Generalized velocity-gauge form of the light-matter interaction Hamiltonian beyond the dipole approximation

Morten Førre* and Aleksander Skjerlie Simonsen†
Department of Physics and Technology, University of Bergen, N-5007 Bergen, Norway
(Received 22 October 2015; revised manuscript received 14 December 2015; published 25 January 2016)

The exact velocity-gauge minimal-coupling Hamiltonian describing the laser-matter interaction is transformed into another form by means of a series of gauge transformations. The Hamiltonian corresponding to this point of view is valid for an arbitrary time- and space-dependent laser field, also known as a nondipole field. In effect, the Hamiltonian represents a generalization of the original velocity-gauge minimal-coupling Hamiltonian in the sense that the particle's (classical) velocity in the laser propagation direction is also explicitly accounted for by a new operator term. Imposing the so-called long-wavelength approximation (LWA) on the field, i.e., assuming the laser wavelength being much larger than the extent of the atomic system, the spatial dependence of the field can be neglected and the interaction Hamiltonian reduces to a simpler form. Nevertheless, the resulting LWA Hamiltonian includes the effect of the magnetic-field component of the laser, which is in clear contrast with the usual dipole approximation Hamiltonian derived by imposing the LWA directly on the initial velocity-gauge minimal-coupling Hamiltonian. As such, the weak-field condition necessary to justify neglecting the magnetic field, and the LWA condition, can be considered independently in this formalism, making it an attractive alternative for a broad range of applications in strong-field physics. We demonstrate that, from a numerical perspective, this form of the light-matter interaction is advantageous compared to its standard velocity-gauge counterpart as it gives rise to faster convergence properties when describing ionization dynamics in superintense fields beyond the dipole approximation.

DOI: 10.1103/PhysRevA.93.013423

The time-dependent dynamics of matter in interaction with a laser field is usually described within the so-called semiclassical approximation. In this approximation, and due to the vast number of photons involved, the laser field may be considered equivalent to a classical electromagnetic field [1], meaning that the formalism of second quantization, i.e., the quantization of the electromagnetic radiation [2], is not necessary in describing the laser-matter interaction. Nevertheless, the dynamics of the atomic system itself is treated fully quantum mechanically by means of the time-dependent Schrödinger equation, or in the relativistic limit, the time-dependent Dirac equation.

The laser-matter interaction is often simplified by imposing the so-called dipole approximation, one of the most frequently used approximations in theoretical physics. Here several equivalent formulations exist, the most common ones being the velocity gauge, the length gauge, and the Kramers-Henneberger frame; see, e.g., [3] and references therein. In the dipole approximation the laser field is treated as a homogeneous time-varying electric field, i.e., any spatial dependences of the electric field as well as the entire magnetic component of the laser are neglected altogether. The dipole approximation is a type of long-wavelength approximation (LWA) in that it is likely to be valid whenever the laser wavelength is much larger than the relevant atomic dimensions in question. However, justifying the dipole approximation approach requires, in addition to the long-wavelength condition, that the laser intensity should not be so high that the homogeneous magnetic-field component [4–11] and/or relativistic effects [12–14] come into play.

It is generally well known that among the equivalent versions of the dipole approximation, the velocity-gauge formulation of quantum mechanics is often beneficial in the description of matter interacting with electromagnetic fields. Now how does this comply with the gauge principle, merely stating that all physical observables are gauge invariant? Of course the gauge principle cannot be violated, meaning that two representations, e.g., the velocity- and length-gauge formulations, must yield identical results in an exact treatment. However, as most practical approaches involve at least *some* level of approximation, that being either an intentional restriction, for instance, the LWA, or simply truncation of the exact problem, the choice of gauge can have critical impact on the modeling of the physical processes. In the case of the velocity gauge, it is often found to be the more convenient choice due to its faster convergence criteria from the point of view of a discrete representation, in particular in the strong-field limit.

