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INVARIANT FORM OF COULOMB CORRECTIONS IN THE THEORY OF NONLINEAR IONIZATION OF ATOMS BY INTENSE LASER RADIATION

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Using the imaginary time method, a new formulation of Coulomb corrections to the amplitude of nonlinear ionization of atoms is given. The Coulomb corrections to the photoelectron action and trajectory are presented in the form independent of the integration path in the imaginary time plane. The obtained representation corrects the previously known results and shows that the subdivision of photoelectron motion into the sub-barrier and after-barrier parts is conditional and does not influence observables. The new correction is particularly relevant in the multiphoton regime of ionization.

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1. INTRODUCTION

The theory of nonlinear ionization of atoms by intense laser radiation originates in the seminal work of Keldysh [1], where an efficient nonperturbative approximation for the amplitude of ionization by an intense low-frequency electromagnetic field was formulated. The term "low-frequency field" means in this content that the ionization potential I_p of an atom is much greater than the photon energy $\hbar\omega$, i. e., the multiquantum parameter is large,

$$K_0 = I_p/\omega \gg 1. \tag{1}$$

Under this condition, ionization can only proceed via a nonlinear mechanism. The Keldysh ionization ansatz can be summarized as follows. In a strong laser field, the electron continuum states can be with reasonable accuracy approximated by Gordon–Volkov waves [2, 3], solutions of the Schrödinger equation (Klein–Gordon or Dirac equation in the relativistic case) for an electron in the field of a plane electromagnetic wave. If the laser field is strong enough, the interaction of a liberated electron with its parent ion can be disregarded in the zeroth approximation. On the other hand, in order to fully ionize a bound atomic level, the electric field strength $\mathcal{E}_0$ well below the characteristic electric field $\mathcal{E}_{ch}$ of this level is usually sufficient, and therefore the condition

$$F = \mathcal{E}_0/\mathcal{E}_{ch} \ll 1 \tag{2}$$

is satisfied for most of the cases. Here, the characteristic field is defined as

$$\mathcal{E}_{ch} = \frac{m^2}{e\hbar}\left(\frac{2I_p}{m}\right)^{3/2} \tag{3}$$

with m and e being the electron mass and the elementary charge. Under conditions (1) and (2), the influence of the laser field on the bound state can be disregarded, and the ionization amplitude can be presented in the form

$$A_0(\mathbf{p}) = -\frac{i}{\hbar}\int d^4x\, \Psi_{\mathbf{p}}^{*(V)}(x) V_{int}(x) \Psi_b(x), \tag{4}$$

where Ψ_b is the bound field-free atomic state, $\Psi_{\mathbf{p}}^{(V)}$ is the Volkov function corresponding to the asymptotic electron momentum equal to $\mathbf{p}$, and V_{int} is the electron–field interaction operator. Equation (4) gives the probability amplitude of nonlinear ionization at arbitrary values of the Keldysh parameter

$$\gamma = \frac{\sqrt{2mI_p}\,\omega}{e\mathcal{E}_0}, \tag{5}$$

where ω is the laser-field frequency.

The theoretical approach based on the above idea is known in the literature as the Keldysh theory or

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strong field approximation (SFA) [4, 5]. Over the time after publication of Keldysh's work [1], it was essentially developed and applied for description of a variety of strong-field phenomena. The present status of the Keldysh theory and SFA was reviewed in Refs. [6–8].

Amplitude (4) does not account for the electron–ion interaction in the continuum. For systems bound by short-range forces, e. g., negative ions, this interaction is small, but still causes observable effects. For a review of theoretical approaches to description of strong-field ionization of negative ions, we refer the reader to [9, 10] and the references therein. For atoms and molecules, the Coulomb force generates significant effects, whose description requires an essential modification of the theory. This was achieved by the introduction of Coulomb corrections into the phase of amplitude (4). Evaluation of these corrections is based on the imaginary time method (ITM) [11], which allows expressing amplitude (4) via the electron action in the field of a plane electromagnetic wave, calculated along a classical trajectory in complex time. In the early paper by Perelomov and Popov [12], the ITM was applied for calculation of the total ionization rate of atoms in the tunneling limit, $\gamma \ll 1$. It was shown there that the Coulomb field enhances the rate of ionization typically by several orders of magnitude. Later, this result was generalized to the case of arbitrary values of the Keldysh parameter [13] (assuming that inequality (1) is satisfied, however). Besides enhancing the total ionization rate, the Coulomb interaction was shown to generate several effects accessible for experimental observation, including the Coulomb asymmetry in elliptically polarized fields [14–16], cusps and double-hump structures [17–19], low-energy structures [20–23], and side lobes [24] in momentum spectra of photoelectrons.

