Application of the gauge-invariant formalism to the description of ionization by ultrastrong few-cycle pulses

Adam Bechler* and M. Ślęczka
Institute of Physics, University of Szczecin, Wielkopolska 15, 70-451 Szczecin, Poland
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We analyze energy spectra of photoelectrons ejected by strong laser pulses, using the manifestly gauge-invariant approach. Gauge-invariant expressions for the ionization amplitude can be obtained by grouping together all terms of the same order in atomic potential in the standard strong-field approximation (SFA) expansion. We calculate energy spectra of photoelectrons for the hydrogen atom, and compare them with results based on numerical solutions of the time-dependent Schrödinger equation. Agreement, at least qualitative, is observed for an electric-field amplitude higher than 1 a.u., and for larger energies of ionized electrons, basically beyond the region, where the standard SFA is usually applied.

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The purpose of this Brief Report is the gauge-invariant description of the ionization of an atom by strong and ultrashort pulses of electromagnetic radiation. The formalism which we explore here was proposed long ago [1] but, to our knowledge, has not been extensively used as a theoretical tool for the description of strong-field ionization in realistic three-dimensional situations. Instead, it was applied to the ionization of one-dimensional model atoms with short-range delta-like potentials [1]. It has been also used to discuss bound-state stabilization through short ultraintense fields [2]. In this Brief Report we present an attempt to use the gauge-invariant approach to calculate the energy spectra of photoelectrons ejected by strong pulses from a hydrogen atom. We also compare the calculations with energy spectra obtained by solving numerically the time-dependent Schrödinger equation (TDSE). Regarding the pulse parameters the main finding is that the present semianalytic gauge-invariant formalism leads to good agreement with TDSE results for very intense pulses with a maximum electric field of the order of 1 a.u. or higher, and for shorter carrier wavelengths of the order of 100 nm or shorter. The agreement is also better of higher photoelectron energies.

Standard semianalytic description of the interaction of strong electromagnetic radiation with matter is based on the strong-field approximation (SFA) [3–5]. Although it constitutes a commonly used well-established theoretical tool, it by no means can be considered as unambiguous. Ambiguity of SFA is due to its dependence on the choice of gauge of electromagnetic potentials of the pulse.

In the case of the two most popular gauges, the velocity gauge (VG) and length gauge (LG), theoretical predictions may differ significantly [6–9]. In general, the length gauge is favored since in many cases it leads to better agreement with experimental results [10–12]. It has been, however, argued by Reiss [8] that the velocity gauge gives better agreement with the results of measurements in the case of detachment from negative fluorine ions [10]. It has been also argued by Cormier and Lambropoulos [13] that "for dynamical reasons" the velocity gauge is more suitable for the description of atomic processes in strong fields. The typical parameters for which SFA shows good agreement with TDSE results should fulfill two conditions: $\omega \ll I_p$ and $\omega \ll U_p$, where ω is the carrier-wave frequency, I_p is the binding potential, and U_p is the ponderomotive energy. Strong gauge dependence of the Keldysh approximation was already noted by Neto and Davidovich [1]. They showed that including corrections to this approximation cancels the standard Keldysh contribution and leaves gauge-invariant expansion in powers of the atomic potentials, which is convergent if the potential is represented by a bounded operator. A related approach was also presented recently [14], where numerical tests were also performed for a one-dimensional model system.

The approach used here is closely related to that of Neto and Davidovich and is based on the appropriate grouping of all terms of the same order in atomic potential in the standard SFA expansion [15]. It can be shown that the gauge-dependent part of the "direct ionization" amplitude is canceled when the next iteration, corresponding to the first "rescattering" correction, is included. In effect, one obtains an expression for the amplitude, independent of the choice of gauge, which contains a zero-order term with no explicit dependence on the atomic potential, and the first-order contribution linear in the potential plus gauge-dependent contribution of the second order in atomic potential. This gauge-dependent term is again canceled when the rescattering correction of second order in atomic potential is accounted for. The iterative procedure leads then to an expression which is manifestly gauge independent order by order [15]. For the sake of completeness we briefly outline the main points of the gauge-invariant formalism.

