omega_L)$ the factor

$$\delta_\lambda(\omega - \omega_L) = \frac{1}{\pi}\frac{\lambda}{(\omega-\omega_L)^2+\lambda^2}, \tag{24}$$

which tends to $\delta(\omega - \omega_L)$ as $\lambda \to 0$.

The expression for the amplitude $\mathcal{U}$ Eq. (17) is divergent twice: first, for the elastic scattering when $\omega = \omega_L$, and second, at the point of resonance, when $\Delta\omega \to 0$. Both divergences can be avoided by introduction of the adiabatic exponent: evaluation of the integral $J_\lambda(\omega, \omega_L)$ performed in the Appendix for arbitrary values of λ results in a finite expression for $J_\lambda(\omega = \omega_L)$. So expression (22) for $J_\lambda(\omega = \omega_L)$ can be used for determination of the amplitude $\mathcal{U}_\lambda(\omega = \omega_L)$ via

$$\mathcal{U}_\lambda(\omega = \omega_L) = AI_\lambda(\omega = \omega_L)\delta_\lambda(\omega - \omega_L), \tag{25}$$

for the determination of reduced amplitude

$$I_\lambda(\omega = \omega_L) = J_\lambda(\omega = \omega_L)\delta_\lambda^{-1}(\omega = \omega_L), \tag{26}$$

and for regularization of Eq. (17) for $\omega = \omega_L$:

$$\mathcal{U}(\omega = \omega_L) = \lim_{\lambda\to 0}\mathcal{U}_\lambda(\omega = \omega_L). \tag{27}$$

The general expression for $J_\lambda(\omega, \omega_L)$ is (see the Appendix)

$$J_\lambda(\omega = \omega_L) = \frac{\lambda^2}{\pi^2}\left(\frac{1}{2}\frac{1}{\left(E_n - E_A - \omega_L^2\right)^2+\lambda^2}\frac{1}{(E_n - E_A - \omega)^2+\lambda^2} - \frac{1}{2i\lambda}\frac{1}{\omega-\omega_L}\left[\frac{1}{E_n - E_A - \omega_L - i\lambda}\frac{1}{\omega - \omega_L - 2i\lambda} - \frac{1}{E_n - E_A - \omega - i\lambda}\frac{1}{\omega_L - \omega - 2i\lambda}\right]\right). \tag{28}$$

Keeping $\lambda \neq 0$, using Eq. (25) and expanding Eq. (28) in terms of $\frac{\omega-\omega_L}{\lambda}$ in the limit $\omega \to \omega_L$ we find (see the Appendix)

$$J_\lambda(\omega = \omega_L) = \frac{1}{2\pi^2}\left(\frac{\lambda^2}{[(E_n - E_A - \omega_L)^2+\lambda^2]^2} - \frac{1}{\lambda}\frac{1}{E_n - E_A - \omega_L - i\lambda}\right), \tag{29}$$

$$I_\lambda(\omega = \omega_L) = \left(-\frac{1}{\Delta\omega - i\lambda} + \frac{\lambda^3}{[\Delta\omega^2+\lambda^2]^2}\right). \tag{30}$$

Now we can set also $\lambda = 0$ and find that for the infinite transit time $\tau = \infty$ we return to the value $I = \frac{1}{\Delta\omega}$ in Eq. (17). For large detuning, $\Delta\omega \gg \lambda$ (as, for example, is required in the experiment proposed in Refs. [2,3]), the standard form of the Lorentz spectral line profile, i.e.,

$$I_\lambda \sim \frac{1}{\Delta\omega - \frac{i}{2}\Gamma_T}, \tag{31}$$

is restored, where $\Gamma_T = 2\lambda$. Transit width Γ_T replaces here the natural width Γ_N. This proof is the main result of our paper. From Eq. (30) we can see that for small detuning (very close to the point of resonance) the dependence of the line profile on the transit time becomes complicated and the Lorentz profile is not valid anymore. The critical relation between $\Delta\omega$ and τ is

$$\Delta\omega\tau > 1. \tag{32}$$

Formally, Eq. (32) coincides with the uncertainty relation, but its sense is different: the uncertainty relation determines the limit for the accuracy of measuring frequency, but Eq. (32) sets the limit for using the Lorentz form for description of the line profile.

