Semirelativistic Schrödinger Equation for Relativistic Laser-Matter Interactions

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A semirelativistic formulation of light-matter interaction is derived using the so called propagation gauge and the relativistic mass shift. We show that relativistic effects induced by a superintense laser field can, to a surprisingly large extent, be accounted for by the Schrödinger equation, provided that we replace the rest mass in the propagation gauge Hamiltonian by the corresponding time-dependent field-dressed mass. The validity of the semirelativistic approach is tested numerically on a hydrogen atom exposed to an intense extreme ultraviolet laser pulse strong enough to accelerate the electron towards relativistic velocities. It is found that while the results obtained from the ordinary (nonrelativistic) Schrödinger equation generally differ from those of the Dirac equation, demonstrating that relativistic effects are significant, the semirelativistic formulation provides results in quantitative agreement with a fully relativistic treatment.

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Triggered by rapid technological advances [1–4] and new infrastructure projects [5] there is an increased interest in the description of quantum systems exposed to superintense laser fields. In spite of the importance to address the relativistic regime [6] there are comparatively few such studies reported in the literature, probably due to the very nature of the time-dependent Dirac equation which is notoriously hard to solve.

Several issues make the Dirac equation tougher to solve numerically than its nonrelativistic counterpart, the Schrödinger equation. The fact that the numerical space is increased by a factor of 4 owing to the four components of the Dirac wave function, as opposed to a scalar wave function in the nonrelativistic case, is but the least of problems. The existence of a negative energy continuum is harder to tackle—for several reasons [7,8]. First, many numerical time integration techniques require a numerical time step restricted by the inverse of the rest-mass energy of the particle at hand, thus rendering calculations for realistic laser pulses infeasible. Second, spurious states may contaminate the spectrum of the numerical representation of the Hamiltonian.

Another, more subtle complication is the fact that inclusion of the spatial dependence of the external field, which is imperative for ultrastrong fields [9], is harder to achieve in a consistent manner for the Dirac equation than is the case for the Schrödinger equation [10]. While this particular challenge to a large extent has been lifted by introducing the so-called *propagation gauge* to the Dirac equation [11–13], a formulation of the Schrödinger equation which allows us to include relativistic effects is still desirable, albeit seemingly too much to hope for. However, as it turns out, the apparently naive approach of simply substituting the rest mass with the relativistic mass of the electron, does in fact, to a surprisingly large degree, accommodate for relativistic effects induced by external electromagnetic fields.

In the following, we outline the theoretical framework. The semirelativistic interaction is derived in three ways: first directly from the classical Hamiltonian function, then from the Dirac equation, and, finally, from the Klein-Gordon equation. While the full spatial dependence of the external field is included in the first and last approach, the derivation from the Dirac Hamiltonian is subject to the so-called *long wavelength approximation* [12,14], which is not to be confused with the *dipole approximation*. Our numerical results, comparing the fully relativistic and semirelativistic approaches, are then presented. Here, we have also included results obtained from nonrelativistic calculations in order to demonstrate that the studied cases indeed feature relativistic effects. Finally, we present our conclusions. Atomic units are used where stated explicitly.

First, we will take the following relativistic Hamiltonian for a particle of charge $q = -e$ as our starting point:

$$H_0 = \sqrt{m^2c^4 + p^2c^2} - mc^2 - e\varphi(\mathbf{r}), \tag{1}$$

which would act on a scalar wave function, as opposed to a bispinor in the Dirac case. For an atom in the absence of any external field, the scalar potential φ is simply provided by the Coulomb potential $V = -e\varphi$.

Of course, both the Dirac and Klein-Gordon formulations are closely related to the above Hamiltonian. The various ways we present for deriving the semirelativistic

form from these formulations are, however, rather different. In all cases it is crucial that the interaction is formulated within the propagation gauge—for reasons which will be transparent shortly.