The time-dependent Hamiltonian describing the interaction of matter with a strong external field can be partitioned in two ways:

$$\hat{H}(t) = \hat{H}_{\rm at} + \hat{F}(t) = \hat{H}_F(t) + \hat{V}_{\rm at}, \tag{1}$$

where

$$\hat{H}_{\rm at} = \hat{H}_0 + \hat{V}_{\rm at}, \tag{2a}$$

$$\hat{H}_F = \hat{H}_0 + \hat{F}(t). \tag{2b}$$

Here $\hat{H}_0 = \hat{\mathbf{p}}^2/2$ is the free Hamiltonian, $\hat{V}_{\rm at}$ denotes the time-independent atomic potential, and the time-dependent part $\hat{F}(t)$ describes the interaction of the atomic electron with

*Corresponding author: adamb@univ.szczecin.pl

a strong external field. The most commonly used length gauge is characterized by $\hat{F}_L(t) = \mathbf{r}\cdot\mathbf{E}(t)$, where $\mathbf{E}(t)$ denotes the electric field of the pulse and is defined by $\mathbf{E}(t) = -\partial\mathbf{A}(t)/\partial t$. In the velocity gauge, $\hat{F}_V(t) = \hat{\mathbf{p}}\cdot\mathbf{A}(t) + (1/2)\mathbf{A}^2(t)$, where $\mathbf{A}(t)$ is the vector potential of the external field. Coordinate dependence is suppressed in the dipole approximation.

With the use of this partitioning of the full Hamiltonian, one obtains two types of integral Dyson equations for the time evolution operator [16]:

$$\hat{U}(t,t') = \hat{U}_{\rm at}(t,t') - i\int_{t'}^{t} dt_1 \hat{U}(t,t_1)\hat{F}(t_1)\hat{U}_{\rm at}(t_1,t'), \quad (3a)$$

$$\hat{U}(t,t') = \hat{U}_F(t,t') - i\int_{t'}^{t} dt_1 \hat{U}(t,t_1)\hat{V}_{\rm at}\hat{U}_F(t_1,t'), \quad (3b)$$

where $\hat{U}_{\rm at}$ and $\hat{U}_F$ are time evolution operators generated by $\hat{H}_{\rm at}$ and $\hat{H}_F$ (1), respectively.

The time-dependent transition amplitude can be written as

$$M(t,t') = \langle\psi_f(t_f)|\hat{U}(t_f,t_i)|\psi_i(t_i)\rangle, \quad (4)$$

with $|\psi_f\rangle$ and $|\psi_i\rangle$ denoting, respectively, the final continuum state of the ejected electron, and the initial bound state, both fulfilling the Schrödinger equation with the atomic Hamiltonian $\hat{H}_{\rm at}$; t_i is the time when we switch on the external field and t_f when we switch it off. The exact transition amplitude, calculated with the use of Eq. (3a), can be written in the form

$$M_{fi} = -i\int_{t_i}^{t_f} dt_1 \langle\psi_f(t_f)|\hat{U}(t_f,t_1)\hat{F}(t_1)|\psi_i(t_1)\rangle, \quad (5)$$

where the relation $\hat{U}_{\rm at}(t_1,t_i)|\psi_i(t_i)\rangle = |\psi_i(t_1)\rangle$ has been used. This expression for the amplitude is gauge independent up to a phase factor. The standard form of the SFA is realized by approximating $\hat{U}(t_f,t_1)$ by $\hat{U}_F(t_f,t_1)$ in Eq. (5), i.e.,

$$M_{fi} \approx M_0 = -i\int_{t_i}^{t_f} dt_1 \langle\psi_f^a(t_f)|\hat{U}_F(t_f,t_1)\hat{F}(t_1)|\psi_i(t_1)\rangle, \quad (6)$$

where the final state is approximated by a plane wave, so that $\langle\psi_f^a(t_f)|\hat{U}_F(t_f,t_1)$ is the Volkov state. This type of approximation is justified better for the detachment from negative ions, when, due to the short-range character of the atomic potential, the final electron can be a good approximation treated as "free." In the case of ionization from a neutral atom, one should, in principle, take into account the distortion of the plane wave by a long-range tail of the residual ion potential, especially for a slow ejected electron. Whereas the exact amplitude (4) is independent of the choice of gauge, the approximated expression (6) does not possess this property. Using the equation fulfilled by $\hat{U}_F$,