With the development of x-ray lasers, for instance, the European XFEL that is currently under construction at the free-electron laser facility at DESY in Hamburg, the wavelength of the laser field will ultimately become so short that the validity of the dipole approximation becomes questionable [15]. The European XFEL will generate extremely intense x-ray pulses in the wavelength range from 0.05 to 4.7 nm, opening up many new opportunities for manipulating and controlling matter at the most fundamental level and paving the way for studies of atomic and molecular systems under extreme nonperturbative conditions. Nondipole effects induced by the magnetic field have already been observed experimentally in laser-matter interactions in the optical regime [16,17]. Motivated by the current developments and the need to include beyond-dipole (nondipole) effects for a proper description of the underlying dynamics, and considering the question of gauge choice in theoretical modeling, we here propose an

*morten.forre@uib.no
†aleksander.simonsen@uib.no

alternative velocity-gauge form of the light-matter interaction valid beyond the dipole approximation, thus revisiting the question of equivalent formulations of electrodynamics.

In this work we transform the nondipole minimal-coupling Hamiltonian of a massive charged particle in an explicitly space- and time-dependent electromagnetic field into another form by executing a sequence of (unitary) gauge transformations. The transformed Hamiltonian takes a relatively simple form that proves to be both useful and appealing in the description of strong-field nondipole dynamics. The Hamiltonian from this alternative point of view contains an operator of the form $\hat{\boldsymbol{k}}\cdot\boldsymbol{p}$ ($\hat{\boldsymbol{k}}$ being a unit vector pointing in the laser propagation direction), whose prefactor happens to be equal to the velocity component along the direction of propagation for the corresponding classical (free) particle subjected only to the laser field. As such, we will refer to the resulting gauge as the propagation gauge.

In the nonrelativistic limit, the evolution of a wave packet representing a particle of mass m and charge q in a (Coulomb) potential V and laser field $\boldsymbol{A}$ is governed by the time-dependent Schrödinger equation $i\hbar\partial_t\Psi_0 = H_0\Psi_0$, with the usual exact minimal-coupling Hamiltonian

$$H_0 = \frac{1}{2m}[\boldsymbol{p} - q\boldsymbol{A}(\eta)]^2 + V(\boldsymbol{r}), \tag{1}$$

where the classical electric $\boldsymbol{E}$ and magnetic $\boldsymbol{B}$ fields are given by $\boldsymbol{E} = -\partial_t\boldsymbol{A}$ and $\boldsymbol{B} = \nabla\times\boldsymbol{A}$, respectively. Here we require that $\boldsymbol{A}(\eta)$ satisfies the wave equation and depends on both space and time coordinates in the following way: $\eta = \omega t - \boldsymbol{k}\cdot\boldsymbol{r}$, where ω is the angular frequency of the field, $\boldsymbol{k} = \omega/c\,\hat{\boldsymbol{k}}$ is the wave vector, and c is the speed of light. Imposing the Coulomb gauge restriction $\nabla\cdot\boldsymbol{A} = 0$ on the field, the Hamiltonian is cast in the form

$$H_0 = \frac{p^2}{2m} + V - \frac{q}{m}\boldsymbol{A}\cdot\boldsymbol{p} + \frac{q^2}{2m}\boldsymbol{A}^2, \tag{2}$$

where the velocity-gauge form of the Hamiltonian is recognized by the $\boldsymbol{A}\cdot\boldsymbol{p}$ interaction term. While the $\boldsymbol{A}^2$ term in the Hamiltonian (2) is not contributing to the light-matter interaction within the dipole approximation, it is well known that it plays a central role in the description of the nondipole dynamics of atoms and molecules in superintense laser fields [4–11,18–22]. In particular, the main nondipole contribution due to the magnetic field is accounted for by this operator. From a theoretical point of view, it is common to assume that the space-dependent vector potential can be expanded in powers of $\boldsymbol{k}\cdot\boldsymbol{r}$, where the term linear in $\boldsymbol{k}\cdot\boldsymbol{r}$ represents the dominant nondipole correction [4–11,15,18–22]. In this approach, the nondipole operator takes a similar form (up to a prefactor) as the ordinary dipole interaction operator as represented in the length-gauge formulation of the light-matter interaction, i.e., $-q\boldsymbol{E}\cdot\boldsymbol{r}$ [3], except that its line of action is now directed along the laser propagation direction instead of the laser polarization axis.

