

**Breakdown of the nonrelativistic approximation in superintense laser-matter interactions**

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We study the breakdown of the nonrelativistic approximation in the multiphoton ionization of atomic hydrogen by some intense x-ray laser pulse, in a regime where the dipole approximation is no longer valid. To this end, both the time-dependent Dirac equation as well as its nonrelativistic counterpart, the time-dependent Schrödinger equation, are solved within an *ab initio* numerical framework. It is demonstrated that a recently developed semirelativistic Schrödinger equation for superintense laser fields [Lindblom *et al.*, *Phys. Rev. Lett.* **121**, 253202 (2018)] yields results in excellent agreement with the fully relativistic treatment. The semirelativistic equation is then used in an investigation of the role of higher-order beyond-dipole corrections to the laser-matter interaction. The result of the present study can be summarized into two main findings: (1) relativistic effects predict a blueshift of the multiphoton ionization spectrum, and (2) higher-order beyond dipole corrections (beyond the leading-order term) indicate a corresponding redshift of the photoelectron spectrum. However, the two shifts turn out to be of the same order of magnitude, effectively leading to a net cancellation of their respective contributions. This apparent cancellation effect raises an important question: Is the distinction between relativistic blueshifts and higher-order beyond dipole redshifts meaningful from an experimental point of view? The result of the present study indicates that the answer is negative because the two effects nonetheless cannot be measured separately. Therefore, instead, we suggest that the present findings should merely be taken as a demonstration that caution should be exercised when higher-order beyond-dipole and relativistic corrections are to be taken into account in approximation schemes in the modeling of superintense laser-matter interactions.

DOI: [10.1103/PhysRevA.99.053410](https://doi.org/10.1103/PhysRevA.99.053410)**I. INTRODUCTION**

There is a growing interest in studying ionization and excitation processes in atoms induced by intense laser fields in regimes where the validity of the celebrated dipole approximation breaks down [1–8]. In the dipole approximation the spatial dependency of the laser field is not considered, and in addition the magnetic-field component is disregarded. The dipole approximation greatly simplifies the modeling of the light-matter interaction and as such it is usually the preferred theoretical approach. The validity of the dipole approximation is commonly studied within the context of nonrelativistic quantum mechanics and the time-dependent Schrödinger equation (TDSE) [9–23]. However, in the limit of very high laser intensities, at some point the nonrelativistic TDSE approach cannot be applied and a fully relativistic treatment of the laser-matter interaction is prerequisite [24–31].

In this work, we go beyond the standard nonrelativistic approximation and study the relativistic ionization dynamics of hydrogen by some intense x-ray laser pulse in a regime where the ordinary nonrelativistic Schrödinger equation is no longer valid. To this end, we consider a hydrogen  $1s$  electron exposed to a 15 cycle 1.36 keV laser pulse of peak intensity  $3.5 \times 10^{22}$  W/cm<sup>2</sup>. At this high intensity the electron may be accelerated to about 15% of the speed of light during the laser interaction, and as such a nonrelativistic description of the ionization process becomes questionable. In a recent

work [32], it was shown that the dipole approximation is generally not valid for such laser parameters, and beyond-dipole (nondipole) effects, i.e., the effect of the magnetic component of the field, must be properly taken into account in the theoretical modeling of the laser-matter interaction. Here we go one step further and also include relativistic corrections to the dynamics. This is achieved using two independent theoretical approaches: (1) solving the fully relativistic time-dependent Dirac equation numerically and (2) modeling the problem by means of a recently developed semirelativistic Schrödinger equation [27].

It is demonstrated that the semirelativistic Schrödinger formulation—even in the absence of any spin-dependent term—provides results in excellent agreement with the fully relativistic treatment, merely demonstrating the power of such a semirelativistic formulation of the light-matter interaction. Considering the energy of the emitted photoelectron, it is found that relativistic effects give rise to an apparent positive shift (blueshift) of the multiphoton ionization spectrum to higher energies, i.e., the electron seems to acquire a higher kinetic energy than what is predicted by the corresponding nonrelativistic theory. But as we shall see below, this shift might not be experimentally observable, because there exists another competing (nonrelativistic) shift that tends to cancel the effect of the relativistic correction. Although the electron's spin degrees of freedom turns out to be of less importance here, it is relatively straightforward to include such effects in the semirelativistic Schrödinger equation, i.e., the spin-orbit interaction, the interaction of the spin with the magnetic field, etc.

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Having justified the validity of the semirelativistic approach, in the last part of this work we apply the semirelativistic time-dependent Schrödinger equation in an investigation of the role of higher-order nondipole effects in the laser-matter interaction, including terms beyond the leading-order magnetic component. One such nonvanishing correction term is identified and its significance seems to be of relativistic order, i.e., its effect turns out to be of equal importance as purely relativistic effects. Furthermore, it is found that this higher-order beyond-dipole correction gives rise to a redshift in the corresponding photoelectron spectrum, i.e., an energy shift in the complete opposite direction than the previously predicted relativistic blueshift. Moreover, the net effect when both shifts are considered seems to be very small. This finding is particularly important and might be decisive for how one should proceed in order to insert magnetic and relativistic corrections into the time-dependent Schrödinger equation in a consistent manner.