III. NATURAL BROADENING

In fact, it can be shown that the transit-time width and radiative width can be taken into account simultaneously. The theory of a spectral line profile dependent on the natural broadening within quantum electrodynamics was first formulated by Low [19] (see further development in Ref. [20]).

In particular, for the process described by the Feynman graph Fig. 1 we need to take into account the infinite chain of the electron self-energy insertions into the internal electron line. The first term of this chain is given by the Feynman diagram Fig. 2. The corresponding S-matrix element can be written as (in case of elastic scattering)

$$S^{(4)} = (-ie)^4\int dx_1dx_2dx_3dx_4\overline{\psi}_A(x_1)\gamma^{\mu_1}\widetilde{A}_{\mu_1}(x_1)S(x_1,x_2) \times \gamma^{\mu_2}S(x_2,x_3)\gamma^{\mu_3}S(x_3,x_4)\gamma^{\mu_4}A^*_{\mu_4}(x_4)\psi_A(x_4) \times D_{\mu_2\mu_3}(x_2,x_3), \tag{33}$$

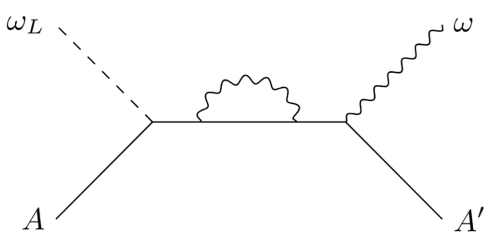

FIG. 2. The Feynman diagram Fig. 1 with one electron self-energy insertion. The internal wavy line denotes the photon propagator, and other notions are the same as used in Fig. 1.

where $D_{\mu\nu}(x_2, x_3)$ is the photon propagator in the Feynman gauge,

$$D_{\mu\nu}(x_2, x_3) = \frac{\delta_{\mu\nu}}{2\pi i r_{23}} \int d\Omega e^{i|\Omega|r_{23} - i\Omega(t_2 - t_3)}, \tag{34}$$

$r_{23} = |\boldsymbol{r}_2 - \boldsymbol{r}_3|$, Ω is the frequency of the vacuum photon, and $\delta_{\mu\nu}$ is the Kronecker delta.

After insertion in Eq. (33) adiabatic exponents we get

$$S_\lambda^{(4)} = \frac{(-ie)^4}{(2\pi i)^4} \int dx_1 dx_2 dx_3 dx_4 d\omega' d\omega'' d\omega''' d\Omega \times \overline{\psi}_A(\boldsymbol{r}_1)\gamma^{\mu_1}\widetilde{A}_{\mu_1}(\boldsymbol{r}_1) \sum_{n_1} \frac{\psi_{n_1}(\boldsymbol{r}_1)\overline{\psi}_{n_1}(\boldsymbol{r}_2)}{E_{n_1}(1 - i0) + \omega'} \sum_{n_2} \frac{\psi_{n_2}(\boldsymbol{r}_2)\overline{\psi}_{n_2}(\boldsymbol{r}_3)}{E_{n_2}(1 - i0) + \omega''}$$

$$\times \sum_{n_3} \frac{\psi_{n_3}(\boldsymbol{r}_3)\overline{\psi}_{n_3}(\boldsymbol{r}_4)}{E_{n_3}(1 - i0) + \omega'''} \times \gamma^{\mu_4} A^*_{\mu_4}(\boldsymbol{r}_4)\psi_A(\boldsymbol{r}_4)\gamma^{\mu_2}\gamma^{\mu_3} \frac{\delta_{\mu_2\mu_3}}{r_{23}} e^{i|\Omega|r_{23}} \times e^{iE_A t_1} e^{i\omega_L t_1} e^{-\lambda|t_1|} e^{i\omega'(t_1 - t_2)} e^{i\omega''(t_2 - t_3)} e^{i\omega'''(t_3 - t_4)} e^{-i\omega t_4}$$