In our alternative formulation of the light-matter interaction, the last term in Eq. (2) is in effect substituted by a velocity-gauge-like operator, acting in the direction of propagation. To this end, we define a recursive sequence of gauge transformations of the form

$$\Psi_n = U_n\Psi_{n-1} = e^{i\alpha_n}\Psi_{n-1}, \tag{3}$$

with

$$\alpha_n(\eta) = a_n\frac{mc^2}{\hbar\omega}\int_{-\infty}^{\eta}\left[\frac{q\boldsymbol{A}(\eta')}{mc}\right]^{2n}d\eta', \tag{4}$$

where a_n are real transformation weights for $n = 1,2,3,\ldots$. Gauge invariance is maintained in each step by transforming the wave function under the unitary transformations, resulting in different corresponding Hamiltonians in the time-dependent Schrödinger equation. In each transformation step, the Hamiltonian from the new point of view is then given recursively by

$$H_n = U_nH_{n-1}U_n^\dagger + i\hbar\dot{U}_nU_n^\dagger, \tag{5}$$

where H_n defines the "new" Hamiltonian that is derived from the "old" Hamiltonian H_{n-1} by means of the transformation U_n. Using the operator identity known as the Baker-Hausdorff lemma [23],

$$e^{ia}be^{-ia} = b + \frac{i}{1!}[a,b] + \frac{i^2}{2!}[a,[a,b]] + \frac{i^3}{3!}[a,[a,[a,b]]] + \cdots, \tag{6}$$

$[a,b] = ab - ba$ denoting the commutation relationship between two Hermitian operators a and b, the effect of the unitary transformation U_n is evaluated. The resulting Hamiltonian H_n is related to its predecessor H_{n-1} by

$$\begin{aligned}H_n =& H_{n-1} - a_nmc^2\left(\frac{q\boldsymbol{A}}{mc}\right)^{2n} + a_n^2\frac{mc^2}{2}\left(\frac{q\boldsymbol{A}}{mc}\right)^{4n}\\ &+ a_n\frac{c}{2}\left\{\left(\frac{q\boldsymbol{A}}{mc}\right)^{2n},\hat{\boldsymbol{k}}\cdot\boldsymbol{p}\right\} + a_nmc^2\sum_{i=1}^{n-1}a_i\left(\frac{q\boldsymbol{A}}{mc}\right)^{2(n+i)},\end{aligned} \tag{7}$$

where curly brackets denote the anticommutator notation $\{a,b\} = ab + ba$.

When applying Eq. (7) to the original velocity-gauge Hamiltonian (2) and choosing $a_1 = 1/2$, the following expression for H_1 is obtained:

$$\begin{aligned}H_1 =& \frac{p^2}{2m} + V - \frac{q}{m}\boldsymbol{A}\cdot\boldsymbol{p} + \frac{1}{4}\left(\frac{q^2\boldsymbol{A}^2}{m^2c}\hat{\boldsymbol{k}}\cdot\boldsymbol{p} + \hat{\boldsymbol{k}}\cdot\boldsymbol{p}\frac{q^2\boldsymbol{A}^2}{m^2c}\right)\\ &+ \frac{1}{8}\frac{q^4}{m^3c^2}\boldsymbol{A}^4.\end{aligned} \tag{8}$$

Comparing Eqs. (2) and (8), we note that the $\boldsymbol{A}^2$ term in the former has been replaced by the $\boldsymbol{A}^2$ ($\hat{\boldsymbol{k}}\cdot\boldsymbol{p}$) and ($\hat{\boldsymbol{k}}\cdot\boldsymbol{p}$) $\boldsymbol{A}^2$ terms in the latter. In addition, due to the transformation, a higher-order term proportional to $\boldsymbol{A}^4$ has emerged in Eq. (8). Now, in order to eventually arrive at a Hamiltonian formulation where only those light-matter interaction terms explicitly containing the momentum operator $\boldsymbol{p}$ are retained, i.e., successively removing terms proportional to $\boldsymbol{A}^{2n}$, the transformation (3) is repeated with $n = 2,3,4,\ldots$. To this end, the corresponding transformation weights must be defined in the following way:

$$a_n = \frac{1}{2}\sum_{i=1}^{n-1}a_ia_{n-i} = \binom{2n}{n}\frac{1}{4^n(2n-1)}, \tag{9}$$