Currently, the method of Coulomb corrections in the theory of strong-field ionization is well developed. This includes classical trajectory simulations, a relativistic version of the Keldysh theory, the trajectory-based SFA, and other approaches. For details, we refer to [6, 8, 25] and the references therein. The aim of this paper is to address one controversial issue inherent to all the above-mentioned methods of evaluation of Coulomb corrections. Namely, the calculation procedure involves the photoelectron tunnel exit — a spatial point where the electron appears in real time and space after ionization. In purely classical simulations (e. g., in Refs.[15, 21, 23]), the tunnel exit is a starting point for a calculation, and the influence of the electron–ion interaction on the electron dynamics before the electron appears at the exit is not considered. In quantum mechanical calculations, including the ITM, both sub-barrier and after-barrier motion of the electron are taken into account. The sub-barrier motion mostly yields the imaginary part of the action and influences the absolute value of the ionization probability. Photoelectron motion after the barrier proceeds in real time and space and influences the real part of the action, and therefore the interference structure of photoelectron spectra. As a result, the tunnel exit enters Coulomb corrections to the photoelectron action. On the other hand, the position of the tunnel exit is not an observable, and hence it must not influence momentum distributions.

The question therefore arises: is it possible to formulate the method of Coulomb corrections in a form that does not involve the tunnel exit, but only depends on the observables of the problem? Such a formulation is given in this paper. It is shown that the Coulomb correction to the photoelectron momentum can be presented in the form of a converging integral in the complex time plane, which only depends on the momentum itself. The integration paths must be chosen taking the analyticity properties of the Coulomb interaction energy in complex space into account. It is then shown that the obtained Coulomb correction reproduces the previously known result in the tunneling regime $\gamma \ll 1$, but this is not the case for $\gamma \gg 1$.

This paper is organized as follows. In Sec. 2, we introduce basic equations and briefly describe the standard approach to the calculation of Coulomb corrections. In Sec. 3, we derive an invariant form of the Coulomb correction to the photoelectron momentum, which does not involve the tunnel exit position. The choice of the integration path in the complex time plane is then discussed. The last section contains conclusions.

2. BASIC EQUATIONS

Using the ITM, ionization amplitude (4) can be represented in the form (here and hereafter, atomic units $\hbar = m = e$ are used) [11, 12]

$$A_0(\mathbf{p}) \propto \exp(iW_0(\mathbf{p}, t_s, T)), \tag{6}$$

where W_0 is the reduced electron action in the laser field $\boldsymbol{\mathcal{E}}(t)$,

$$W_0(\mathbf{p}, t_s(\mathbf{p}), T) = \\ = \int_{t_s}^{T} \left\{ \frac{1}{2}\mathbf{v}_0^2 - \boldsymbol{\mathcal{E}}(t)\cdot\mathbf{r}_0 - I_p \right\} dt - \mathbf{r}_0\cdot\mathbf{v}_0 \Big|_{t_s}^{T}, \tag{7}$$

and the trajectory $\mathbf{r}_0(t)$ satisfies the Newton equation

$$\ddot{\mathbf{r}}_0 = -\boldsymbol{\mathcal{E}}(t), \tag{8}$$

with the initial and final conditions

$$\mathbf{v}_0^2(t_s) = -2I_p, \quad \mathbf{r}_0(t_s) = 0, \quad \mathbf{v}_0(T) = \mathbf{p}. \tag{9}$$