$$\times e^{-\lambda|t_4|} e^{-iE_A t_4} e^{-i\Omega(t_2 - t_3)}. \tag{35}$$

The adiabatic exponents are inserted only in the x_1 and x_4 vertices and not in the vertices x_2 and x_3, corresponding to the interaction of the atomic electron with the vacuum photon, since this kind of interaction always takes place in the infinite time interval. In the resonance approximation only one term in the sum over n_1 with $n_1 = n$ remains. Similarly, only one term $n_3 = n$ in the sum over n_3 remains. Here it may be worth it to mention the validity of the resonance approximation which is used throughout this paper. The NR correction to the energy levels in atoms was considered by many authors, including, for example, in Ref. [20]. In the hydrogen atom these corrections may reach the level of a few kHz and influence the accuracy of determination of the transition frequencies in hydrogen [21]. Still in such systems as the ThO molecule which are of most interest for us, the NR corrections are of the order $\Delta_{\rm NR} = \Gamma_N^2/\omega_0$. With $\Gamma_N \approx 1$ Hz for the metastable state and $\omega_0 = E_N - E_A \approx 10^{16}$ Hz these corrections become negligible. After integration over time and frequency variables in Eq. (35) in the resonance approximation a diagonal matrix element of the electron self-energy can be separated out. This matrix element in the general case can be presented as [20]

$$\langle u|\hat{\Sigma}(\xi)|d\rangle = e^2 \sum_m \frac{i}{2\pi} \int d\omega \frac{I_{ummd}(\omega)}{\xi - \omega - E_n(1 - i0)}, \tag{36}$$

$$I_{u_1u_2d_1d_2}(\omega) = \int d\boldsymbol{r}_1 d\boldsymbol{r}_2 \overline{\psi}_{u_1}(\boldsymbol{r}_1)\overline{\psi}_{u_2}(\boldsymbol{r}_2)\gamma^{\mu_1}\gamma^{\mu_2} \frac{\delta_{\mu_1\mu_2}}{r_{12}}$$
$$\times e^{i|\omega|r_{12}} \psi_{d_1}(\boldsymbol{r}_1)\psi_{d_2}(\boldsymbol{r}_2). \tag{37}$$

Then in the resonance approximation in case $\lambda = 0$ Eq. (35) can be written as

$$S_{AA}^{(4)} = S_{AA}^{(2)} \frac{\langle n|\hat{\Sigma}(E_n)|n\rangle}{\Delta\omega}, \tag{38}$$

where $\Delta\omega$ is detuning $\Delta\omega = E_n - E_A - \omega_L$, and n is the resonant state.

Similarly, the contributions of many electron self-energy insertions in the resonance approximation can be introduced. As a result, the following consequence will arise [19,20]:

$$\mathcal{U} \sim S^{(2)} \left(1 + \frac{\Sigma_{nn}}{\Delta\omega} + \left(\frac{\Sigma_{nn}}{\Delta\omega}\right)^2 + \dots\right), \tag{39}$$

where $\Sigma_{nn} \equiv \langle n|\hat{\Sigma}(E_n)|n\rangle$. This consequence represents the geometric progression and can be converted to

$$\mathcal{U} \sim \frac{1}{\Delta\omega} \frac{1}{1 + \frac{\Sigma_{nn}}{\Delta\omega}} = \frac{1}{\Delta\omega + \Sigma_{nn}}. \tag{40}$$

In Eq. (40) the matrix element Σ_{nn} can be presented traditionally as $\Sigma_{nn} = L_n - \frac{i}{2}\Gamma_n$, where L_n is the Lamb shift of the resonant level n and $\Gamma_n = \Gamma_N$ is the radiative (natural) width.