with $a_1 = 1/2$ and $\binom{a}{b} = \frac{a!}{b!(a-b)!}$ being the binomial coefficient. Then, by applying the recursion formula (7) n times, the corresponding transformed Hamiltonian is simply given by

$$H_n = \frac{p^2}{2m} + V - \frac{q}{m}\boldsymbol{A}\cdot\boldsymbol{p} + \frac{c}{2}\sum_{i=1}^{n} a_i \left\{ \left(\frac{q\boldsymbol{A}}{mc}\right)^{2i}, \hat{\boldsymbol{k}}\cdot\boldsymbol{p} \right\} + \frac{mc^2}{2}\sum_{i=1}^{n}\left(\frac{q\boldsymbol{A}}{mc}\right)^{2(n+i)} \sum_{j=1}^{n-i+1} a_{i+j-1}a_{n-j+1}. \quad (10)$$

In a nonrelativistic treatment, one must require that $|q\boldsymbol{A}|/m < c$, i.e, the velocity of the particle in the field is never allowed to exceed the speed of light. This means that all the terms in the last (double) sum in Eq. (10) can be made arbitrarily small for increasing value of n, i.e., in the limit $n \to \infty$ the corresponding sum vanishes and the Hamiltonian (10) simplifies to

$$H \equiv \lim_{n\to\infty} H_n = \frac{p^2}{2m} + V - \frac{q}{m}\boldsymbol{A}\cdot\boldsymbol{p} + \frac{c}{2}\sum_{i=1}^{\infty} a_i \left\{ \left(\frac{q\boldsymbol{A}}{mc}\right)^{2i}, \hat{\boldsymbol{k}}\cdot\boldsymbol{p} \right\}. \quad (11)$$

The Hamiltonian (11) constitutes the main result of this work. It is valid for an arbitrary time- and space-dependent (nondipole) laser field $\boldsymbol{A}(\eta)$ provided the condition $|q\boldsymbol{A}|/m < c$ holds. However, since this condition is violated only for laser fields strong enough to accelerate the corresponding classical particle to superrelativistic velocities, *any* description of the light-matter interaction within the (nonrelativistic) time-dependent Schrödinger picture is defective *per se*. So, without loss of generality, the full Hamiltonian (11) is equivalent to the ordinary minimal-coupling Hamiltonian in Eq. (2), in the sense that they can be used interchangeably and their corresponding wave functions are related exactly by the unitary transformations (3). Comparing Eqs. (2) and (11), we note that the last term proportional to $\boldsymbol{A}^2$ in the first Hamiltonian has been replaced by an operator series with terms proportional to $\hat{\boldsymbol{k}}\cdot\boldsymbol{p}$ in the second. As a matter of fact, the sum over all the prefactors in the series expansion in Eq. (11) turns out to be equal to the component of the velocity in the propagation direction of the corresponding free (classical) particle moving in the laser field, i.e., the velocity in the propagation direction is simply given as

$$\boldsymbol{v}_{\hat{\boldsymbol{k}}}(\eta) = c\hat{\boldsymbol{k}}\sum_{i=1}^{\infty} a_i \left[\frac{q\boldsymbol{A}(\eta)}{mc}\right]^{2i}. \quad (12)$$

This is an important result that provides an interpretation of the terms in the Hamiltonian (11). Note the similarity between the two interaction terms $-q/m\boldsymbol{A}\cdot\boldsymbol{p}$ and $(\boldsymbol{v}_{\hat{\boldsymbol{k}}}\cdot\boldsymbol{p} + \boldsymbol{p}\cdot\boldsymbol{v}_{\hat{\boldsymbol{k}}})/2$, where $-q/m\boldsymbol{A}$ and $\boldsymbol{v}_{\hat{\boldsymbol{k}}}$ correspond to the velocity components of a classical particle subjected to the laser, along the directions of polarization and propagation, respectively. As such, we suggest that the Hamiltonian (11) represents a generalization of the original velocity-gauge minimal-coupling Hamiltonian (2), in that the velocity in the laser propagation direction is explicitly taken into account in the present formulation. Therefore, we will adopt the notion of the propagation gauge for the corresponding light-matter interaction scheme.