$$-i(\partial/\partial t_1)\hat{U}_F(t_f,t_1) = \hat{U}_F(t_f,t_1)[\hat{H}_0 + \hat{F}(t_1)], \quad (7)$$

and performing integration by parts we can rewrite expression (6) in the form

$$M_0 = \langle\psi_f^a(t_f)|\hat{U}_F(t_f,t_i)|\psi_i(t_i)\rangle - \langle\psi_f^a(t_f)|\psi_i(t_f)\rangle - i\int_{t_i}^{t_f} dt_1 \langle\psi_f^a(t_f)|\hat{U}_F(t_f,t_1)\hat{V}_{\rm at}|\psi_i(t_1)\rangle. \quad (8)$$

The second term in Eq. (8) will be absent if no approximation for the final state is used. The transition amplitude of Eq. (8) can be written in the new gauge by implementation of the unitary transformation $\hat{u}(\mathbf{r},t) = \exp[i\chi_g(\hat{\mathbf{r}},t)]$, where χ_g consistent with dipole approximation can be at most linear in $\mathbf{r}$. Denoting the transformed vector potential, scalar potential, and evolution operator by $\mathbf{A}_g$, φ_g, and $\hat{U}_g(t,t')$, respectively, we have

$$\mathbf{A}_g = \mathbf{A} - \nabla\chi_g, \quad \varphi_g = \varphi + \frac{\partial\chi_g}{\partial t}, \qquad \hat{U}_g(t,t') = e^{i\chi_g(\mathbf{r},t)}\hat{U}(t,t')e^{-i\chi_g(\mathbf{r},t')}. \quad (9)$$

The transformed Hamiltonian reads as

$$\hat{H}_g(t) = e^{i\chi_g(\mathbf{r},t)}\hat{H}(t)e^{-i\chi_g(\mathbf{r},t)} - \frac{\partial\chi_g(\mathbf{r},t)}{\partial t}. \quad (10)$$

The transformed interaction Hamiltonian is given by

$$\hat{F}_g(t) = \hat{F}(t) - (1/2)(\hat{\mathbf{p}}\cdot\nabla\chi_g + \nabla\chi_g\cdot\hat{\mathbf{p}}) + (1/2)(\nabla\chi_g)^2 + i[\chi_g,\hat{F}(t)]. \quad (11)$$

Assuming that $\chi(\mathbf{r},t)$ vanishes for $t < t_i$ and $t > t_f$ one can see that first two terms in Eq. (8) are independent of the choice of gauge, whereas the third one is not, since $\hat{U}(t_f,t_1)$ depends on $\chi_g(t_1)$ for transient times.

A procedure to obtain the gauge-invariant formulation of the Keldysh approximation, which will be sketched below, is closely related to that of Neto and Davidovich [1], and Milonni [17]. To obtain a gauge-invariant version of Eq. (8) we consider the next iteration of Eq. (3a) [15], i.e.,

$$M_1 = (-i)^2\int_{t_i}^{t_f} dt_1\int_{t_1}^{t_f} dt_2 \langle\psi_f^a(t_f)|\hat{U}_F(t_f,t_2) \times \hat{V}_{\rm at}\hat{U}_F(t_2,t_1)\hat{F}(t_1)|\psi_i(t_1)\rangle. \quad (12)$$

Changing the order of integration, using Eq. (7), and integrating by parts, we get

$$M_1 = i\int_{t_i}^{t_f} dt_2 \langle\psi_f^a(t_f)|\hat{U}_F(t_f,t_2)\hat{V}_{\rm at}|\psi_i(t_2)\rangle - i\int_{t_i}^{t_f} dt_2 \langle\psi_f^a(t_f)|\hat{U}_F(t_f,t_2)\hat{V}_{\rm at}\hat{U}_F(t_2,t_i)|\psi_i(t_i)\rangle + (-i)^2\int_{t_i}^{t_f} dt_2\int_{t_i}^{t_2} dt_1 \langle\psi_f^a(t_f)|\hat{U}_F(t_f,t_2)\hat{V}_{\rm at}\hat{U}_F(t_2,t_1) \times \hat{V}_{\rm at}|\psi_i(t_1)\rangle. \quad (13)$$