We take our external field $\mathbf{A}$ to be linearly polarized and satisfying the Coulomb gauge restriction,

$$\mathbf{A} = A(\eta)\hat{\mathbf{A}} \quad \text{with} \quad \eta = \omega t - \mathbf{k}\cdot\mathbf{r}, \tag{2}$$

where the unit vectors $\hat{\mathbf{k}}$ and $\hat{\mathbf{A}}$ are orthogonal. The propagation gauge formulation of the interaction is obtained from the usual minimal coupling formulation by imposing the following gauge transformation [11–13]:

$$\boldsymbol{A} \to \boldsymbol{A} + \nabla\xi \quad \text{and} \quad \varphi \to \varphi - \frac{\partial}{\partial t}\xi, \quad \text{with}$$
$$\xi(\eta) = -\frac{e}{2m\omega}\int_{-\infty}^{\eta}[A(\eta')]^2 d\eta'. \tag{3}$$

The corresponding kinetic momentum is now

$$\mathbf{d} = \mathbf{p} + e\mathbf{A} + \frac{e^2}{2mc}A^2\hat{\mathbf{k}}. \tag{4}$$

This momentum shift is such that a free, nonquantum mechanical electron starting at zero momentum, remains at zero momentum. This applies both to the polarization direction and the propagation direction of the external field, not only to the polarization direction, as is the case in the usual minimal coupling formulation.

By introducing the interaction with the external field via minimal coupling, $\mathbf{p} \to \mathbf{p} + e\mathbf{A}$, and then imposing the gauge transformation Eq. (3), the Hamiltonian of Eq. (1) takes the form

$$H = \sqrt{m^2c^4 + d^2c^2} - \left(m + \frac{e^2}{2mc^2}A^2\right)c^2 + V$$
$$= \sqrt{\mu^2c^4 + q^2c^2} - \mu c^2 + V, \tag{5}$$

where we have introduced

$$\mu = m\left(1 + \frac{e^2}{2m^2c^2}A^2\right) \quad \text{and} \tag{6}$$

$$q^2 = p^2 + 2e\mathbf{A}\cdot\mathbf{p} + \frac{e^2}{2mc}\{A^2, \hat{\mathbf{k}}\cdot\mathbf{p}\}. \tag{7}$$

The η-dependent effective mass μ, which is not to be confused with the reduced mass, coincides with the time-dependent relativistic mass of a free, classical electron in the field initially at rest [15]. We now expand the square root in a manner which ensures hermicity term by term:

$$H = V + \frac{c^2}{2}\left(\mu\sqrt{1 + \mu^{-2}\frac{q^2}{c^2}} + \sqrt{1 + \frac{q^2}{c^2}\mu^{-2}}\,\mu\right) - \mu c^2$$
$$= V + \frac{1}{2}\left[\left\{\frac{1}{2\mu}, q^2\right\} - \left\{\frac{1}{8\mu c^2}, q^2\mu^{-2}q^2\right\} + \cdots\right]. \tag{8}$$

Truncation at lowest order yields

$$H \approx \frac{1}{2}\left\{\frac{1}{2\mu}, p^2\right\} + \frac{e}{\mu}\mathbf{A}\cdot\mathbf{p} + \frac{e^2}{8mc}\left\{\frac{1}{\mu}, \{A^2, \hat{\mathbf{k}}\cdot\mathbf{p}\}\right\} + V, \tag{9}$$

where the anticommutators persist due to the fact that the relativistic mass μ is spatially dependent and, hence, does not commute with the momentum operator.

We argue that the leading order term in Eq. (8) indeed includes most of the relativistic correction to the ionization probability. Now, one may rightfully question how an expansion of the kinetic energy can meaningfully be truncated at lowest order in p^2 in the relativistic region. This is precisely why it is crucial that the interaction is formulated in the propagation gauge. Note that for an initial wave packet with $\langle\mathbf{p}\rangle = \mathbf{0}$ in the absence of any Coulomb potential, the expectation value of the canonical momentum will remain identical to zero at all times in the propagation gauge. In the presence of the Coulomb potential this is, of course, no longer true, and one can only assume that $\langle\mathbf{p}\rangle \simeq 0$. However, in the strong field limit, the Coulomb potential eventually represents only a small perturbation, with the result that one can safely neglect higher order terms in the momentum operator with respect to the leading order term. Thus, we expect that the validity of the Hamiltonian of Eq. (9) in fact will *increase* with increasing intensity. This is also reflected in the fact that the expansion parameters in Eq. (8), i.e., $\mu^{-2}q^2/c^2$ and $q^2/c^2\cdot\mu^{-2}$, decrease with increasing field strength.

In the present approach, relativistic corrections to the Coulomb interaction and spin are not accounted for. However, we do expect it to correctly accommodate for transient relativistic effects induced by the external laser field as long as the probability of real pair production is negligible.