Replacing $S^{(2)}$ in Eq. (38) by $S_\lambda^{(2)}$ and using again Eqs. (31) and (40) for the case of large detuning ($\Delta\omega \gg \lambda$) we obtain for the reduced amplitude

$$I_\lambda \approx \frac{1}{\Delta\omega - \frac{i}{2}\Gamma_T - \frac{i}{2}\Gamma_N}. \tag{41}$$

Here we neglect the Lamb shift, since we are interested only in broadening.

IV. TRANSVERSE DOPPLER BROADENING

Having the absorption amplitude for the atomic (molecular) beam crossing the laser beam in the form Eq. (41) it is easy to take into account the transverse Doppler width Γ_D. This broadening arises due to the chaotic (thermal) motion of atoms within the beam in the direction orthogonal to the direction of the beam. For this purpose we have to introduce the Doppler effect, replacing the frequency ω in Eq. (41) by $\omega(1 - \frac{v_\perp}{c})$ and the velocity $v_\perp$ by the average velocity of chaotic motion of atoms. This averaging is commonly performed by convolution of amplitude with the Maxwell distribution of atomic velocities. Also commonly the natural broadening is incorporated in the process of this averaging. It is important that in our approach the dependence of the amplitude on Γ_T is exactly the same as the dependence on Γ_N and that Γ_T did arrive in the energy denominator.

Then the result of averaging we can present in the form (see, for example, Ref. [22])

$$\left\langle \frac{1}{\omega\left(1 - \frac{v_\perp}{c}\right) - \omega_0 - \frac{i}{2}(\Gamma_N + \Gamma_T)} \right\rangle_{AV}$$
$$= \frac{1}{\omega - \omega_0}[g(u, v) - if(u, v)], \tag{42}$$

where $\omega_0 = E_n - E_A$, $\omega - \omega_0 = \Delta\omega$, and variables u and v are defined as

$$u = \frac{\omega - \omega_0}{\Gamma_D}, \tag{43}$$

$$v = \frac{\Gamma_N + \Gamma_T}{2\Gamma_D}, \tag{44}$$

$$\Gamma_D = \omega_0 \frac{v_\perp}{c}, \tag{45}$$

$$v_\perp = \sqrt{\frac{2kT}{M}}, \tag{46}$$

where k is the Boltzmann constant, T is the absolute temperature, and M is the mass of an atom (molecule). The functions $g(u, v)$ and $f(u, v)$ are

$$g(u, v) = \mathrm{Im}\sqrt{\pi} e^{-w^2}[1 - \Phi(-iw)], \tag{47}$$

$$f(u, v) = \mathrm{Re}\sqrt{\pi} e^{-w^2}[1 - \Phi(-iw)], \tag{48}$$

where $w = u + iv$, $\Phi(z)$ is the error function [23]. In case of $u \gg 1$ and $v \ll 1$ the asymptotic expressions are valid:

$$g(u, v) \approx \frac{1}{u}, \tag{49}$$

$$f(u, v) \approx \sqrt{\pi e^{-u^2}} \frac{v}{u^2}. \tag{50}$$

With Eqs. (42)–(50) we come to the final expression for the reduced absorption amplitude dependent on the parameters $\Delta\omega$, Γ_N, Γ_T, and Γ_D:

$$I_\lambda = \frac{1}{\omega - \omega_0}[g(u, v) - if(u, v)]. \tag{51}$$

Absorption corresponds to the imaginary part of Eq. (51). The real part is corresponding to the dispersion. Note that Eq. (51) is valid only when Eq. (31) is valid, i.e., when $\Gamma_T \ll \Delta\omega$. For the very fast particles close to the resonance ($\lambda \gg \Delta\omega$) the dependence on the transit time becomes more complicated [see Eq. (30)].