In the sum of M_0 [Eq. (8)] and M_1, the gauge-dependent contributions of the first order in atomic potential [i.e., the third term in Eq. (8) and first term in Eq. (13)] cancel each other and then, up to first order in the atomic potential, $M \approx M^{(0)} + M^{(1)}$, where

$$M^{(0)} = \langle\psi_f^a(t_f)|\hat{U}_F(t_f,t_i)|\psi_i(t_i)\rangle - \langle\psi_f^a(t_f)|\psi_i(t_f)\rangle, \quad (14a)$$

$$M^{(1)} = -i\int_{t_i}^{t_f} dt_1 \langle\psi_f^a(t_f)|\hat{U}_F(t_f,t_1)\hat{V}_{\rm at}\hat{U}_F(t_1,t_i)|\psi_i(t_i)\rangle. \quad (14b)$$

It can be easily seen that both $M^{(0)}$ and $M^{(1)}$ are independent of the choice of gauge, whereas the third, gauge-dependent contribution to the right-hand side of Eq. (13) is of higher order in the atomic potential and, as such, has to be combined with the next iteration of Eq. (3a). However, in a similar way

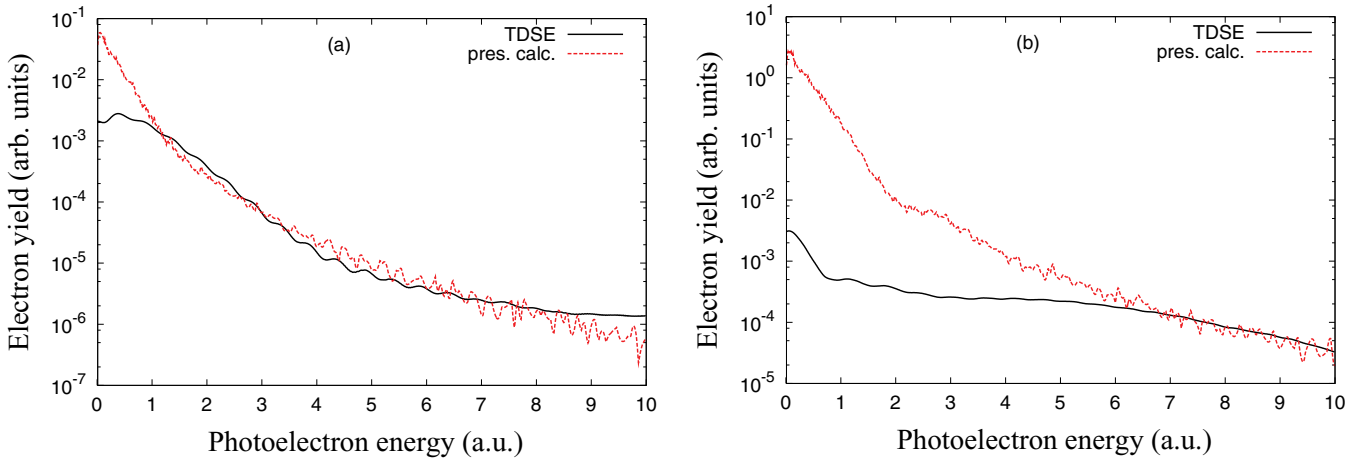

FIG. 1. (Color online) Energy distribution of photoelectrons for two-cycle laser pulses with $\omega = 0.57$ a.u., corresponding to a wavelength of 80 nm, and with (a) $E_0 = 3$ a.u. and (b) $E_0 = 10$ a.u. Solid black lines corresponds to TDSE results, dashed red lines corresponds to present calculations. The results were scaled to fit TDSE for photoelectron energy equal to 8 a.u.

as in the case of M_0 [Eq. (8)] and M_1 [Eq. (12)], this term will be canceled by an analogous boundary term appearing from integration by part. It should be also noted that we can obtain the equivalent form of transition amplitude expressed by Eq. (1) using directly the second form for the time evolution operator (3b).