Atomic units (a.u.) are used where stated explicitly.

## II. THEORY AND METHODOLOGY

All results in the present work are obtained within the so-called propagation gauge formulation of the light-matter interaction [33–35]. Of course, in an exact treatment the prediction for any observable is gauge independent. Nonetheless, the reason for using the propagation gauge here is that it has proven to exhibit superior performance in terms of convergence in actual numerical applications, in particular for computations of laser-matter interactions in the superintense-field regime beyond the dipole approximation [33–35]. Furthermore, the propagation gauge Hamiltonian has the clear advantage that the long-wavelength approximation (LWA), i.e., disregarding the spatial dependence in the vector potential  $\mathbf{A}$ , may be imposed without making any assumptions about the importance of the magnetic component of the laser field [33], as this is taken into account by a separate (propagation gauge) term in the interaction. This stands in stark contrast to the usual minimal-coupling (velocity gauge) formulation of the light-matter interaction where spatial dependency in the vector potential must be explicitly introduced in order to take magnetic-field effects into account [11,18,36].

In the first part of this work we present a derivation of the semirelativistic Schrödinger equation in the propagation gauge [27]. We start out with the minimal-coupling Klein–

Gordon equation for a spinless particle of mass  $m$  and charge  $q$  in an electromagnetic field, as described by some vector potential  $\mathbf{A}$  and scalar potential  $\varphi$ ,

$$\left(i\hbar\frac{\partial}{\partial t} - q\varphi\right)^2 X(\mathbf{r}, t) = [m^2c^4 + c^2(\mathbf{p} - q\mathbf{A})^2]X(\mathbf{r}, t). \quad (1)$$

Here  $c$  is the speed of light. To investigate the nonrelativistic limit of this equation, it is useful to introduce a new wave function given by the transformation  $\Psi(\mathbf{r}, t) = X(\mathbf{r}, t)e^{imc^2t/\hbar}$ , effectively shifting the energies downwards by the rest mass energy  $mc^2$ . Then, adopting the propagation gauge point of view [33,34], we consistently let the potentials transform according to  $\mathbf{A} \rightarrow \mathbf{A} + \nabla\xi$  and  $\varphi \rightarrow \varphi - \partial_t\xi$  with

$$\xi(\eta) = \xi(\omega t - \mathbf{k} \cdot \mathbf{r}) = \frac{q}{2m\omega} \int_{-\infty}^{\eta} A^2(\eta') d\eta', \quad (2)$$

where  $\omega$  is the central angular frequency of the electromagnetic field and  $\mathbf{k} = \omega/c \hat{\mathbf{k}}$  is the wave vector. With these transformations, the Klein–Gordon equation is cast into the form

$$\left[-\hbar^2\frac{\partial^2}{\partial t^2} + i\hbar\frac{\partial}{\partial t}B + Bi\hbar\frac{\partial}{\partial t} - C\right]\Psi' = 0, \quad (3)$$

with the operators  $B$  and  $C$  defined as follows:

$$\begin{aligned} B &= mc^2 + \frac{q^2}{2m}A^2 - q\varphi, \\ C &= p^2c^2 - c^2\left(q\mathbf{A} - \frac{q^2}{2mc}A^2\hat{\mathbf{k}}\right) \cdot \mathbf{p} \\ &\quad - c^2\mathbf{p} \cdot \left(q\mathbf{A} - \frac{q^2}{2mc}A^2\hat{\mathbf{k}}\right) \\ &\quad + q\varphi\left(2mc^2 + \frac{q^2}{m}A^2 - q\varphi\right). \end{aligned} \quad (5)$$

We have here assumed transverse electromagnetic fields so that  $\hat{\mathbf{k}} \cdot \mathbf{A} = 0$ . Furthermore, the wave function  $\Psi'$  in the propagation gauge representation is related to the original wave function  $\Psi$  by the transformation  $\Psi' = e^{i\xi}\Psi$ . Now, insisting that  $i\hbar\frac{\partial}{\partial t}$  is the operator for the total energy, i.e., the Hamiltonian operator  $H$ , it is straightforward to show that Eq. (3) has a solution if and only if  $H = -B \pm (B^2 + C)^{1/2}$ . Keeping only the solution corresponding to positive-energy states, the resulting Hamiltonian becomes

$$H = mc^2 \sqrt{\left(1 + \frac{q^2}{2m^2c^2}A^2\right)^2 + \frac{2}{mc^2}\left[\frac{p^2}{2m} - \frac{q}{2m}\left\{\mathbf{A} - \frac{q}{2mc}A^2\hat{\mathbf{k}} \cdot \mathbf{p}\right\}\right]} - mc^2 - \frac{q^2}{2m}A^2 + q\varphi. \quad (6)$$

In this equation, the curly brackets denote the anticommutator defined by  $\{a, b\} = ab + ba$ . Note at this point that the vector potential  $\mathbf{A}(\eta) = \mathbf{A}(\omega t - \mathbf{k} \cdot \mathbf{r})$  satisfies the wave equation and generally depends on both space and time coordinates. Hence the order of the operators is important and  $A^2\hat{\mathbf{k}} \cdot \mathbf{p} \neq \hat{\mathbf{k}} \cdot \mathbf{p}A^2$ .