The expansion of Eq. (8) becomes simpler when q^2 and μ commute. This is the case when Eq. (5) is taken to be a classical function rather than an operator. It is also the case within the long wavelength approximation (LWA), in which the spatial dependence of the vector potential $\mathbf{A}$ and, thus, also in μ is neglected:

$$H = V + \frac{q^2}{2\mu} - \frac{q^4}{8c^2\mu^3} + \cdots. \tag{10}$$

If we, again, retain only the leading term in kinetic energy, we obtain

$$H \approx \frac{q^2}{2\mu} + V = \frac{p^2}{2\mu} + \frac{e}{\mu}\mathbf{A}\cdot\mathbf{p} + \frac{e^2}{2\mu mc}A^2\hat{\mathbf{k}}\cdot\mathbf{p} + V. \tag{11}$$

We emphasize that the LWA is far less restrictive than the much applied dipole approximation. While the assumption of a homogeneous vector potential imposed within the propagation gauge does not distort the leading magnetic interaction, no magnetic interaction at all is included with a homogeneous vector potential imposed within the minimal coupling, or *velocity gauge*, formulation.

The Hamiltonian equations (9) and (11) coincide with the nonrelativistic Schrödinger Hamiltonian in the propagation gauge upon the substitution $\mu \to m$. The dynamics corresponding to these Hamiltonians is thus the same as that of the nonrelativistic equations, albeit with the increased effective mass that the electron acquires in the field rather than the bare rest mass. Note, however, that the opposite substitution, i.e., $m \to \mu$ in the nonrelativistic Schrödinger Hamiltonian, would *not* provide Eq. (11) due to the remaining dependence on the electron rest mass in the third term in Eqs. (9) and (11). This term accounts for the leading magnetic interaction in the strong field limit and its rest-mass dependence originates from the momentum induced in the direction of the electromagnetic field, i.e., the third term on the right-hand side of Eq. (4).

It is worth mentioning that the interaction form of Eq. (11) may rather easily be transformed into other more familiar forms, analogous to, e.g., the velocity gauge, the length gauge, or the Kramers-Henneberger frame [16,17].

Next, we will outline how the above Hamiltonian may be derived from the Dirac equation. In propagation gauge form it reads [13]

$$i\hbar\frac{d}{dt}\Psi = H\Psi \quad \text{with}$$
$$H = c\alpha\cdot\mathbf{d} + mc^2\beta + \left(V - \frac{e^2}{2m}A^2\right)\mathbb{1}_4, \quad (12)$$

where $\mathbf{d}$ is defined in Eq. (4). The wave function Ψ now is a four-component bispinor,

$$\Psi = \begin{pmatrix}\Phi\\ X\end{pmatrix}. \quad (13)$$

The upper spinor Φ is typically referred to as the *large* component for states with positive energy, while the lower spinor X is coined the *small* component. We apply the usual formulation in terms of Pauli matrices $\boldsymbol{\sigma}$ and identity matrices for the α and β matrices.

In a classic paper [18] Foldy and Wouthuysen showed how the Dirac equation for a free fermion may be transformed in a manner which decouples the large and the small component. If we, within the LWA, generalize their transformation by replacing the canonical momentum $\mathbf{p}$ with the time-dependent kinetic momentum $\mathbf{d}$, the transformation reads

$$T = e^S \quad \text{with} \quad S = \frac{1}{2d}\tan^{-1}\left(\frac{d}{mc}\right)\beta\alpha\cdot\mathbf{d}, \quad (14)$$

where $\mathbf{d}$, which is defined in Eq. (4), depends explicitly on the external field $\mathbf{A}$. With this, the Dirac Hamiltonian (12) is cast into

$$H' = e^S H e^{-S} + i\hbar\frac{d}{dt}S$$
$$= \beta\sqrt{m^2c^4 + d^2c^2} + e^S V e^{-S} - \frac{e^2}{2m}A^2 + i\frac{\hbar}{2}\beta\alpha\cdot\boldsymbol{\lambda}, \quad (15)$$

$$\text{where } \boldsymbol{\lambda} = \frac{d}{dt}\frac{\mathbf{d}}{d}\tan^{-1}\left(\frac{d}{mc}\right). \quad (16)$$

This form contains two nondiagonal terms: the transformed Coulomb potential and the last term, which originates from the time dependence of the transformation. If we disregard relativistic corrections to the Coulomb potential and keep only the diagonal leading term,

$$e^S V e^{-S} \approx V, \quad (17)$$

we neglect contributions such as the Darwin term and spin-orbit coupling. This should, however, be admissible for cases in which the dominant relativistic effects are the ones induced by the external field $\mathbf{A}$.