Here, $T \to \infty$ is the time instant when the electron, having the velocity $\mathbf{v}_0$ and momentum $\mathbf{p}$ is observed at the detector. A preexponential factor not important for our purposes is omitted in (6). Its particular form is determined by the initial-state wave function [6, 7, 26]. The first equation in (9) shows that the initial time t_s of electron motion is always complex, while the second equation assumes that before the ionization event, the electron was confined in the atom. Introducing the laser field vector potential such that $\boldsymbol{\mathcal{E}}(t) = -\dot{\mathbf{A}}(t)$, we can represent the first equation in (9) in the form

$$[\mathbf{p} + \mathbf{A}(t_s)]^2 = -2I_p, \tag{10}$$

which determines a complex saddle point $t_s(\mathbf{p})$. The ITM equations provide us with a physically appealing picture of ionization: the electron starts from the origin at a complex time instant $t_s = t_0 + i\tau_0$, having an imaginary initial velocity $v_0(t_s) = \pm i\sqrt{2I_p}$. As time arrives to the real axis, $t = t_0$, the velocity also becomes real. The electron coordinate $\mathbf{b} \equiv \mathbf{r}_0(t_0)$ is also real for the most probable trajectory that minimizes the imaginary part of action (7). This point $\mathbf{b}$ is interpreted as the tunnel exit. For a linearly polarized monochromatic field

$$\boldsymbol{\mathcal{E}}(t) = \boldsymbol{\mathcal{E}}_0 \cos\varphi, \quad \varphi = \omega t, \tag{11}$$

we obtain

$$\begin{aligned} \mathbf{v}_0(\varphi) &= \mathbf{p} - \frac{\boldsymbol{\mathcal{E}}_0}{\omega}\sin\varphi, \\ \mathbf{r}_0(\varphi) &= \frac{\mathbf{p}}{\omega}(\varphi - \varphi_s) + \frac{\boldsymbol{\mathcal{E}}_0}{\omega^2}(\cos\varphi - \cos\varphi_s). \end{aligned} \tag{12}$$

For the most probable trajectory $\mathbf{p} = 0$, $\varphi_s = i\,\mathrm{Arcsh}\,\gamma$, and the tunnel exit point is given by

$$b = \frac{\mathcal{E}_0}{\omega^2}\left(\sqrt{1+\gamma^2} - 1\right). \tag{13}$$

In the tunneling limit $\gamma \ll 1$, this gives the standard potential barrier width in a static field $b = I_p/\mathcal{E}_0$; in the opposite multiphoton regime, $b = \sqrt{2I_p}/\omega$.

The procedure introducing Coulomb corrections to amplitude (6) is as follows [13, 25].

1. The Coulomb-free trajectory is replaced by a corrected one:

$$\mathbf{v}_0(t) = \mathbf{p} + \mathbf{A}(t) \to \mathbf{v}_0(t) + \mathbf{v}_1(t), \quad \dot{\mathbf{v}}_1 = -\frac{Z\mathbf{r}_0}{r_0^3}, \tag{14}$$

where Z is the atomic residual charge ($Z = 1$ for ionization of neutral atoms and $Z = 0$ for negative ions).

2. The photoelectron momentum is no longer conserved, and therefore its value $\mathbf{p}'$ at the tunnel exit is different from the one measured by a detector. Thus, the initial drift momentum is to be found from

$$\mathbf{v}(T) = \mathbf{p}' + \mathbf{v}_1(T) = \mathbf{p}. \tag{15}$$

3. The corresponding saddle point $t_s' = t_s(\mathbf{p}')$ is calculated from the same saddle-point equation (10) with $\mathbf{p}'$ instead of $\mathbf{p}$. The momentum $\mathbf{p}'$ is defined such that $\mathbf{v}_1(t_0) = 0$.