Next, assuming that  $q\varphi = V$  is a Coulombic potential and expanding the square-root in the Hamiltonian (6) in a Maclaurin series that explicitly ensures

the Hermiticity of the operators term by term, i.e., writing  $(a^2 + b)^{1/2} = \frac{1}{2}a(1 + a^{-2}b)^{1/2} + \frac{1}{2}(1 + ba^{-2})^{1/2}a = \frac{1}{2}a(1 + \frac{1}{2}a^{-2}b - \dots) + \frac{1}{2}(1 + \frac{1}{2}ba^{-2} - \dots)a$ , the Hamiltonian to leading order in  $1/c$  reads

$$H = \frac{p^2}{2m} + V - \frac{q}{m}\mathbf{A} \cdot \mathbf{p} + \frac{q^2}{4m^2c}\{A^2, \hat{\mathbf{k}} \cdot \mathbf{p}\} + O(1/c^2), \quad (7)$$

where it has been assumed that the electromagnetic field satisfies the Coulomb gauge restriction  $\nabla \cdot \mathbf{A} = 0$ . Equation (7) defines the (nonrelativistic) light-matter interaction Hamiltonian as obtained within the so-called propagation gauge [33,34].

Higher-order (relativistic) corrections to the Hamiltonian can now readily be introduced simply by accounting for higher-order terms in the Maclaurin series expansion of the square-root in Eq. (6). Furthermore, by using  $(1 + \frac{q^2}{2m^2c^2}A^2)^{-1} = 1 - \frac{q^2}{2m^2c^2}A^2 + O(1/c^4)$ , the propagation gauge Hamiltonian correct to order  $1/c^2$  becomes

$$H = \frac{p^2}{2m} + V - \frac{q}{m}\mathbf{A} \cdot \mathbf{p} + \frac{q^2}{4m^2c} \{A^2, \hat{\mathbf{k}} \cdot \mathbf{p}\} - \frac{p^4}{8m^3c^2} + \frac{q}{4m^3c^2} \{A \cdot \mathbf{p}, p^2\} + \frac{q^3}{2m^3c^2} A^2 \mathbf{A} \cdot \mathbf{p} - \frac{q^2}{8m^3c^2} \{A^2, p^2\} - \frac{q^2}{2m^3c^2} (\mathbf{A} \cdot \mathbf{p})^2 + O(1/c^3). \quad (8)$$

The final Hamiltonian (8), which accounts for relativistic effects at the lowest order in momentum, i.e., relativistic corrections of order  $1/c^2$ , is fully equivalent to the semirelativistic equation derived in Ref. [27]. At last, the temporal evolution of a particle of mass  $m$  and charge  $q$ , as represented by a wave function  $\Psi$  and moving in some potential  $V$  and electromagnetic field  $\mathbf{A}$ , is governed by the time-dependent Schrödinger equation (TDSE),  $i\hbar \frac{\partial}{\partial t} \Psi = H\Psi$ , with the Hamiltonians (7) and/or (8) in the nonrelativistic and (weakly) relativistic limits, respectively.

Note that only relativistic corrections to the kinetic energy of the particle have been included in Eq. (8), and that the usual spin-orbit and Darwin terms have not been considered. Therefore, to obtain a fully relativistic description of the laser-matter interaction, the corresponding solutions to the time-dependent Dirac equation (TDDE) are to be obtained. The TDDE formally takes the same form as the TDSE, i.e.,  $i\hbar \frac{\partial}{\partial t} \Psi = H\Psi$ , but now  $\Psi$  becomes a four-component bispinor wave function instead, simply due to the introduction of the electron-spin degrees of freedom as well as the positive- and negative-energy solutions. Furthermore, neglecting retardation effects in the electron-nucleus interaction and taking the nuclear mass to be infinite, the Dirac Hamiltonian in the propagation gauge is given by [35]

$$H = c\boldsymbol{\alpha} \cdot \left( \mathbf{p} - q\mathbf{A} + \frac{q^2}{2mc} A^2 \hat{\mathbf{k}} \right) + \beta mc^2 + V - \frac{q^2}{2m} A^2, \quad (9)$$

with

$$\boldsymbol{\alpha} = \begin{pmatrix} 0 & \boldsymbol{\sigma} \\ \boldsymbol{\sigma} & 0 \end{pmatrix}$$

and

$$\beta = \begin{pmatrix} I & 0 \\ 0 & -I \end{pmatrix},$$

and where  $I$  is the unit two-by-two matrix and  $\boldsymbol{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$  are the three Pauli two-by-two spin matrices.

Both the nonrelativistic and semirelativistic Schrödinger Hamiltonians (7) and (8), respectively, as well as the exact Dirac Hamiltonian (9), are all formulated within the propagation gauge representation of the light-matter interaction

[33–35]. In this representation the main contribution due to the magnetic field is accounted for by the last operator in Eq. (7), as well as the third term in the parentheses in the Dirac Hamiltonian (9).