The last term in Eq. (15) is negligible in most cases. In order to see this, we rewrite the Dirac equation, Eq. (12), with the transformed Hamiltonian Eq. (15) in terms of the two components:

$$i\hbar\frac{d}{dt}\begin{pmatrix}\Phi\\ X\end{pmatrix} = H'\begin{pmatrix}\Phi\\ X\end{pmatrix}, \quad \text{with}$$
$$H' = \begin{pmatrix}\sqrt{\mu^2c^4 + q^2c^2} & i\frac{\hbar}{2}\boldsymbol{\sigma}\cdot\boldsymbol{\lambda}\\ -i\frac{\hbar}{2}\boldsymbol{\sigma}\cdot\boldsymbol{\lambda} & -\sqrt{\mu^2c^4 + q^2c^2}\end{pmatrix}$$
$$+ (V - \mu c^2)\mathbb{1}_4. \quad (18)$$

Here, we have shifted the energy downwards by mc^2 and used the same steps as in connection with Eq. (5). We now approximate

$$\left(-\sqrt{\mu^2c^4 + q^2c^2} - \mu c^2 + V - i\frac{d}{dt}\right)X \approx -2\,\mu c^2 X; \quad (19)$$

i.e., for the small component we neglect higher order terms in q^2, the Coulomb potential and the time derivative upon comparison with the dominating "dynamical mass energy" μc^2. Note, however, that the Coulomb potential in Eq. (18) is still included to leading order.

With this, Eq. (18) decouples and we are left with the following effective Hamiltonian for the large component Φ:

$$\sqrt{\mu^2c^4 + q^2c^2} - \mu c^2 + V + \frac{\hbar^2}{8\mu c^2}\lambda^2. \quad (20)$$

Here, we notice that the three first terms are identical to the right-hand side of Eq. (5). The remaining term, $\sim\lambda^2$, is to

leading order a purely time-dependent one, which can be removed by a phase transformation. The next to leading term is of order $m^{-4}c^{-5}$ and thus the last term of Eq. (20) may safely be neglected in view of the approximations already made when using Eq. (19). With this the Hamiltonian of Eq. (5) is reproduced for the large component Φ, albeit within the LWA. We emphasize that the equivalence with the scalar Hamiltonian of Eq. (5) implies the neglect of all spin-dependent interactions.

Finally, we will outline how Eq. (5) may be obtained from the Klein-Gordon equation. Within the propagation gauge, it takes on the form

$$\left(-\hbar^2\frac{d^2}{dt^2}+i\hbar\frac{d}{dt}K+Ki\hbar\frac{d}{dt}\right)\Psi=L\Psi, \tag{21}$$

where we have defined the operators

$$K=mc^2+\frac{e^2}{2m}A^2-V \quad \text{and} \tag{22}$$

$$L=p^2c^2+\left(e\mathbf{A}+\frac{e^2}{2mc}A^2\hat{\mathbf{k}}\right)\cdot\mathbf{p}c^2 + \mathbf{p}\cdot\left(e\mathbf{A}+\frac{e^2}{2mc}A^2\hat{\mathbf{k}}\right)c^2+V\left(2mc^2+\frac{e^2}{m}A^2-V\right), \tag{23}$$

and, again, shifted the energy downwards by the rest-mass energy mc^2. By insisting that the Hamiltonian H equals $i\hbar d/dt$, we arrive at

$$(H^2+HK+KH-L)\Psi=0. \tag{24}$$

The operator on the left-hand side above is identical to the zero operator if and only if

$$H=-K\pm\sqrt{K^2+L}. \tag{25}$$

Note that this Hamiltonian, just like the Dirac Hamiltonian, provides both a positive and a negative energy spectrum. By selecting the form corresponding to positive energies we, again, arrive at Eq. (5), as may be verified by inspection.