An important difference between the propagation gauge (11) and the velocity gauge (2) is revealed when we assume the LWA, i.e., $\boldsymbol{A}$ becomes a purely time-dependent function. Then Eq. (11) reduces to

$$H^{\mathrm{LWA}} = \frac{p^2}{2m} + V - \frac{q}{m}\boldsymbol{A}_0\cdot\boldsymbol{p} + \frac{c}{2}\left(\frac{q\boldsymbol{A}_0}{mc}\right)^2 \times \left[1 + \frac{1}{4}\left(\frac{q\boldsymbol{A}_0}{mc}\right)^2 + \frac{1}{8}\left(\frac{q\boldsymbol{A}_0}{mc}\right)^4 + \cdots\right]\hat{\boldsymbol{k}}\cdot\boldsymbol{p}, \quad (13)$$

where $\boldsymbol{A}_0 \equiv \boldsymbol{A}(t)$. Furthermore, neglecting all terms but the first one in the square brackets in Eq. (13), the reduced Hamiltonian takes the same form as the one used in Refs. [6,21,22,24]. Adopting instead the LWA directly to the minimal-coupling Hamiltonian (2), the usual dipole approximation Hamiltonian is retrieved,

$$H_0^{\mathrm{LWA}} = \frac{p^2}{2m} + V - \frac{q}{m}\boldsymbol{A}_0\cdot\boldsymbol{p}, \quad (14)$$

where the purely time-dependent and unimportant $\frac{q^2}{2m}\boldsymbol{A}_0^2(t)$ term effectively cancels out by a phase (gauge) transformation [10].

Comparing the two versions of the LWA interaction Hamiltonians (13) and (14), the first one contains an extra operator term of the form $\hat{\boldsymbol{k}}\cdot\boldsymbol{p}$, i.e., a velocity-gauge-like operator acting along the propagation direction of the laser. This term plays the role of the radiation pressure originating from the combined effect of the electric and magnetic fields, as represented by the Poynting vector in Maxwell's classical theory of electromagnetism. Radiation pressure is obviously not supported by the usual dipole approximation Hamiltonian (14) where any field-driven motion along the direction of propagation is neglected altogether. At first sight, the appearance of the additional operator in Eq. (13) that has no counterpart in the ordinary dipole approximation Hamiltonian (14) might seem inconsistent, given that both Hamiltonians have been derived from the very same assumption, namely, that the vector potential is taken to be purely time dependent.

Then the natural question arises: How is it possible that the very same assumption on the laser field, i.e., the LWA, can have such a different impact? The answer is that it is a question of choice of gauge and the fact that the validity range of an imposed approximation generally depends on this choice. The Hamiltonians (2) and (11) are both of general validity but they correspond to two different points of view (gauges) from which the time-dependent Schrödinger equation may be represented. As such, in a complete treatment they would always yield identical observables, i.e., the results are gauge invariant. However, once an approximation has been adopted, e.g., the LWA on the field, the resulting forms (13) and (14) are no longer equivalent and they would generally yield diverging results. We would here like to note the similarity to the so-called gauge problem that has been identified in connection with the strong-field approximation (SFA) for describing the ionization and high-order harmonic generation dynamics of atoms and molecules in intense laser fields [25]. It is well known that the length- and velocity-gauge formulations of the SFA are likely to yield quite different results, often differing by several orders of magnitude. This

problem is merely an example of the more general principle that states that within an approximate framework the prediction of physical observables become gauge dependent [3]. The radiation pressure is a phenomenon that depends on the laser intensity *per se*, rather than its frequency, and therefore it *should* be considered independently of the extension of the system and long-wavelength assumptions. Therefore, we suggest that the LWA Hamiltonian in the propagation gauge form (13) is of more general validity than its velocity-gauge counterpart (14) and this in spite of the fact that they are indeed both derived by imposing the very same restriction on the vector potential.