4. The Coulomb interaction $U_C = -Z/r$ is added to the action.

The Coulomb-corrected ionization amplitude is

$$\begin{aligned} A(\mathbf{p}) &\sim \exp(iW(\mathbf{p}, t_s', T)), \\ W &= \int_{t_s'}^{T} \left\{\frac{1}{2}\mathbf{v}^2 - \boldsymbol{\mathcal{E}}(t)\cdot\mathbf{r} + \frac{Z}{r} - I_p\right\} dt - \mathbf{r}\cdot\mathbf{v}\Big|_{t_s'}^{T}. \end{aligned} \tag{16}$$

Taking into account that the correction $\mathbf{v}_1$ in (14) is found perturbatively and assuming that the Coulomb perturbation is small compared to the value of the field-induced action W_0, we can keep only contributions linear in the charge Z in (16). We note that being small compared to the action W_0, Coulomb corrections are usually much greater than unity in absolute value; this determines their significant effect on the ionization probability and momentum distributions.

The first-order expansion of action (16) yields

$$\begin{aligned} W(\mathbf{p}, t_s', T) \approx W_0(\mathbf{p}', t_s', T) + \\ + \int_{t_s}^{T} \frac{Z}{r_0(\tau)} d\tau - \mathbf{r}_0\cdot\mathbf{v}_1\Big|_{t_s}^{T}. \end{aligned} \tag{17}$$

The first term is the Coulomb-free action calculated along the new trajectory, and the second is the Coulomb action calculated along the Coulomb-free trajectory. The last term originates from the correction to the photoelectron trajectory and has a finite nonzero value at the lower limit.

The Coulomb integral

$$W_C = \int_{t_s}^{T} \frac{Z}{r_0(\tau)} d\tau \tag{18}$$

is logarithmically divergent at the lower limit and requires regularization. It is performed by replacing $t_s \to t_*$ (Fig. 1) and matching the result of integration to the asymptotic form of the atomic bound-state

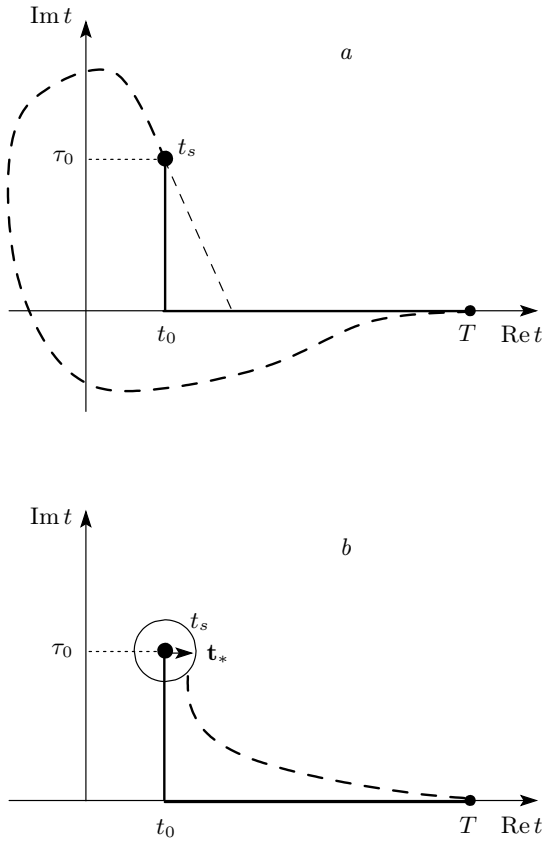

Fig. 1. Complex time plane with the saddle point $t_s = t_0 + i\tau_0$: (a) the standard integration path connecting the saddle point with the real time of observation T is shown by a solid line. Other possible paths giving the same result for W_0 are shown by dashed lines; (b) the circle of the radius $|t_* - t_s|$ characterizing the matching region. In the tunneling regime, the radius of this curve is greater than τ_0, and hence the real part of the stationary point can be chosen as a starting point for integration in Eq. (21)

wave function [11–13]. The matching point satisfies the condition

$$1/K_0 \ll \omega|t_* - t_s| \ll 1. \tag{19}$$

The regularized result has the form [12]

$$W_C = -in_* \ln(2iK_0T) + \int_{t_s}^{T} \left\{ \frac{Z}{\sqrt{r_0^2(t)}} + \frac{in_*}{t - t_s} \right\} dt, \tag{20}$$

