

Relativistic calculation of the electron-momentum shift in tunneling ionization

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We describe a procedure for the solution of the time-dependent Dirac equation. The procedure is based on the relativistic generalization of the matrix iteration method. We use this procedure to study electron-momentum distribution along the laser-beam propagation direction for the process of the tunneling ionization of a hydrogen atom. We found, in agreement with the experimental observations [C. T. L. Smeenk, L. Arissian, B. Zhou, A. Mysyrowicz, D. M. Villeneuve, A. Staudte, and P. B. Corkum, *Phys. Rev. Lett.* **106**, 193002 (2011)], that relativistic effects lead to appreciable deviation of the distribution from the strict left-right symmetry present in the nonrelativistic case. The expectation value of the momentum along the laser-beam propagation direction grows linearly with intensity and follows closely the behavior of the expectation value of the kinetic energy divided by the speed of light. These features agree with the experimental results [C. T. L. Smeenk, L. Arissian, B. Zhou, A. Mysyrowicz, D. M. Villeneuve, A. Staudte, and P. B. Corkum, *Phys. Rev. Lett.* **106**, 193002 (2011)].

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Keldysh's pioneering work [1] and its subsequent developments [2,3] laid out a basis of our current understanding of the main features of the strong field ionization of atoms and molecules. According to this theory ionization phenomena can be separated into two broad classes: the multiphoton and tunneling ionization. Whether an ionization process can be characterized as a multiphoton or a tunneling one depends on the value of the Keldysh parameter $\gamma = w\sqrt{2I}/E$ (where w and E are the frequency and strength of the laser field, respectively, and I is the ionization potential of the target in atomic units). Values of $\gamma \ll 1$ correspond to the tunneling regime, which interests us in the present work.

Original Keldysh theory relied on a completely nonrelativistic approach. A relativistic modification of this theory was proposed in [4]. It is clear that such relativistic modification is indispensable if our goal is the proper description of the ionization phenomena occurring for the field intensities of the order of several units of 1×10^{22} W/cm² which are currently available [5], or if we are interested in ionization driven by high-frequency electromagnetic fields. However, even for the infrared (IR) laser fields of much lower intensity, relativistic effects can manifest themselves. Even though the momentum of a photon is small, for the processes with participation of a large number of photons the total momentum delivered to the atom can be non-negligible. Smeenk *et al.* [6] report results of the study of the ionization process driven by the laser pulses with wavelengths of 800 and 1400 nm under experimental conditions where a large number of photons (30–50) is absorbed. The experiment demonstrated that the average electron-momentum component along the laser propagation direction grows linearly with the laser field intensity and is nearly equal to the average photoelectron energy divided by the speed of light. Another example is provided by the recent study [7] of momentum sharing between the photoelectron and the parental ion in the processes of single- and multiphoton ionization. It was found [7] that momentum sharing is very different for the two processes, with intense transfer of the momentum from the field to the ion-electron system occurring

for field intensities of several units of 1×10^{14} W/cm². A detailed study of the relativistic features of the tunneling ionization was reported in [8]. Introducing a gauge-invariant notion of the potential barrier for the process of the relativistic tunneling and reducing the problem to a one-dimensional tunneling model, the authors showed that there is a clear manifestation of the relativistic effects in tunneling ionization. Even for moderate field intensities an ionized electron acquires a non-negligible momentum shift along the direction of the laser-pulse propagation.

This is not the only instance where relativistic effects manifest themselves in the tunneling ionization even for moderately high field intensities. It was shown in [8,9] that correct treatment of the relativistic effects may prove important for the solution of the problem of the “tunneling time”—the time it takes a bound electron to tunnel through the barrier and become ionized. A rapid progress of the attosecond angular streaking techniques [10,11] offers a unique opportunity to study this question experimentally. An interesting and important issue arising in relation to the problem of the tunneling time is whether one can consider sub-barrier motion of the electron as instantaneous, an assumption which contradicts the special theory of relativity [12]. Another manifestation of the relativistic effects in an apparently nonrelativistic setting was found in [13]. The authors used a relativistic version of the intense field S -matrix approach [2] to study the spin-flip process in a hydrogen atom in circularly polarized near-infrared and XUV laser fields. Considerable intensity-dependent spin-flip asymmetry was found, which was shown to survive even when relativistic effects, such as retardation and spin-orbit effects, were very small.