Having derived a Schrödinger-like equation for the relativistic laser-matter interactions, we now aim at demonstrating its capabilities by performing a calculation on a concrete time-dependent problem and compare its result with the exact one as obtained by the Dirac equation. To this end, we will investigate the following scenario: A hydrogen atom with the electron initially prepared in the ground state is exposed to a laser pulse, defined as

$$\mathbf{A}(\eta)=A(\eta)\hat{\mathbf{z}}=\frac{E_0}{\omega}f(\eta)\sin(\eta+\phi)\hat{\mathbf{z}}, \tag{26}$$

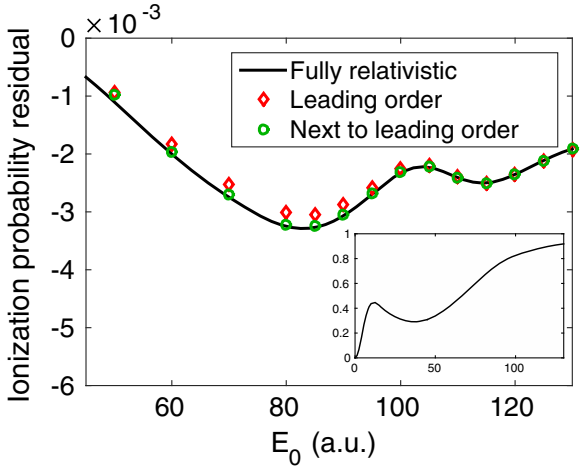

FIG. 1. A hydrogen atom initially prepared in the ground state is exposed to a 15 cycle laser pulse with a central frequency of $\omega = 3.5$ a.u. The peak field strength E_0 ranges from 50 to 130 a.u. The black curve is the *difference* in ionization probability predicted by the fully relativistic Dirac equation and the nonrelativistic Schrödinger equation, within the long wavelength approximation. The red diamonds show the corresponding difference for results obtained using the semirelativistic approach with the kinetic energy truncated at lowest order, Eq. (11), while the green circles are obtained with the next to leading order correction in Eq. (10) included. The inset shows the *total* ionization probability.

where the envelope function f is chosen to be a sine squared.

Both the Dirac equation and the Schrödinger equation are solved within the LWA. In the latter case, calculations have been performed both with and without the relativistic mass shift included in order to see whether this shift can accommodate for relativistic effects. For details on the implementation, see Refs. [10,13]. The validity of the LWA was demonstrated explicitly in the latter.

The results for a 15 cycle laser pulse with photon energies in the extreme ultraviolet region are shown in Fig. 1. We find relativistic corrections of about 0.5% in the ionization probability. We see that the bulk of the correction is indeed provided by Eq. (11), while the next order corrections from Eq. (10) in fact vanishes beyond maximum field strengths E_0 of about 100 a.u. We have also verified numerically that the spin-orbit coupling, which was neglected in Eq. (17), is indeed unimportant.

Beyond the validity of the LWA, we have performed classical trajectory Monte Carlo simulations; i.e., we have resolved the dynamics according to a classical Hamiltonian function for a microcanonical ensemble of initial states corresponding to the ground state energy and estimated ionization probabilities by analyzing the outcome statistically [19,20]. This has been done for the same three cases as above: fully relativistic, fully nonrelativistic, and semirelativistic. In the semirelativistic case, our Hamiltonian function takes the same form as in Eq. (11), albeit with full spatial dependence in $\mathbf{A}$ and μ

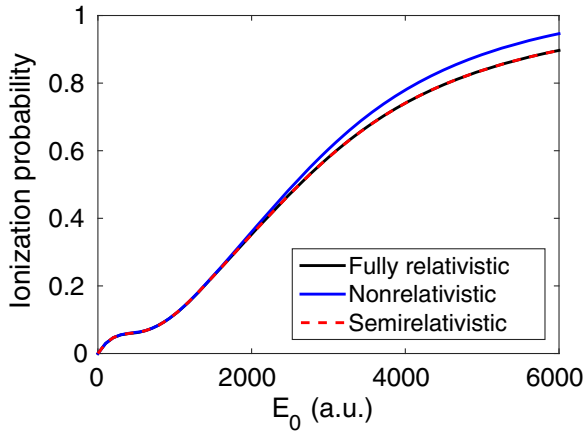

FIG. 2. The ionization probability as a function of peak electric field strength E_0, obtained by classical trajectory Monte Carlo calculations. The atom has been exposed to a 15 cycle laser pulse with central frequency $\omega = 50$ a.u. The calculations have been done both fully relativistically, fully nonrelativistically and by using the semirelativistic Hamiltonian function of Eq. (11) with the full spatial dependence of the laser field included.

in this case. In Fig. 2 we present ionization probabilities obtained from a calculation with a central frequency of $\omega = 50$ a.u., which corresponds to photons in the soft x-ray region. As we can see, the semirelativistic results are virtually indistinguishable from the fully relativistic ones, thus providing strong evidence of the validity of the present approach.