where the effective principal quantum number $n_* = Z/\sqrt{2I_p}$ is introduced. The integration path in (20) is usually chosen as shown in Fig. 1a by a solid line: from the stationary point t_s to its projection t_0 on the real axis and then along the real axis. The respective contributions can be interpreted as the sub-barrier and after-barrier corrections. For the most probable photoelectron momentum, the first integral is imaginary and gives the Coulomb correction to the rate of ionization [12, 13], while the second is real and corrects the phase of the probability amplitude and thus the interference structure of momentum distributions [25]. For other final momenta, both sub-barrier and after-barrier corrections respectively acquire a real and an imaginary part. The sub-barrier correction for arbitrary final momenta was first calculated in Ref. [27]. It was shown that taking it into account essentially corrects the interference structure of momentum distributions, improving their agreement with results of *ab-initio* numerical solutions of the time-dependent Schrödinger equation. For circularly polarized fields, this phase correction was analyzed in Refs. [28–31].

To calculate the first term in (17), we must find the renormalized momentum $\mathbf{p}'(\mathbf{p})$. This calculation meets a principal difficulty. In the first order with respect to the Coulomb force,

$$\mathbf{v}_1 = -Z \int_{t_s}^{T} \frac{\mathbf{r}_0}{r_0^3}\, dt. \tag{21}$$

This integral is divergent at the lower integration limit, and a similar problem appears with differential equation (14), because the particle starts at the origin where the Coulomb force is divergent. A commonly accepted way of avoiding this difficulty is to start integration from the real time instant t_0 when the electron is at the tunnel exit $\mathbf{b} = \mathbf{r}_0(t_0)$. In other words, the influence of the Coulomb force on the photoelectron momentum is taken into account only after the barrier. To the best of our knowledge, this ansatz was used in all the works with calculations of Coulomb corrections to photoelectron trajectories. This approach yields the high-frequency correction to the rate of ionization [13] and allows reproducing the Coulomb asymmetry in elliptically polarized fields [15, 16], the low-energy structure [21, 22], and other effects in photoionization spectra. In the recent series of works [32–34], this method for calculating the Coulomb-induced correction to the photoelectron final momentum was used for quantitative description of experiments on attosecond streaking.

Despite good agreement with experimental data and exact numerical solutions, the above-described approach to the calculation of the final momentum is obviously controversial. The tunnel exit is not a physically distinguished spatial point and nothing special happens

to the electron there. Moreover, because laser-induced action (7) is an analytic function of the complex time, any integration path connecting the points t_s and T, can be chosen, such that the time instant when the electron velocity becomes real is arbitrary (see the illustration in Fig. 1 *a*). Hence, the starting point for integration in real time cannot be defined in the theory. As a result, any proper expression for the Coulomb-corrected action must involve not t_0 but only the saddle point $t_s(\mathbf{p})$. The questions then occur: (i) how to make integral (21) meaningful and path-independent, and (ii) why do the previously obtained results based on an incorrect regularization of (21) yield good agreement with the data? These questions are answered in the next section, where an invariant expression for the Coulomb correction to the final photoelectron momentum is derived.

3. INVARIANT FORM OF THE COULOMB CORRECTION

Coulomb correction (20) is already presented in the form independent of the integration path, if this path does not intersect cuts of the function $1/r_0(t)$. Positions of the branch points and the corresponding cuts are discussed at the end of this section.