V. RESULTS AND DISCUSSION

As the results of our paper in this section we present the numeric dependence of absorption probability in the experiments with crossing the atomic (molecular) beam and laser beams on the transit time $\tau = \Gamma_T^{-1}$ and on the frequency detuning $\Delta\omega$. This probability is represented by the ratio $f(u, v)/(\omega - \omega_0)$ [see Eq. (42)] and is depicted in Fig. 3. As a particular example we employ the proposed experiment [2,3] for observation of space- and time-parity odd Faraday rotation on the ThO molecule. In this experiment the transition from the ground state to the metastable excited state is chosen for the observation of the resonant optical rotation in an external electric field. The natural width $\Gamma_N \approx 1\,\mathrm{s}^{-1}$ is considered to be negligible compared to Γ_T. The velocities of the molecules in the beam are assumed to be about $v_{||} \approx 10^4\,\mathrm{cm\,s}^{-1}$. The transverse Doppler width is $\Gamma_D \approx 10^8\,\mathrm{s}^{-1}$ and detuning is of the order of several Γ_D. In this experiment the absorption represents the parasite effect and should be made as small as possible compared to the dispersion responsible for the optical rotation. The dispersion is represented by the ratio $g(u, v)/(\omega - \omega_0)$ in Eq. (42). In Fig. 4 the ratio $f(u, v)/g(u, v)$ is plotted as a function of τ for different $\Delta\omega = \omega - \omega_0$ values. Qualitatively the dependencies in Figs. 3 and 4 correlate with the corresponding dependencies obtained in Ref. [10] on the basis of nonrelativistic quantum-mechanical calculations. The behavior of $f(u, v)/g(u, v)$ in Fig. 4 confirms the choice of the parameters in the experiment [3].

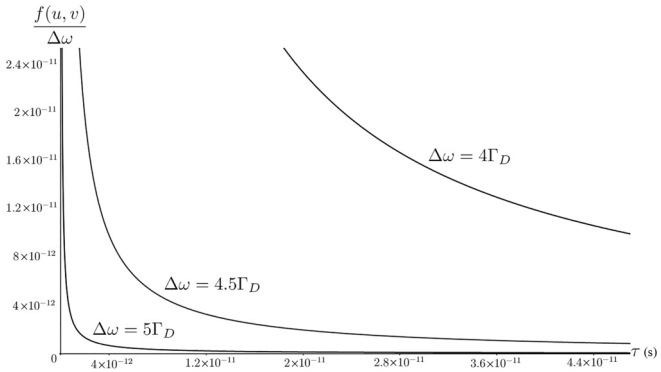

FIG. 3. The absorption probability vs transit time for different detunings $\Delta\omega = \omega - \omega_0$.

VI. CONCLUSIONS

Now we can state once more that the main goal of our paper is achieved: this is the proof that in the description of the process of photon absorption by atoms within crossing of atomic and laser beams the transit width plays the same role as the natural width and may be inserted in the Lorentz energy denominator for the resonant absorption. Our aim was to find out whether this insertion is always justified. In particular, it was important for the proposed experiment on the search for the electric dipole moment of the electron by observation of the Faraday optical rotation in an electric field.

The resolution of this problem is given by Eq. (30). Conditions necessary for the experiment to remain within the Lorentz picture of the spectral line profile look like $\Delta\omega\tau > 1$. Fortunately, these conditions are compatible with the detuning value $\Delta\omega \approx 5\Gamma_D$ considered as optimal in Refs. [2,3].

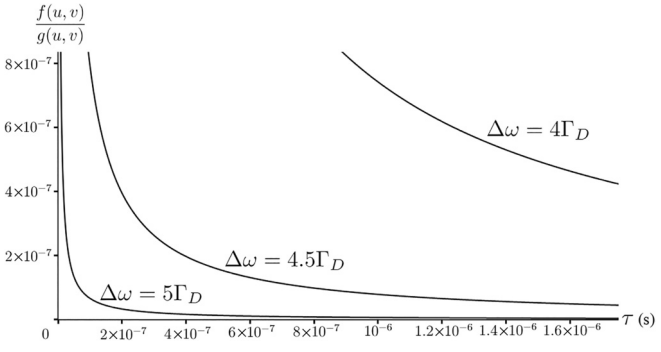

FIG. 4. Ratio absorption and dispersion as a function of transit time for different detunings.