The laser pulse is modeled in terms of the vector potential

$$\mathbf{A}(\eta) = \frac{E_0}{\omega} f(\eta) \sin \eta \hat{\mathbf{u}}_p, \quad (10)$$

where  $E_0$  is the electric-field strength at peak intensity,  $f(\eta)$  defines the laser pulse profile, and  $\hat{\mathbf{u}}_p$  is a unit vector pointing in the laser polarization direction. Solving the TDSE and/or the TDDE with the full vector potential (10) typically results in an intractable computational problem due to the coupling of space and time coordinates. Therefore, to simplify the theoretical treatment, it is common to assume that the space-dependent vector potential can be expanded in powers of  $\mathbf{k} \cdot \mathbf{r}$  [10,11,18], where the term linear in  $\mathbf{k} \cdot \mathbf{r}$  represents the leading-order correction.

A recent nonrelativistic study showed that the LWA works quite well for the current laser parameters [32], providing results in fair agreement with the exact ones. Therefore, to begin with, we will also adopt the LWA approach here. In the propagation gauge, the LWA is simply obtained by the substitution  $\mathbf{A}(\eta) \rightarrow \mathbf{A}(t)$ . In this approximation, the anticommutation rules in Eqs. (7) and (8) become superfluous as the operators now commute. Furthermore, the very last term in the Dirac Hamiltonian (9) becomes a purely time-dependent factor that effectively cancels out via a global phase transformation [18]. Note again that setting  $\mathbf{A}(\eta) \simeq \mathbf{A}(t)$  in the propagation gauge is not equivalent to assuming the dipole approximation, as the major contribution from the magnetic field is still accounted for by some separate term in the (propagation gauge) Hamiltonians (7)–(9), respectively. At first sight, it might seem counterintuitive that these Hamiltonians still support a magnetic field, despite the vector potential being taken to only depend on time and not on spatial degrees of freedom. The explanation lies in the gauge transformation and the fact that the magnetic field effectively cancels in the propagation gauge representation of the light-matter interaction, to the benefit of the electric field.

Finally, we will go beyond the LWA and also introduce higher-order beyond dipole corrections to the dynamics, the main finding being that such corrections might be crucial for an accurate treatment of the relativistic light-matter interaction. Nonetheless, we have found that the linear approximation is sufficient for the laser parameters considered here [32]. In this approximation the vector potential takes the form

$$\mathbf{A}(\eta) \simeq \mathbf{A}(t) + \frac{1}{c} \hat{\mathbf{k}} \cdot \mathbf{r} \mathbf{E}(t), \quad (11)$$

where  $\mathbf{E} = -\frac{\partial}{\partial t} \mathbf{A}$  is the electric field. We further adopt a sine-squared carrier envelope for the laser field, i.e.,

$$f(t) = \begin{cases} \sin^2\left(\frac{\pi t}{T}\right), & 0 < t < T \\ 0, & \text{otherwise.} \end{cases} \quad (12)$$

Using the expanded potential (11), the Hamiltonian (8) is cast into the approximate form

$$\begin{aligned}
H \simeq & \frac{p^2}{2m} + V - \frac{q}{m} \left[ \mathbf{A}(t) + \frac{1}{c} \hat{\mathbf{k}} \cdot \mathbf{r} \mathbf{E}(t) \right] \cdot \mathbf{p} + \frac{q^2}{2m^2 c} A^2(t) \hat{\mathbf{k}} \cdot \mathbf{p} + \frac{q^2}{2m^2 c^2} \mathbf{A}(t) \cdot \mathbf{E}(t) \{ \hat{\mathbf{k}} \cdot \mathbf{r}, \hat{\mathbf{k}} \cdot \mathbf{p} \} \\
& - \frac{p^4}{8m^3 c^2} + \frac{q}{2m^3 c^2} p^2 \mathbf{A}(t) \cdot \mathbf{p} + \frac{q^3}{2m^3 c^2} A^2(t) \mathbf{A}(t) \cdot \mathbf{p} - \frac{q^2}{4m^3 c^2} A^2(t) p^2 - \frac{q^2}{2m^3 c^2} [\mathbf{A}(t) \cdot \mathbf{p}]^2,
\end{aligned} \quad (13)$$

where all (non-important) terms of order  $1/c^3$  and higher have been omitted.

Both the TDSE and the TDDE are solved by using the spectral method where the wave function is expanded in a basis composed of the eigenstates of the time-independent (field-free) Hamiltonian. In the TDSE approach the equation is discretized following the procedure described in Ref. [18], i.e., the states are written as products of hydrogenic radial wave functions,  $R_{kl}$ , and spherical harmonics,  $Y_{lm}$ ,

$$\Psi(\mathbf{r}, t) = \sum_{klm} c_{klm}(t) R_{kl}(|\mathbf{r}|) Y_{lm}(\hat{\mathbf{r}}). \quad (14)$$

Both continuum (scattering) and bound states are included in the expansion. The radial wave functions are obtained by solving the corresponding hydrogenic radial equation in a  $B$ -spline basis. Finally, the time-dependent matrix elements of the TDSE Hamiltonians (7), (8), and (13) are computed, and the resulting system of ordinary differential equations is solved by using the predictor-corrector method developed by Shampine and Gordon [37].