To obtain a finite expression for the Coulomb-corrected momentum, we consider Eq. (14) at a time t_* that satisfies (19). Under this condition, the electron is far from the atom, and hence the Coulomb force is small compared with the laser one, but its excursion is still small compare to the electron quiver amplitude in the laser field, $\mathcal{E}_0/\omega^2$. Then $\mathbf{r}_0$ from (12) can be expanded in a series in $t - t_s$:

$$\mathbf{r}_0(t) \approx [\mathbf{p} + \mathbf{A}(t_s)](t - t_s) - \frac{1}{2}\boldsymbol{\mathcal{E}}(t_s)(t - t_s)^2.$$

Omitting the terms that are small under condition (19), after a simple algebra, we obtain

$$\mathbf{v}_1 = \int_{t_s}^{T} \left\{ -\frac{Z\mathbf{r}_0(t)}{r_0^3(t)} + \mathbf{f}(t - t_s) \right\} dt + \\ + iZF\mathbf{g}(\mathbf{p}, \gamma) \ln(T), \quad (22)$$

where

$$\mathbf{f} = iZF\omega \left\{ \frac{\mathbf{q} + \mathbf{a}}{(\varphi - \varphi_s)^2} - \frac{\mathbf{g}(\mathbf{q}, \varphi_s)}{\varphi - \varphi_s} \right\} \quad (23)$$

and

$$\mathbf{g} = \frac{1}{2}\left[\mathbf{e} + \frac{3}{\gamma^2}(\mathbf{q} + \mathbf{a})(\mathbf{e} \cdot (\mathbf{q} + \mathbf{a})) \right] \cos\varphi_s. \quad (24)$$

Here, the dimensionless quantities are $\mathbf{q} = \mathbf{p}/p_F$ and $\mathbf{a} = \mathbf{A}(t_s)/p_F$, with the field-induced momentum $p_F = \mathcal{E}_0/\omega$ and the unit vector $\mathbf{e}$ along the polarization direction. The integral in (22) converges and its value is independent of the integration path if it does not intersect the cuts of the function $\mathbf{r}_0/r_0^3$. The obtained value of $\mathbf{v}_1$ corrects the initial complex photoelectron velocity, such that

$$\mathbf{v}(t_s) = \mathbf{p} + \mathbf{A}(t_s') - \mathbf{v}_1 \equiv \mathbf{p}' + \mathbf{A}(t_s'). \quad (25)$$

Equations (22)–(25) define the action in (17) and thus the ionization amplitude and present the main result in the paper. They improve the previously accepted equation (21) for the Coulomb corrected photoelectron momentum.

Unlike laser-induced action (7), Coulomb integral (18) is not an analytic function in the whole complex plane. The analytic function $\mathbf{r}_0^2$ from (12) in general has an infinite number of first-order zeros, which generate corresponding branch points and cuts of the functions $1/\mathbf{r}_0$ and $\mathbf{r}_0/r_0^3$. A proper integration path in Eqs. (20) and (22) should connect t_s and T without intersecting the cuts. A map of cuts is shown on Fig. 2 for some typical set of parameters. The roots of the equation

$$\mathbf{r}_0^2(t_n) = 0 \quad (26)$$

come in pairs, which merge for the electron momenta parallel to the polarization axis. In this special case, the t_n are second-order zeros, like the saddle point t_s always is. Then the Coulomb potential energy has no branch points but first-order poles only. The presence of even a small lateral momentum component converts these poles into pairs of branch points, which recede from another with as $p_\perp$ increases. Cuts are defined as the lines in the complex time plane where $\mathbf{r}_0^2 < 0$. For trajectory (12), their asymptotic forms are given by

$$\varphi_n = \frac{\pi}{2} + \pi n, \quad n = 0, 1, 2, \ldots \quad (27)$$

As is seen from Fig. 2, it is possible to connect the time instants t_s and T by a curve avoiding the cuts. A more detailed study shows that this is always the case. This curve may even coincide with the standard path (solid curve in Fig. 1) associated with the tunnel exit, but not for all final momenta: for small longitudinal and large perpendicular momenta, a branch point approaches this line (see Fig. 2 *b*); for $p_x = 0$, it is clearly seen that it lies exactly between t_s and t_0. Indeed, the equation

$$\frac{p_\perp \omega}{\mathcal{E}_0}(\psi - \psi_0) = \operatorname{ch}\psi - \operatorname{ch}\psi_0$$

always has a solution $0 < \psi_b < \psi_0$ if $p_\perp$ is sufficiently large or if $\gamma \ll 1$.