Additionally, we have calculated classical trajectories for electrons, initially at rest, only subject to the interaction with the external laser field, i.e., without any Coulomb interaction. This was done for a wide range of laser frequencies ω, ranging from the infrared region via the optical to the x-ray region. The same three approaches as above were applied. While the nonrelativistic Hamiltonian produced results deviating substantially from the fully relativistic ones, the semirelativistic formalism consistently gave trajectories virtually indistinguishable from the fully relativistic ones.

In conclusion, we have demonstrated that by substituting the mass with the *relativistic* mass in the adequate manner, the validity of the Schrödinger equation is extended into the relativistic region. The validity of the approach has been demonstrated by direct comparison with fully relativistic calculations, providing quantitative agreement. This high degree of accuracy is a surprising finding, and a very useful one indeed: within the long wavelength approximation, adjustment of the mass is easily implemented, thus extending the validity of theoretical studies considerably with little extra effort when it comes to implementation and numerics. The coincidence between the fully relativistic and the semirelativistic calculations shows that the relativistic corrections may be attributed to the increased inertia induced by the external laser field.

As a next step, a derivation based on the Dirac equation beyond the long wavelength approximation will, hopefully, provide a relativistic Hamiltonian of Schrödinger type which also features spin dynamics.

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[1] C. I. Moore, A. Ting, S. J. McNaught, J. Qiu, H. R. Burris, and P. Sprangle, Phys. Rev. Lett. **82**, 1688 (1999).
[2] A. D. DiChiara, I. Ghebregziabher, R. Sauer, J. Waesche, S. Palaniyappan, B. L. Wen, and B. C. Walker, Phys. Rev. Lett. **101**, 173002 (2008).
[3] H. Yumoto, H. Mimura, T. Koyama, S. Matsuyama, K. Tono, T. Togashi, Y. Inubushi, T. Sato, T. Tanaka, T. Kimura *et al.*, Nat. Photonics **7**, 43 (2013).
[4] H. Yoneda, Y. Inubushi, M. Yabashi, T. Katayama, T. Ishikawa, H. Ohashi, H. Yumoto, K. Yamauchi, H. Mimura, and H. Kitamura, Nat. Commun. **5**, 5080 (2014).
[5] A. Thoss, *ELI White Book*, edited by G. A. Mourou, G. Korn, W. Sandner, and J. L. Collier (ELI, Berlin, 2011).
[6] A. Di Piazza, C. Müller, K. Z. Hatsagortsyan, and C. H. Keitel, Rev. Mod. Phys. **84**, 1177 (2012).
[7] S. Selstø, E. Lindroth, and J. Bengtsson, Phys. Rev. A **79**, 043418 (2009).
[8] Y. V. Vanne and A. Saenz, Phys. Rev. A **85**, 033411 (2012).
[9] H. R. Reiss, Phys. Rev. A **63**, 013409 (2000).
[10] T. Kjellsson, S. Selstø, and E. Lindroth, Phys. Rev. A **95**, 043403 (2017).
[11] A. S. Simonsen and M. Førre, Phys. Rev. A **93**, 063425 (2016).
[12] M. Førre and A. S. Simonsen, Phys. Rev. A **93**, 013423 (2016).
[13] T. Kjellsson, M. Førre, A. S. Simonsen, S. Selstø, and E. Lindroth, Phys. Rev. A **96**, 023426 (2017).
[14] J. R. Vázquez de Aldana, N. J. Kylstra, L. Roso, P. L. Knight, A. Patel, and R. A. Worthington, Phys. Rev. A **64**, 013411 (2001).
[15] E. S. Sarachik and G. T. Schappert, Phys. Rev. D **1**, 2738 (1970).
[16] S. Selstø and M. Førre, Phys. Rev. A **76**, 023427 (2007).
[17] A. D. Bandrauk, F. Fillion-Gourdeau, and E. Lorin, J. Phys. B **46**, 153001 (2013).
[18] L. L. Foldy and S. A. Wouthuysen, Phys. Rev. **78**, 29 (1950).
[19] L. N. Gaier and C. H. Keitel, Phys. Rev. A **65**, 023406 (2002).
[20] S. Borbély, K. Tőkési, and L. Nagy, Phys. Rev. A **77**, 033412 (2008).