To be able to address these questions we need to include relativistic effects in the theory describing the evolution of the atomic system under the action of the laser pulse in time. Below we describe such an approach based on the numerical solution of the time-dependent Dirac equation for an atom in the presence of an external electromagnetic field. The Dirac equation for a single electron provides the framework allowing one to take into account relativistic effects in an *ab initio* way, as long as the effects of quantum electrodynamics can be neglected. In the literature the Dirac equation is often solved

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using approximations similar to the well-known nonrelativistic strong-field approximation (SFA), where the effect of the atomic potential on the ionized electron is neglected [7,14]. In [15,16] approaches to the solution of the full time-dependent Dirac equation based on the use of the so-called split operator method have been described. We propose and describe below a procedure which is based on the relativistic generalization of the so-called matrix iteration method [17]. We used a nonrelativistic implementation of this procedure to study the evolution of atomic systems in strong fields of arbitrary polarization [18–20] and found this method to be accurate, fast, and reliable. We try to show below that relativistic generalization of the matrix iteration procedure inherits many of these features. Atomic units with $\hbar = 1$, $e = 1$, $m = 1$, and $c \approx 137.036$ (here e and m are charge and mass of the electron, and c is the speed of light) are used throughout the paper.

I. THEORY

We solve the time-dependent Dirac equation

$$i \frac{\partial \Psi(\mathbf{r}, t)}{\partial t} = \hat{H} \Psi(\mathbf{r}, t) \quad (1)$$

for the bispinor $\Psi(\mathbf{r}, t)$ with the Hamiltonian operator

$$\hat{H} = \hat{H}_{\text{atom}} + \hat{H}_{\text{int}}, \quad (2)$$

with

$$\hat{H}_{\text{atom}} = c \boldsymbol{\alpha} \cdot \hat{\mathbf{p}} + c^2(\beta - I) + I V(r) \quad (3)$$

and

$$\hat{H}_{\text{int}} = c \boldsymbol{\alpha} \cdot \mathbf{A} \quad (4)$$

In these expressions,

$$\boldsymbol{\alpha} = \begin{pmatrix} \mathbf{0} & \boldsymbol{\sigma} \\ \boldsymbol{\sigma} & \mathbf{0} \end{pmatrix}, \quad \beta = \begin{pmatrix} I & \mathbf{0} \\ \mathbf{0} & -I \end{pmatrix}, \quad I = \begin{pmatrix} I & \mathbf{0} \\ \mathbf{0} & I \end{pmatrix},$$

$\boldsymbol{\sigma}$ are Pauli matrices, $\mathbf{0}$ and I are 2×2 null and identity matrices, and c is the speed of light. To facilitate comparison with the

nonrelativistic calculations, we subtracted the constant term Ic^2 corresponding to the rest mass energy of the electron from the field-free atomic Hamiltonian (3). In Eq. (3) $V(r) = -\frac{1}{r}$ is the Coulomb potential. We use Coulomb gauge, interaction of the atom, and the external electromagnetic field is described by means of the vector potential \mathbf{A} in Eq. (4). We employ here the definition of the vector potential commonly used in atomic physics so that electric field and vector potential are related as $\mathbf{E} = -\frac{\partial \mathbf{A}}{\partial t}$.

To solve Eq. (1) we modify the strategy used in [18,19,21] for solution of the nonrelativistic time-dependent Schrödinger equation (TDSE). The solution is represented as a series in basis bispinors:

$$\Psi(\mathbf{r}, t) = \sum_{l=j \pm 1/2}^j \sum_{M=-j}^j \Psi_{j l M}(\mathbf{r}, t), \quad (5)$$

where each basis bispinor is

$$\Psi_{j l M}(\mathbf{r}, t) = \begin{pmatrix} g_{j l M}(r, t) \Omega_{j l M}(\mathbf{n}) \\ f_{j l M}(r, t) \Omega_{j l' M}(\mathbf{n}) \end{pmatrix}, \quad (6)$$

and two-component spherical spinors are defined as

$$\Omega_{j l M}(\mathbf{n}) = \begin{pmatrix} C_{l M - \frac{1}{2} \frac{1}{2}}^{j M} Y_{l, M - \frac{1}{2}}(\mathbf{n}) \\ C_{l M + \frac{1}{2} \frac{1}{2}}^{j M} Y_{l, M + \frac{1}{2}}(\mathbf{n}) \end{pmatrix},$$

where $C_{l m \frac{1}{2} \mu}^{j M}$ are the Clebsch-Gordan coefficients, $Y_{lm}(\mathbf{n})$ the spherical harmonics, and $\mathbf{n} = \mathbf{r}/r$. Parameters l and l' in Eq. (5) must satisfy the relation $l + l' = 2j$.