We now turn to an application of the propagation gauge (PG) to a relevant physical problem and show that numerical calculations yield identical, yet faster converging results compared to those obtained within the already well established velocity-gauge (VG) description. In order to compare the PG and the VG on an equal footing, we resort to a first-order spatial expansion of the vector potential in the latter case (see, e.g., Ref. [10]); this in order to include a beyond-dipole interaction term describing the radiation pressure, which is already inherently accounted for in the PG picture. For this demonstration, we retain only the first leading-order term from the LWA Hamiltonian (13) such that the VG and PG Hamiltonians become equivalent (within an exact treatment) and take the forms

$$H_{1\mathrm{st}}^{\mathrm{VG}} = \frac{p^2}{2m_e} + V + \frac{e}{m_e}\boldsymbol{A}_0 \cdot \boldsymbol{p} - \frac{e^2}{m_e c}\left(\boldsymbol{A}_0 \cdot \frac{d\boldsymbol{A}_0}{dt}\right)\hat{\boldsymbol{k}} \cdot \boldsymbol{r}, \tag{15}$$

$$H_{1\mathrm{st}}^{\mathrm{PG}} = \frac{p^2}{2m_e} + V + \frac{e}{m_e}\boldsymbol{A}_0 \cdot \boldsymbol{p} + \frac{c}{2}\left(\frac{e\boldsymbol{A}_0}{m_e c}\right)^2 \hat{\boldsymbol{k}} \cdot \boldsymbol{p}, \tag{16}$$

where m_e and $-e$ are the electron mass and charge, respectively, and the system under study is chosen to be atomic hydrogen. The time-dependent Schrödinger equation (TDSE) is discretized by expanding the three-dimensional wave function on a product basis of B-spline functions B_k and spherical harmonics Y_{lm},

$$\Psi(\boldsymbol{r},t) = \sum_{k=1}^{k_{\max}} \sum_{l=0}^{l_{\max}} \sum_{m=-l}^{l} c_{klm}(t) \frac{B_k(|\boldsymbol{r}|)}{|\boldsymbol{r}|} Y_{lm}(\hat{\boldsymbol{r}}), \tag{17}$$

where a total number of $k_{\max}$ B splines is distributed on some radial domain $|\boldsymbol{r}| \in [0, R_{\max}]$. We would here like to point out that the resulting Hamiltonian matrix systems are of the same size and complexity when discretization of the Hamiltonians (15) and (16) is performed. Any difference when it comes to computational performance relates to different convergence criteria with respect to the basis sizes $k_{\max}$ and $l_{\max}$ and possibly the propagation time step. The matrix system of ordinary differential equations is propagated in time with the same scheme as described in Refs. [10,11]. For the laser field, we choose a linearly polarized XUV pulse of sine-square shape with a central photon energy $\hbar\omega = 3.5$ a.u. At maximum intensity, the peak electric-field strength reaches $E_0 = 45$ a.u. and the pulse lasts for a total of 40 optical cycles. Such laser parameters have recently been found to induce beyond-dipole effects associated with the radiation pressure [10] and *that*

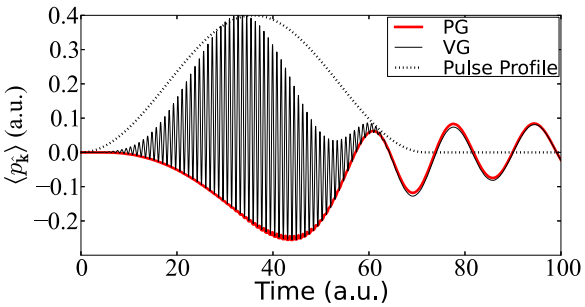

FIG. 1. Momentum expectation value of the electron along the laser propagation direction vs time during the laser-matter interaction between a hydrogen atom and an XUV laser pulse, as obtained with the velocity-gauge (thin black line) and propagation gauge (thick red line) versions of the interaction Hamiltonian, i.e., Eqs. (15) and (16), respectively. The electric-field strength $E_0 = 45$ a.u. at peak intensity, the photon energy $\hbar\omega = 3.5$ a.u., and the pulse is modeled by a sine-square envelope profile (dashed black line) lasting for a total of 40 optical cycles, i.e., 71.8 a.u.

without introducing significant corrections due to spatial variations in the electric and magnetic fields themselves, thus the long-wavelength approximation should remain valid.