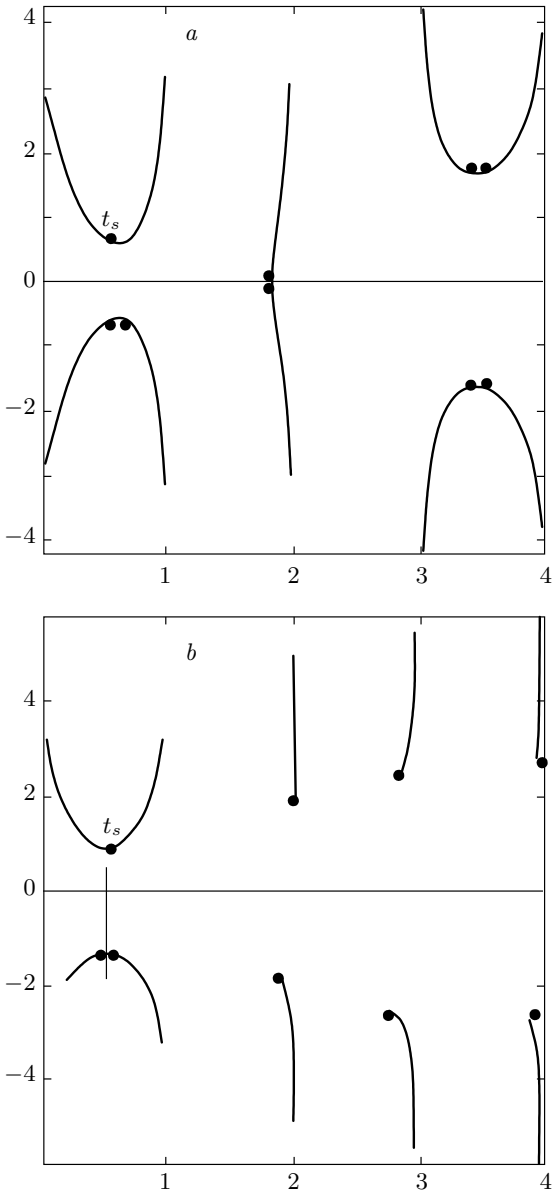

Fig. 2. Branch points (solid dots) and cuts (lines) of the function $1/r_0(t)$ in the dimensionless complex time plane for emission at 0.1 rad (a) to the polarization axis and (b) in the perpendicular direction. The parameters are $\mathcal{E}_0 = \omega = 0.05$, $p = 1.0$. Branch points come in pairs. The stationary point t_s is a first-order pole

4. CONCLUSIONS

To conclude, we have derived a new expression for the Coulomb-corrected photoelectron momentum for a strong field ionization of atoms. The main advantage of Eqs. (22)–(24) compared to the previously known result is that their form is invariant with respect to the choice of the integration path, i. e., the Coulomb-corrected action remains a function of a complex variable as it was in the Coulomb-free theory. In the new formulation, the tunnel exit does not play the role of a physically significant spatial point.

For their applicability, Eqs. (22)–(24) require the condition

$$4K_0^2 F \gg 1, \tag{28}$$

which follows from (19). It is equivalent to $2K_0 \gg \gamma$ and imposes an upper bound on the Keldysh parameter.

The question may arise: why do the calculations based on the meaningless regularized form of expression (21) with $t_s \to t_0$ in many cases yield good agreement with experimental data and numerical results? The reason becomes clear if we compare the barrier width with the distance between the electron and the ion at the matching time instant t_*. In the tunneling regime, the two length parameters $r_0(t_*)$ and b are of the same order, and therefore the tunnel exit can be taken as a starting point for electron motion, as can be any other spatial point from the matching circle shown in Fig. 1*b*. In the opposite limit $\gamma \gg 1$, we have $r_0(t_*) \ll b$ because $b \gg \mathcal{E}_0/\omega^2$. Then the integral in (22) contains an essential "sub-barrier" contribution, omitted in the standard expression for the Coulomb-corrected momentum. Thus, our results are virtually identical to the previously known ones in the regime of tunneling and provide new expressions for the Coulomb-corrected momentum and action in the multiphoton limit.

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