We analyze the dependence of electron spectra, calculated with the use of Eq. (1), on the laser-field parameters and photoelectron energy. We consider a hydrogen atom initially in the ground state ionized by an intense linearly polarized laser pulse characterized by an electric field with a $\cos^2$-type envelope:

$$\mathbf{E}(t) = \begin{cases} \mathbf{e}E_0(t)\cos(\omega t + \varphi)\cos^2\left(\frac{\pi t}{\tau}\right) & \text{for} \quad -\frac{\tau}{2} < t < \frac{\tau}{2}, \\ 0 & \text{otherwise}, \end{cases} \tag{15}$$

where E_0 is the electric-field amplitude, τ is the pulse duration time, and φ is the carrier-envelope phase (CEP). The pulse duration time is $\tau = nT$, where n is the number of optical cycles and $T = 2\pi/\omega$ where ω is the carrier-wave frequency. Energy distributions of photoelectrons for a few values of electric-field strength and for n equal to 2 and 10 were compared with the numerical solution of the time-dependent Schrödinger equation. We consider an H-atom interacting with a pulse with $\omega = 0.5$ a.u., corresponding to carrier-wave length $\lambda \approx 91$ nm, and with CEP equal to zero. The Keldysh parameter is smaller than unity (from 0.05 to 0.5) for cases considered here, which suggests domination of the tunneling ionization. One has to remember, however, that we consider here a very short pulse, whereas as a criterion for the dominant ionization mechanism, the value of the Keldysh parameter is more suitable for many-cycle pulses and long wave trains.

We calculate the energy spectrum of photoelectrons along the polarization direction, i.e. when the angle θ between the electron momentum and direction of field propagation is equal to zero, using the angle and energy resolved probability distribution [18]

$$\frac{dw}{d\Omega_{\mathbf{p}} d\mathcal{E}} = \frac{p}{(2\pi)^3}|M|^2, \tag{16}$$

where $\mathcal{E}$ is the photoelectron energy and $\Omega_{\mathbf{p}}$ is the solid angle in the direction of electron momentum. To simplify further numerical calculation we used a plane wave as a final state $|\psi_f^a\rangle$ of a photoelectron. This approximation is well justified for short-range potentials, and for ionic potentials with long-range Coulomb tails it is better suited for more energetic final electrons.

As mentioned previously, we consider the energy distribution of the photoelectron in the direction of a linearly polarized field, i.e., for $p_\perp = 0$, and compare it with TDSE results obtained with the QPROP package [19]. First we analyzed [Fig. 1(a)] the spectrum for $E_0 = 3$ a.u., i.e., intensity of the order of 3.1×10^{21} W m^{-2} and $n = 2$. For this value of the maximum electric field of the pulse, our spectrum agrees with the TDSE result in the higher range of the energy spectrum, i.e. from 6 to 10 a.u, which is confirmed also for stronger fields, e.g., $E_0 = 10$ a.u., i.e., intensity equal to 3.5×10^{22} W m^{-2} [Fig. 1(b)]. The better agreement of our results with TDSE can be observed for longer laser pulses, i.e. for $n = 10$, where the agreement between our approach and the TDSE result can been seen for almost all range of energies (Fig. 2). In general, we observe better agreement of our results with TDSE for stronger fields and largest energies. Our observation is also that the present approach reproduces better the TDSE result for the values of parameters basically beyond the region of applicability of the standard SFA.