The corresponding scheme for solving the TDDE with the Hamiltonian (9) is computationally more involved. Similarly to the TDSE case, the time-dependent wave function is now expanded in terms of the eigenstates of the field-free Dirac Hamiltonian,

$$H = c\boldsymbol{\alpha} \cdot \mathbf{p} + \beta mc^2 + V, \quad (15)$$

where the relativistic hydrogenic eigenstates are computed by diagonalizing the Hamiltonian (15) in the  $B$ -spline basis. But now special attention must be paid to the appearance of both positive- and negative-energy states in the spectrum as well as the possible occurrence of so-called spurious (artificial) states contaminating the spectrum. In contrast to the TDSE, such spurious states may occur in the relativistic theory when the Dirac equation is expanded in terms of finite basis sets, and several procedures have been suggested to resolve this problem [38–42]. Here we have chosen to employ the method of dual kinetic balance as developed by Shabaev *et al.* [40].

Once the eigenstates have been obtained from the diagonalization procedure, the TDDE is discretized by expanding the time-dependent four-component bispinor wave function as follows:

$$\Psi(\mathbf{r}, t) = \sum_{k,\kappa,j,m} c_{k,\kappa,j,m}(t) \psi_{k,\kappa,j,m}(\mathbf{r}), \quad (16)$$

with

$$\psi_{k,\kappa,j,m}(\mathbf{r}) = \frac{1}{r} \begin{pmatrix} P_{k,\kappa}(r) X_{\kappa,j,m}(\hat{\mathbf{r}}) \\ i Q_{k,\kappa}(r) X_{-\kappa,j,m}(\hat{\mathbf{r}}) \end{pmatrix}, \quad (17)$$

and where  $P_{k,\kappa}(r)$  and  $Q_{k,\kappa}(r)$  are the radial functions associated with the positive- and negative-energy components, respectively. Furthermore,  $X_{\kappa,j,m}$  represents the  $ls$ -coupled spherical spinor

$$X_{\kappa,j,m} = \sum_{m_s, m_l} \langle l_{\kappa}, m_{l_{\kappa}}; s, m_s | j, m \rangle Y_{l_{\kappa}, m_{l_{\kappa}}} \chi_{s, m_s}, \quad (18)$$

with the relativistic quantum number of angular momentum

$$\kappa = \begin{cases} -(l+1) & \text{for } j = l + 1/2 \\ l & \text{for } j = l - 1/2. \end{cases} \quad (19)$$

Finally, the time-dependent matrix elements of the Dirac Hamiltonian (9) are calculated in the basis (17), and the system is evolved in time. To this end, we combine Krylov subspace methods and the Arnoldi–Lanczos algorithm in order to perform matrix exponentiation of the time-dependent Hamiltonian and to propagate the system in time [30,43,44].

### III. RESULTS AND DISCUSSION

We consider a hydrogen  $1s$  electron exposed to a 1.36 keV x-ray laser pulse of angular frequency  $\omega = 50$  a.u., which corresponds to the photon energy 1.36 keV. The total duration of the pulse is set to 15 cycles, i.e., we let  $T = 15 \times 2\pi/\omega$  in Eq. (12). Furthermore, the peak intensity was fixed to  $3.5 \times 10^{22}$  W/cm<sup>2</sup>, corresponding to the choice  $E_0 = 1000$  a.u. for the electric-field amplitude in Eq. (10). At this high intensity the electron is likely to reach velocities of about 15% of the speed of light throughout the laser-matter interaction and, as such, relativistic effects are expected to have some impact on the dynamics. The values of the basis parameters in the expansions of the wave function in Eqs. (14) and (16) were varied systematically in order to control the accuracy and reliability of the computed data. For the TDDE calculations accurate results were obtained with a radial box of extension  $R_{\max} = 65$  a.u. and with the maximum attainable electronic continuum energy truncated at 610 a.u. Note that both positive- and negative-energy states were included in the expansion for completeness. Furthermore, the number of angular momenta included in the basis set was limited up to  $l = l_{\max} = 17$ .

Figure 1 shows the energy spectrum of the emitted electron as obtained by solving the nonrelativistic TDSE both within and beyond the dipole approximation. The probability distribution differential in energy  $E$  is calculated by the equation

$$\frac{dP}{dE} = \frac{1}{k} \frac{dP}{dk} = \frac{1}{k} \sum_{lm} \left| \langle \Phi_{klm}^C(\mathbf{r}) | \Psi(\mathbf{r}, t = T) \rangle \right|^2, \quad (20)$$