The action of the operators \hat{H}_{atom} and \hat{H}_{int} on the basis bispinors $\Psi_{j l M}(\mathbf{r}, t)$ in Eq. (6) can be easily found using well-known formulas [22,23]. To save the space we give here only one of the formulas, the explicit form of which we need below:

$$\hat{H}_{\text{atom}} \Psi_{j l M}(\mathbf{r}, t) = \begin{pmatrix} \left\{ i c \left(\frac{1}{r} \frac{d(f_{j l M}(r, t))}{dr} - \chi \frac{f_{j l M}(r, t)}{r} \right) + V(r) g_{j l M}(r, t) \right\} \Omega_{j l M}(\mathbf{n}) \\ \left\{ i c \left(\frac{1}{r} \frac{d(g_{j l M}(r, t))}{dr} + \chi \frac{g_{j l M}(r, t)}{r} \right) + (V(r) - 2c^2) f_{j l M}(r, t) \right\} \Omega_{j l' M}(\mathbf{n}) \end{pmatrix}, \quad (7)$$

where $\chi = -j - 1/2$ for $j = l + 1/2$, and $\chi = j + 1/2$ for $j = l - 1/2$.

In the most general case the vector potential \mathbf{A} in Hamiltonian (4) is a function of temporal and spatial variables. Taking as an example a linearly polarized pulse traveling in the direction of a unit vector $\hat{\mathbf{k}}$, we may write the vector potential as

$$\mathbf{A}(\mathbf{r}, t) = \hat{\mathbf{e}} h(\zeta), \quad (8)$$

where $\hat{\mathbf{e}} \cdot \hat{\mathbf{k}} = 0$, $\zeta = t - \frac{r \cdot \hat{\mathbf{k}}}{c}$, and $h(\zeta)$ has a compact support [i.e., it is zero outside an interval $(0, T_1)$] for a pulse of finite temporal duration and spatial extension. The logic of the calculational procedure we describe dictates that dependence of the vector potential on the spatial variables should be treated by means of an expansion in spherical harmonics. At every

step during time integration, therefore, we expand the vector potential as

$$\mathbf{A}(\mathbf{r}, t_n) = \hat{\mathbf{e}} \sum_{kq} h_{kq}(r, t_n) Y_{kq}(\mathbf{r}), \quad (9)$$

with

$$h_{kq}(r, t_n) = \int Y_{kq}^*(\mathbf{r}) h(\zeta) d\Omega. \quad (10)$$

These integrals are computed numerically using Gaussian quadratures. The radial functions $g_{j l M}(r, t)$ and $f_{j l M}(r, t)$ in Eq. (5) and Hamiltonian operator \hat{H} in Eq. (1) are discretized on the grid with the step size δr in a box of size R_{max} . Substitution of the expansion (5) in the discretized Dirac equation gives a set of coupled equations on the radial

amplitudes $g_{jIM}^n(t) = g_{jIM}(r_n, t)$, and $f_{jIM}^n(t) = f_{jIM}(r_n, t)$. These equations are solved using the matrix iteration method (MIM) [17]. This well-known technique is often applied for the solution of the nonrelativistic TDSE for atoms in the laser field [18–20,24]. For the present case of the Dirac equation some aspects of the application of this technique differ from the nonrelativistic case. Therefore, we describe the application of the matrix iteration method for the solution of the time-dependent Dirac equation in more detail.

As in the nonrelativistic case, the starting point for the development of the MIM procedure is the expression for the short-time Crank-Nicolson (CN) propagator [25] (this expression can also be regarded as a Cayley representation of the unitary short-time propagator):

$$\Psi(\mathbf{r}, t_n + \delta) = \frac{1 - i\hat{H}(t_n + \delta/2)\delta/2}{1 + i\hat{H}(t_n + \delta/2)\delta/2} \Psi(\mathbf{r}, t_n). \quad (11)$$

The most difficult and time-consuming part of the calculation is finding the inverse of the operator in the denominator in Eq. (11). This operator can be represented as $1 + i\hat{H}(t_n + \delta/2)\delta/2 = \hat{A} + \hat{B}$, where $\hat{A} = 1 + i\hat{H}_{\text{atom}}\delta/2$ and $\hat{B} = i\hat{H}_{\text{int}}(t_n + \delta/2)\delta/2$.