We have developed a gauge-invariant formalism which can be used for the unambiguous gauge-independent calculation of ionization amplitudes in the field of strong and short laser pulses and, contrary to, e.g., [20,21], does not require gauge-dependent partitioning of the total Hamiltonian into "free" and "interacting" parts. This approach is based on grouping together all terms in the standard SFA expansion that are of the same order with respect to atomic potential, and provides an expansion of the ionization amplitude which is independent of the choice of gauge order-by-order. The energy spectra of electrons ejected in the direction of the polarization vector of a linearly polarized pulse, calculated up to first order, show a qualitative agreement with predictions following from "exact" numerical solutions of TDSE, and the agreement improves

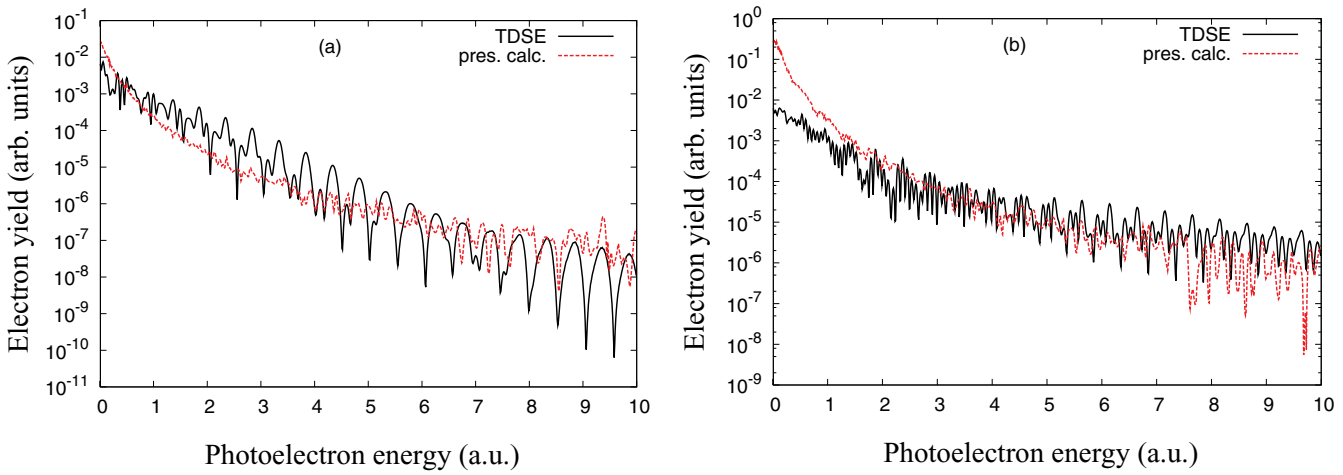

FIG. 2. (Color online) Same as Fig. 1, but for ten-cycle laser pulses with $\omega = 0.5$ a.u., corresponding to a wavelength of 91 nm, and with (a) $E_0 = 1$ a.u. and (b) $E_0 = 3$ a.u.

with increasing amplitude of the pulse electric field as well as with increasing energy of ejected electrons. Previously an approach related to that discussed in this paper was applied to ionization of a one-dimensional model atom with delta-like potential [1,14,22]. Discussion of the excitation and ionization of hydrogen atoms by ultraintense fields, performed along lines similar to the present work mainly in the context of tracing evolution of the ground-state atom under the influence of a rectangular pulse, can be also found in the paper by Geltman [23], and upper bounds on the ionization probabilities have been discussed in [2], where also the issue of convergence was briefly discussed.

Expansion of the transition amplitude in powers of the atomic potential, which is analogous to the Born series in scattering theory [23], is absolutely convergent for absolutely integrable potentials in the one-dimensional case, and, without limitation to a one-dimensional case, for potentials represented by bounded operators [1]. It is a sufficient condition of convergence which is fulfilled, e.g., by short-range potentials regular at origin, but is not fulfilled by typical atomic potentials. This does not mean that the series cannot be either convergent or asymptotic. In general, one may expect that the first approximation, or the series truncated at some further term, would give better results for stronger external fields and higher energies of ejected electrons, similar to the case of Born series in scattering theory. It has been conjectured in Ref. [14] that a dimensionless parameter controlling the expansion should be chosen as $(E_{\mathrm{at}}/E_0)\omega\tau$, where E_{at} is the atomic electric field ($\approx$1 a.u.), E_0 is the amplitude of the laser electric field, ω is the carrier-wave frequency, and τ is the pulse duration time. However, the question of whether for potentials represented by unbounded operators the series (1) is convergent or asymptotic is still unanswered and requires further consideration.

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