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APPENDIX: EVALUATION OF THE INTEGRAL EQ. (22)

For evaluating the integral Eq. (22) we apply the Cauchy theorem, closing the contour of integration in the lower half plane of the complex variable ω' (see Fig. 5). Our scheme for calculating the integral is the following. First, we set $\delta = 0$, and then pole 1 is located on the real axis. Second, we integrate in the complex plane using the contour depicted in Fig. 5. This contour runs along the real axis surrounding pole 1 by a semicircle. This explains the arrival of the factor $1/2$ in the contribution of pole 1. Then

$$J = 2\pi i\left[\tfrac{1}{2}\mathrm{Res}_1 + \mathrm{Res}_2 + \mathrm{Res}_3\right], \tag{A1}$$

where Res_i $(i = 1, 2, 3)$ are the residues in the corresponding poles. The integral over the large half circle vanishes since it behaves like $\frac{1}{R^4}$, where $R = |\omega'|$, when $R \to \infty$. The poles are $\omega'_1 = -E_n + i\delta$, $\omega'_2 = -E_A - \omega_L - i\lambda$, and $\omega'_3 = -E_A - \omega - i\lambda$. The corresponding residues are

$$\mathrm{Res}_1 = \frac{\lambda^2}{\pi^2}\frac{1}{(E_n - E_A - \omega_L)^2 + \lambda^2}\frac{1}{(E_n - E_a - \omega)^2 + \lambda^2}, \tag{A2}$$

$$\mathrm{Res}_2 = \frac{\lambda^2}{\pi^2}\frac{1}{E_n - E_A - \omega_L - i\lambda} \times \left(-\frac{1}{2i\lambda}\right)\frac{1}{\omega - \omega_L - 2i\lambda}\frac{1}{\omega - \omega_L}, \tag{A3}$$

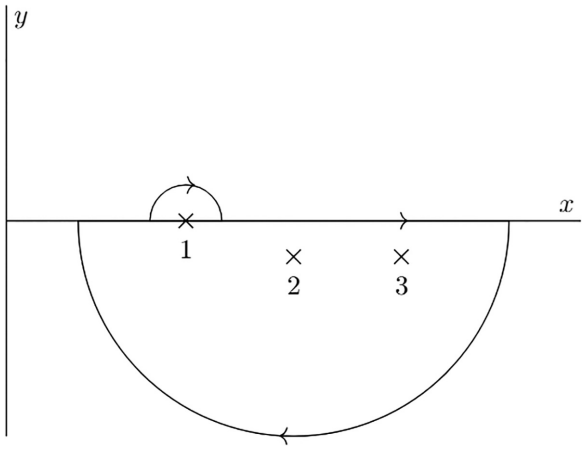

FIG. 5. Integration of Eq. (22) in the complex plane ω'. The poles are denoted by numbers 1, 2, and 3. The residue for pole 1 is located in the upper half plane, since the energy E_n of the resonant level n is positive.

$$\mathrm{Res}_3 = \frac{\lambda^2}{\pi^2}\frac{1}{E_n - E_A - \omega - i\lambda} \times \left(-\frac{1}{2i\lambda}\right)\frac{1}{\omega_L - \omega - 2i\lambda}\frac{1}{\omega_L - \omega}. \tag{A4}$$

From Eqs. (A1)–(A4) follows Eq. (29) in the main text.

Next we expand Eq. (29) in terms of the parameter $\frac{\omega-\omega_L}{\lambda}$ by a finite value of λ and $\omega \to \omega_L$ up to the first-order terms in $\frac{\omega-\omega_L}{\lambda}$. Then using Eq. (26) we find

$$I_\lambda = \left(\frac{\lambda^3}{[(E_n - E_A - \omega_L)^2 + \lambda^2]^2} - \frac{1}{E_n - E_A - \omega_L - i\lambda}\right), \tag{A5}$$

which coincides with Eq. (30) in the main text.