Using the Neumann expansion for $(\hat{A} + \hat{B})^{-1}$,

$$(\hat{A} + \hat{B})^{-1} = \hat{A}^{-1} - \hat{A}^{-1}\hat{B}\hat{A}^{-1} + \hat{A}^{-1}\hat{B}\hat{A}^{-1}\hat{B}\hat{A}^{-1} - \dots, \quad (12)$$

one reduces the problem of computing the inverse of the operator $1 + i\hat{H}(t_n + \delta/2)\delta/2$ in Eq. (11) to the repeated computation of the inverse of the operator \hat{A} . We note that \hat{A} inherits all its properties from the atomic Hamiltonian \hat{H}_{atom} . This means, in particular, that \hat{A} is diagonal in quantum numbers jIM . Operator \hat{A} , therefore, can be inverted separately in every subspace spanned by the bispinors with a given jIM . As Eq. (7) shows, to find the action of the atomic Hamiltonian on the basis bispinor $\Psi_{jIM}(\mathbf{r}, t)$, we have to compute first derivatives of the functions $g_{jIM}(r, t)$ and $f_{jIM}(r, t)$. We use central differences to represent these derivatives in the discretized version of Eq. (7). In order to compute the action of the atomic Hamiltonian on the basis bispinor at a given grid point r_n we need, therefore, to know $g_{jIM}^k(t)$ and $f_{jIM}^m(t)$ with $k = n \pm 1$, $m = n \pm 1$. One can see now that, reordering, if necessary, the grid variables $g_{jIM}^k(t)$ and $f_{jIM}^m(t)$, we may describe the action of the atomic Hamiltonian on the grid variables by means of a band matrix. The same statement is true, of course, for the operator \hat{A} . Its inversion, therefore, requires inversion of a band matrix which can be easily and efficiently done using standard algorithms. Convergence of the Neumann expansion (12) can be monitored, choosing the time step size δ appropriately [17].

Electron spectra $P(E)$ were obtained by projecting the solution of the time-dependent Dirac equation after the end of the pulse on the set of the continuum states $\Psi_{jIM}(\mathbf{r})$ of Hamiltonian (3) and summing over all possible jIM . Photoelectron angular distributions (PADs) were calculated by computing ionization amplitudes $a(\mu, \mathbf{p})$, where μ is polarization (which can be visualized as the spin direction in the electron's rest frame) and \mathbf{p} is asymptotic electron momentum. The amplitudes were obtained by projecting the solution of the time-dependent Dirac equation after the end of the pulse on the set of the ingoing relativistic scattering states

$\Psi_{\mu, \mathbf{p}}^-(\mathbf{r})$ of the hydrogen atom [22]:

$$\Psi_{\mu, \mathbf{p}}^-(\mathbf{r}) = \sum_{jIM} i^l e^{-i\delta_{jl}(p)} \langle \Omega_{jIM}(\hat{\mathbf{p}}) | v_{\mu} \rangle \Psi_{pjIM}(\mathbf{r}), \quad (13)$$

where $\Psi_{pjIM}(\mathbf{r})$ are continuous-spectrum wave functions of the Dirac Hamiltonian normalized to $\delta(p - p')$, $\delta_{jl}(p)$ is the relativistic Coulomb phase shift, $\Omega_{jIM}(\hat{\mathbf{p}})$ is a two-component spherical spinor, and v_{μ} is a two-component spinor describing the polarization state.

Of particular interest to us was the distribution of the electron-momentum component along the laser-beam propagation direction. We use below the geometry in which the beam propagates along the x axis. The distribution was, therefore, computed as