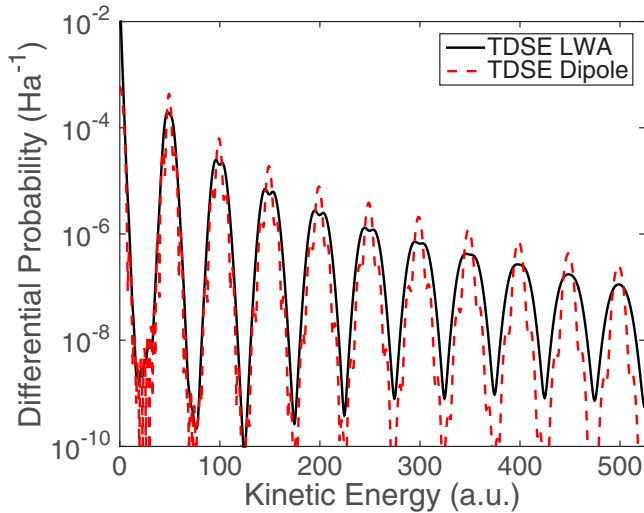


FIG. 1. Kinetic-energy spectrum of the emitted photoelectron, as obtained by Eq. (20), for a 15 cycle laser pulse with  $\omega = 50$  a.u. and  $E_0 = 1000$  a.u. Note that the abbreviation Ha on the y axis refers to hartree, the atomic unit of energy. Solid black line: the beyond dipole result obtained by solving the TDSE with the Hamiltonian in Eq. (7) within the LWA. Dashed red line: the corresponding dipole approximation result as obtained by solving the TDSE with the Hamiltonian (7) but omitting the last (diamagnetic) term. The total ionization probability equals 0.047 and 0.0043 in the beyond-dipole and dipole cases, respectively.

where  $\Psi(\mathbf{r}, t = T)$  is the wave function at the end of the laser pulse,  $\Phi_{klm}^C(\mathbf{r})$  is the Coulomb wave function (normalized on the  $k$  scale), and  $k = \sqrt{2E}$  is the wave number. A total number of ten (multiphoton) peaks corresponding to the net absorption of 1–10 photons from the field are depicted in the figure. The beyond-dipole result is calculated by means of the Hamiltonian (7) within the LWA, meaning that the spatial dependence in the vector potential has been omitted, i.e., setting  $\mathbf{A}(\eta) \simeq \mathbf{A}(t)$ . As such, the contribution of the magnetic component of the laser field is taken into account through the last (propagation-gauge) term in the Hamiltonian. The beyond-dipole result (black solid line) is compared with the corresponding dipole calculation result (red dashed line), which is obtained by neglecting the last (diamagnetic) term in Eq. (7). Figure 1 depicts multiphoton resonances up to tenth order, and clear differences in the calculated dipole and beyond-dipole results are identified, merely demonstrating that the dipole formulation of the laser-atom interaction Hamiltonian is not valid in the present ionization scenario and must therefore be abandoned. Note also the great difference in the total ionization yields, i.e., 0.047 and 0.0043 in the beyond-dipole and dipole limits, respectively. As it turns out, the relatively large nondipole ionization yield is primarily associated with the emission of low-energy photoelectrons [18,32].

Having settled that beyond-dipole corrections must be taken into account, we now proceed to investigate the role of relativistic effects in the underlying dynamics. This is accomplished by solving the TDDE within the LWA by using the propagation gauge Hamiltonian (9). The results are

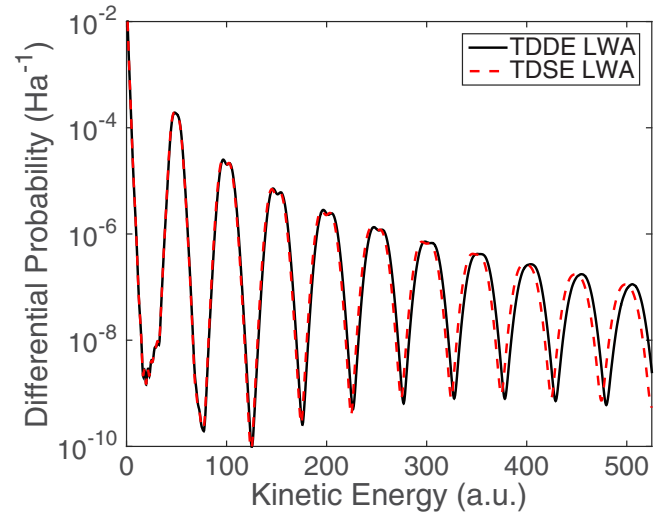


FIG. 2. Kinetic-energy spectrum of the emitted photoelectron for a 15 cycle laser pulse with  $\omega = 50$  a.u. and  $E_0 = 1000$  a.u. Solid black line: the fully relativistic result obtained by solving the TDDE with the Hamiltonian in Eq. (9) within the LWA. Dashed red line: the corresponding nonrelativistic result as obtained by solving the TDSE with the Hamiltonian in Eq. (7) within the LWA.

shown in Fig. 2 for both the relativistic and nonrelativistic calculation, respectively. As a matter of fact, the figure clearly manifests that relativistic corrections are important in that the multiphoton spectrum is effectively shifted to higher energies, i.e., the electron effectively acquires a higher energy than what is predicted in the corresponding nonrelativistic theory. Furthermore, the figure shows that this positive relativistic energy shift (blueshift) becomes more and more pronounced the higher the order of the multiphoton resonance.