[1] V. Andreev, D. G. Ang, D. DeMille, J. M. Doyle, G. Gabrielse, J. Haefner, N. R. Hutzler, Z. Lasner, C. Meisenhelder, B. R. O'Leary *et al.* (ACME collaboration), Nature (London) **562**, 355 (2018).

[2] V. N. Kutuzov, D. V. Chubukov, L. V. Skripnikov, A. N. Petrov, and L. N. Labzowsky, Ann. Phys. (NY) **434**, 168591 (2021).

[3] S. D. Chekhovskoi, D. V. Chubukov, L. V. Skripnikov, A. N. Petrov, and L. N. Labzowsky, Phys. Rev. A **108**, 052819 (2023).

[4] D. Budker, D. F. Kimball, and D. P. DeMille, *Atomic Physics, An Exploration through Problems and Solutions*, 2nd ed. (Oxford University, New York, 2008).

[5] W. Demtröder, *Laser Spectroscopy, Vol. 1: Basic Principles*, 4th ed. (Springer-Verlag, Berlin, 2008).

[6] V. S. Letokhov, Zh. Eksp. Teor. Fiz. **56**, 1748 (1969) [Sov. Phys. JETP **29**, 937 (1969)].

[7] S. G. Rautian and A. M. Shalagin, Zh. Eksp. Teor. Fiz. Pis'ma Red. **9**, 686 (1969) [JETP Lett. **9**, 427 (1969)]; Zh. Eksp. Teor. Fiz. **58**, 962 (1970) [Sov. Phys. JETP **31**, 518 (1970)].

[8] J. L. Hall, in *Lectures in Theoretical Physics*, edited by W. E. Brittin and K. T. Mahanthappa (Gordon and Breach, New York, 1973), p. 161.

[9] C. J. Bordé and J. L. Hall, in *Laser Spectroscopy*, edited by R. G. Brewer and A. Mooradian (Plenum, New York, 1974).

[10] C. J. Bordé, J. L. Hall, C. V. Kunasz, and D. G. Hummer, Phys. Rev. A **14**, 236 (1976).

[11] D. V. Chubukov, I. A. Aleksandrov, L. V. Skripnikov, and A. N. Petrov, Phys. Rev. A **108**, 053103 (2023).

[12] W. H. Furry, Phys. Rev. **81**, 115 (1951).

[13] S. Schweber, *An Introduction to Relativistic Quantum Field Theory* (Row, Peterson, Evanston, 1961).

[14] P. J. Mohr, G. Plunien, and G. Soff, Phys. Rep. **293**, 227 (1998).

[15] A. I. Akhiezer and V. B. Berestetskii, *Quantum Electrodynamics* (Wiley, New York, 1965).

[16] M. Gell-Mann and F. Low, Phys. Rev. **84**, 350 (1951).

[17] J. Sucher, Phys. Rev. **107**, 1448 (1957).
[18] L. N. Labzowsky, G. L. Klimchirskaya, and Yu. Yu. Dmitriev, *Relativistic Effects in the Spectra of Atomic Systems*, 1st ed. (CRC, Boca Raton, FL, 1993).
[19] F. Low, Phys. Rev. **88**, 53 (1952).
[20] O. Yu. Andreev, L. N. Labzowsky, G. Plunien, and D. A. Solovyev, Phys. Rep. **455**, 135 (2007).
[21] A. Grinin, A. Matveev, D. C. Yost, L. Maisenbacher, V. Wirthl, R. Pohl, T. W. Hänsch, and T. Udem, Science **370**, 1061 (2020).
[22] I. B. Khriplovich, *Parity Nonconservation in Atomic Phenomena* (Gordon and Breach, New York, 1991).
[23] M. Abramowitz and I. A. Stegun, *Handbook of Mathematical Functions: With Formulas, Graphs, and Mathematical Tables* (NBS, USA, 1964).