$$W(p_x) = \sum_{\mu} \int |a(\mu, \mathbf{p})|^2 dp_y dp_z. \quad (14)$$

II. RESULTS

Before presenting our results for the tunneling ionization we would like to discuss briefly an issue which makes the Dirac problem rather different from the nonrelativistic case. This issue arises from the well-known fact of the absence of the lower bound for the Hamiltonian operator (3). This fact has far-reaching consequences and led, ultimately, to the discovery of a positron. We are concerned here with purely numerical aspects of this fact. Negative energy states have energies hugely different from those with positive energies. In the atomic units system we are using, the difference is approximately $2c^2 = 2 \times 137.036^2$. If, therefore, we have in the initial state a superposition of the states with positive and negative energies, we will have two hugely different time scales in the problem. This fact is responsible for the so-called *Zitterbewegung* [26]. To reproduce this phenomenon faithfully we must use an integration time step $\Delta \ll 1/c^2$. Figure 1 illustrates the *Zitterbewegung* for two observables: operator r , and \hat{H} for the evolution starting from the initial state prepared as a superposition (with equal weights) of the bispinor describing the ground $1s_{\frac{1}{2}}$ state, and the bispinor obtained if upper and lower components are swapped (such a state has an energy below the value of $-2c^2$).

Results shown in Fig. 1 have been obtained for the time step $\Delta t = 1 \times 10^{-6}$ a.u. and show the expected oscillations of the expectation values. We should note that this issue

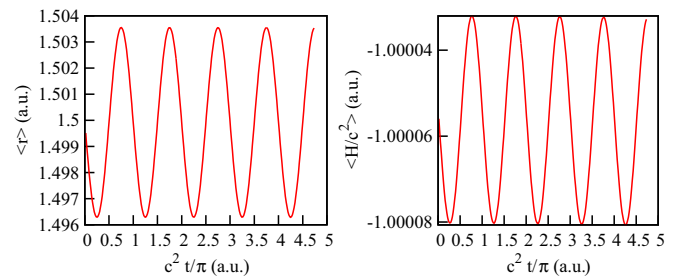


FIG. 1. (Color online) Time dependence of the expectation values of operators r and \hat{H} . Evolution starts from the initial superposition of states with energies which differ by approximately $2c^2$.

is not central to our paper; we touch upon it very briefly only to illustrate that our method is accurate enough to reproduce these tiny features. Of course, in any calculation of tunneling ionization, using time steps of the order of 1×10^{-6} a.u. would be utterly impossible. Fortunately, we do not have to do that. The situation here is similar to the one encountered when stiff (the ones having very different time scales) systems of equations are solved numerically. The proper choice of the stable integration method ensures that while very tiny features (due to the smaller time scale) cannot be reproduced, the overall behavior (governed by the larger time scale) can be reproduced faithfully. In the calculations of the tunneling ionization reported below we use the time step $\Delta t = 1 \times 10^{-1}$ a.u., and matrix iteration procedure of order 20 [i.e., we use the first 20 terms of the Neumann series (12) to compute the operator inverse].

We performed calculations for the linearly polarized pulse with polarization vector along the z axis, propagating in the positive direction of the x axis. To describe such a pulse we used the following expression for the vector potential:

$$\mathbf{A}(\mathbf{r}, t) = -\hat{z} \frac{E_0}{\omega} T(\zeta) \sin^2\left(\frac{\Omega\zeta}{2}\right) \sin(\omega\zeta), \quad (15)$$

where $\zeta = t - x/c$, \hat{z} is a unit vector in the z direction, $T(\zeta)$ is a rectangular window function, such that $T(\zeta) = 1$ for $\zeta \in (0, T_1)$ and zero outside this interval, and $\Omega = 2\pi/T_1$. The parameters in Eq. (15) were carrier frequency $\omega = 0.057$ a.u., corresponding to the wavelength of 800 nm; parameter T_1 (total duration of the pulse) was chosen as $T_1 = 2T$, where $T = 2\pi/\omega = 2.667$ fs is the duration of an optical cycle.

We used atomic hydrogen as a target. The Hamiltonian operator was discretized on a grid with the step size $\delta r = 0.1$ a.u., and the radial variable was restricted to an interval $(0, R_{\max})$, with $R_{\max} = 400$ a.u. The maximum value of the parameter j in Eq. (5) was $J_{\max} = 60\frac{1}{2}$. The initial state was prepared by solving the eigenvalue problem for the discretized Hamiltonian, which resulted in the ground-state energy of -0.500006661 a.u., which is to be compared with the value -0.500006657 a.u. given by the Dirac formula (we remind that, to facilitate comparison with the nonrelativistic calculations, we subtract the rest mass energy term mc^2 from the Hamiltonian).