As a next step, we now demonstrate the usefulness of the semirelativistic formulation for studying relativistic laser-matter interactions. Therefore, in Fig. 3 we show a comparison of the kinetic-energy spectrum of the emitted photoelectron as obtained by solving the TDSE with the semirelativistic Hamiltonian given in Eq. (8), and the corresponding result predicted by the fully relativistic Dirac equation. Both results are obtained beyond the dipole approximation but within the LWA. The agreement between the two independent calculations is astonishingly good, bearing in mind that we are actually comparing TDSE and TDDE calculations on an equal footing, both approaches revealing a highly relativistic ionization behavior of the system. The almost perfect match between the two independent calculations clearly proves the usefulness of the semirelativistic Schrödinger formulation via the Hamiltonian (8) for studying relativistic laser-matter interactions, an approach that was only recently established [27].

In solving the Dirac equation within the LWA, the spin-orbit coupling as well as the interaction of the electron's spin with the magnetic field are all taken into account in lowest order. As such, one could interpret the close agreement between the semirelativistic Schrödinger result and Dirac result in Fig. 3 as an indirect proof that the electron's spin degrees of freedom is of less importance in the present ionization context, i.e., electron's spin might be influenced by the field

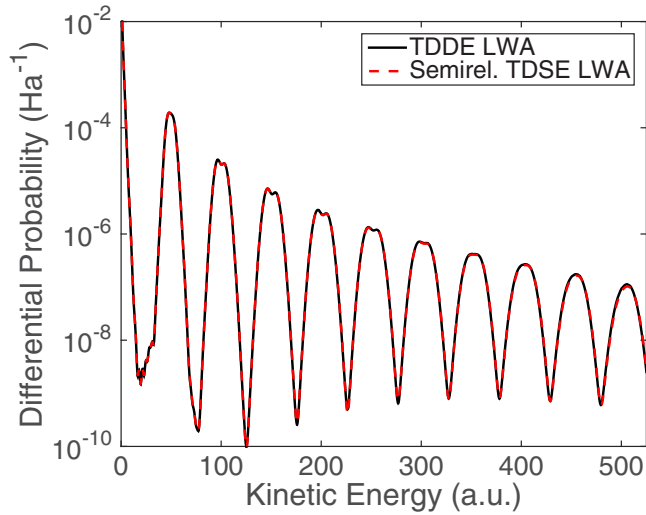


FIG. 3. Kinetic-energy spectrum of the emitted photoelectron for a 15 cycle laser pulse with  $\omega = 50$  a.u. and  $E_0 = 1000$  a.u. Solid black line: the fully relativistic result obtained by solving the TDDE with the Hamiltonian in Eq. (9) within the LWA. Dashed red line: the corresponding semirelativistic result as obtained by solving the TDSE with the Hamiltonian in Eq. (8) within the LWA.

but nevertheless this does not alter the relevant observable, in this case the energy of the photoelectron.

Figure 4 shows the updated energy distribution as obtained when going beyond the LWA and employing the Hamiltonian (13). Also shown is the corresponding LWA result. Both spectra are obtained with the semirelativistic Schrödinger equation, i.e., relativistic effects are taken into account. The

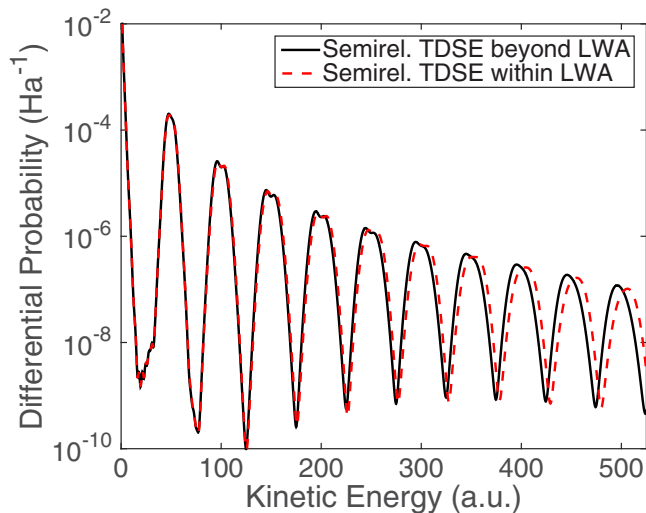


FIG. 4. Kinetic-energy spectrum of the emitted photoelectron for a 15 cycle laser pulse with  $\omega = 50$  a.u. and  $E_0 = 1000$  a.u. Solid black line: the semirelativistic result obtained by solving the TDSE with the Hamiltonian in Eq. (8) beyond the LWA, i.e., by applying the Hamiltonian (13). Dashed red line: the corresponding semirelativistic result as obtained by solving the TDSE with the Hamiltonian in Eq. (8) within the LWA.

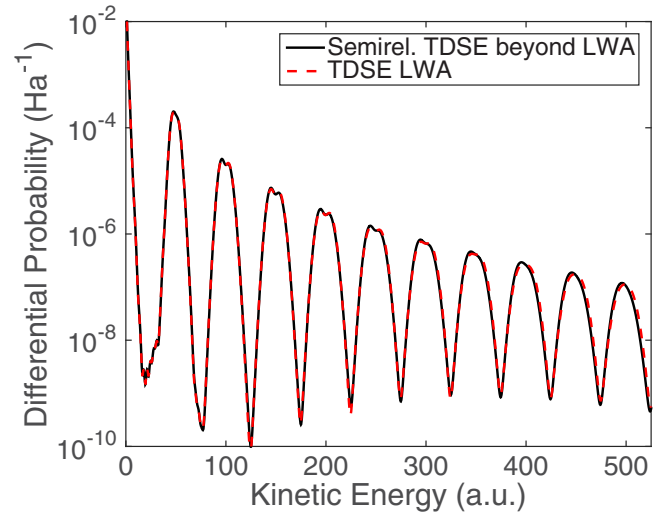


FIG. 5. Kinetic-energy spectrum of the emitted photoelectron for a 15 cycle laser pulse with  $\omega = 50$  a.u. and  $E_0 = 1000$  a.u. Solid black line: the semirelativistic result obtained by solving the TDSE with the Hamiltonian in Eq. (8) beyond the LWA, i.e., by applying the Hamiltonian (13). Dashed red line: the corresponding nonrelativistic result as obtained by solving the TDSE with the Hamiltonian in Eq. (7) within the LWA.

figure clearly shows that the beyond-LWA spectrum (solid black line) is shifted to lower energies as compared with the corresponding LWA result (red dashed line), i.e., the main effect of including the spatial dependence in the  $\mathbf{A} \cdot \mathbf{p}$  operator is to impose a redshift on the spectrum. This finding is in clear opposition to the relativistic blueshift observed in Fig. 2. Furthermore, comparing Figs. 2 and 4 it is seen that the positive relativistic energy shift is of the same order of magnitude as the negative beyond-LWA shift—but in opposite directions, i.e., they therefore partly cancel each other out. As it turns out, this is an extremely interesting finding in that we have now actually demonstrated that relativistic effects, whose relative importance is usually considered to be of order  $1/c^2$  (in atomic units), in fact turn out to be equally important as the leading-order beyond-dipole correction in the  $\mathbf{A} \cdot \mathbf{p}$  operator, a correction whose significance is usually considered to be of order  $1/c$ . This observation is imperative and might have implications for how one should proceed in order to include beyond-dipole and relativistic corrections terms to the Hamiltonian in a self-consistent manner, i.e., imposing spatial dependence in the  $\mathbf{A} \cdot \mathbf{p}$  operator should generally not be pursued unless the impact of relativistic effects have been examined—and vice versa.

Finally, we now explicitly demonstrate that the two effects, i.e., the relativistic blueshift and the (nonrelativistic) beyond-LWA redshift, are off the same order of magnitude and add destructively. To this end, Fig. 5 depicts a comparison between the fully relativistic result as obtained beyond the LWA by applying the Hamiltonian (13) and the corresponding nonrelativistic LWA result obtained with the Hamiltonian (7). Noticing the clear resemblance of the two results and keeping in mind that the black curve includes both the relativistic blueshift and the beyond-LWA redshift—while both effects

are neglected in the red dashed line curve—it becomes evident that the two shifts, to a large extent, cancel each other.

#### IV. CONCLUSION

Using two independent *ab initio* numerical approaches, we have studied the breakdown of the nonrelativistic approximation in the multiphoton ionization of ground-state hydrogenic atoms by some intense x-ray laser pulse. First, the time-dependent Dirac equation is solved for the laser-atom interaction within the so-called long-wavelength approximation and the propagation gauge. Then, the results of the fully relativistic calculations are compared with the corresponding results obtained with the time-dependent Schrödinger equation and within the framework of a recently developed semirelativistic approximation [27]. The main finding is that relativistic effects are important in the present ionization scenario—merely demonstrating the breakdown of the nonrelativistic approach. Furthermore, it is shown that the semirelativistic Schrödinger equation yields results in excellent agreement with those of the Dirac equation. It is found that relativistic effects are primarily responsible for an apparent blueshift (positive-energy shift) of the resulting multiphoton ionization spectrum.

Next, going beyond the long-wavelength approximation, it is shown that higher-order nondipole effects beyond the lowest-order magnetic contribution cannot be neglected in the

present ionization dynamics. We show that these higher-order effects give rise to an energy shift in the complete opposite direction than the relativistic blueshift, i.e., the spectrum becomes redshifted due to higher-order beyond-dipole corrections. Finally, we demonstrate that the relativistic blueshift and the higher-order beyond-dipole redshift are of equal order of magnitude, i.e., the two shifts effectively cancel each other out and the net effect is only very small. Hence, the distinction made in the present work between a relativistic blueshift and a corresponding beyond-dipole redshift might only be of theoretical interest, as the two shifts could nevertheless hardly be disentangled experimentally.

Nonetheless, we believe that the observed cancellation effect is important and has clear implications for how one should proceed in order to impose relativistic and higher-order beyond-dipole corrections into the dynamical equations of motion in a consistent manner. For example, imposing spatial dependence in the vector potential in the  $\mathbf{A} \cdot \mathbf{p}$  operator in Eq. (7) should generally not be pursued until the impact of relativistic effects have been examined. Likewise, the long-wavelength approximation should generally not be assumed in Eq. (8) before the role of higher-order beyond dipole corrections has been ruled out.

Although in this work we have only considered ionization in the x-ray regime at very high laser intensities, we do believe that similar conclusions may also be drawn for longer-wavelength light.

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