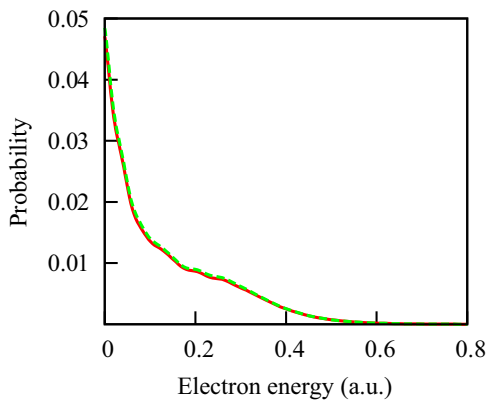


FIG. 2. (Color online) Relativistic (solid red line) and nonrelativistic (dashed green line) energy spectra for the field intensity of 1×10^{14} W/cm².

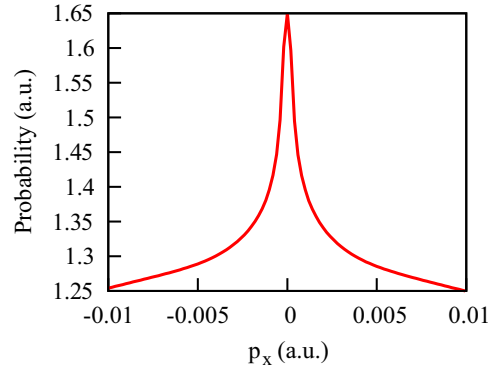


FIG. 3. (Color online) Distribution $W(p_x)$ for the field intensity of 5.05×10^{14} W/cm² a.u.

The energy spectrum for the field intensity of 1×10^{14} W/cm², obtained using the computational procedure we described above, is shown in Fig. 2. We present results of both relativistic calculation, performed using the approach described above, and the nonrelativistic calculation relying on the nonrelativistic implementation of the matrix iteration method [18]. The results of both relativistic and nonrelativistic calculations for the energy spectra are virtually identical. A finer characteristic, allowing one to actually see the relativistic effects, is $W(p_x)$, the electron-momentum distribution in the direction of the laser-beam propagation. This distribution, computed according to Eq. (14), is shown in Fig. 3 for the laser field intensity of 5.05×10^{14} W/cm². Figure 3 shows a slight asymmetry, which can be conveniently characterized by the first moment of the distribution $W(p_x)$, the expectation value $\langle p_x \rangle$.

Figure 4 shows the expectation value $\langle p_x \rangle$ as a function of the laser intensity. $\langle p_x \rangle$ grows linearly with intensity and follows very closely the curve showing dependence of the average kinetic energy divided by the speed of light, which is the behavior observed in the experimental work [6].

III. CONCLUSION

We described a procedure for the solution of the time-dependent Dirac equation. The procedure is based on the relativistic generalization of the well-known matrix iteration

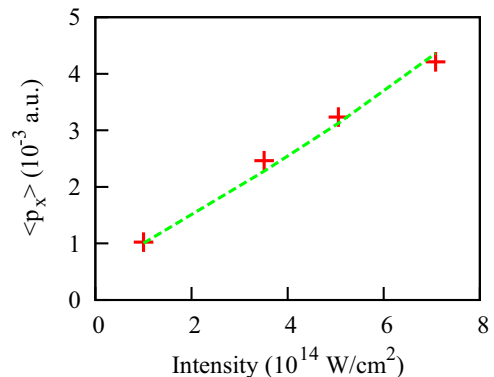


FIG. 4. (Color online) Expectation value $\langle p_x \rangle$ (red crosses) as a function of the field intensity; $\frac{1}{c} \langle E_{\text{kin}} \rangle$ (green dashed line).

method [17]. The method has been often used in nonrelativistic calculations [17,18,24] and is known to be reliable, accurate, and easily adaptable to complicated field geometries, such as fields of arbitrary ellipticity [19,20]. We described application of the relativistic matrix iteration method to the case of the linearly polarized laser pulse; the procedure can also be applied for the driving field of arbitrary polarization.

We used this procedure to study the electron-momentum distribution along the laser-beam propagation direction for the process of tunneling ionization of a hydrogen atom. We found, in agreement with experimental observations [6], that relativistic effects lead to appreciable deviation of the

distribution from the strict left-right symmetry present in the nonrelativistic case. The first moment of this distribution, the expectation value of the momentum along the laser beam propagation direction, grows linearly with intensity and mimics closely the behavior of another observable, the expectation value of the kinetic energy divided by the speed of light. These features agree with the experimental results [6].

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