When comparing the propagation-gauge frame [Eq. (16)] with the usual (first-order expansion) velocity-gauge form of the nondipole interaction [Eq. (15)], very different dynamics can be observed if we track the momentum expectation value along the direction of propagation during the full extent of the laser-atom interaction. The results are shown in Fig. 1. It is shown that the expectation value $\langle p_{\hat{\boldsymbol{k}}} \rangle$ oscillates rapidly in the VG picture (with an angular frequency 2ω), which stands in the direct opposition to the smooth variation with time observed in the PG case. The origin of this fundamentally different behavior relates directly to the previous discussion and the fact that the Hamiltonian (16) represents a different point of view, as compared to Eq. (15). The much smoother temporal behavior of the momentum in the PG case simply calls for a less time consuming propagation scheme with less restrictive convergence criteria.

In order to test the performance of the two discretized Hamiltonians when it comes to the prediction of physical observables, we present the kinetic energy distribution of the ionized electron in Fig. 2. The top panel shows a comparison of the photoelectron spectra as obtained when applying the LWA directly to the velocity-gauge Hamiltonian, thus arriving at the dipole approximation [Eq. (14)], and when applying the LWA to the propagation-gauge Hamiltonian, i.e., Eq. (16). Clear differences between the dipole and beyond-dipole results are revealed in the spectra, in that the multiphoton resonances in the dipole situation exhibit a pronounced oscillatory structure not seen in the corresponding beyond-dipole case. These fringes are a result of the interference between the wave packets being ejected during the laser ramp-on and ramp-off, respectively. As it turns out, the electron's displacement along the laser propagation direction during the laser-matter interaction has a great impact on the underlying interference phenomenon with the result that the interference pattern is suppressed. As such, an exceedingly accurate description of the quantum mechanical radiation pressure and the associated

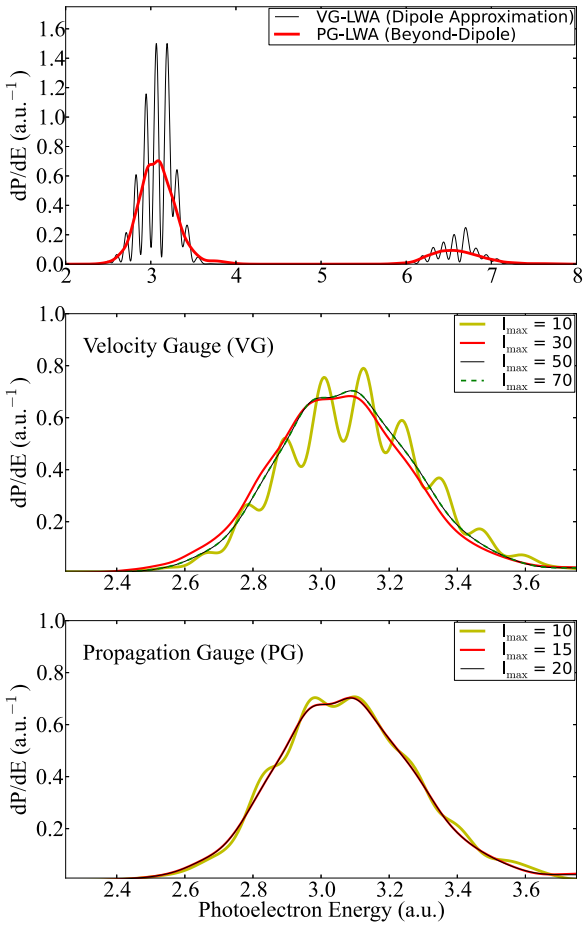

FIG. 2. Probability distribution with respect to the kinetic energy of the ionized electron following a laser-assisted ionization process with the laser field described in Fig. 1. The top panel shows a comparison between the spectra as obtained within the LWA applied directly to the velocity gauge Hamiltonian [Eq. (14)], i.e., the dipole approximation result, and the corresponding result obtained when imposing the LWA to the PG Hamiltonian [Eq. (16)]. The results are obtained with $l_{max} = 30$. In the middle panel, the beyond-dipole velocity-gauge results [Eq. (15)] are shown for four different (angular) truncation sizes, i.e., $l_{max} = 10$ (thick yellow line), $l_{max} = 30$ (red line), $l_{max} = 50$ (thin black line), and $l_{max} = 70$ (dashed green line). In the bottom panel, the corresponding results obtained with the propagation gauge form of the laser-matter interaction [Eq. (16)] are displayed for the basis sizes $l_{max} = 10$ (thick yellow line), $l_{max} = 15$ (red line), and $l_{max} = 20$ (thin black line).

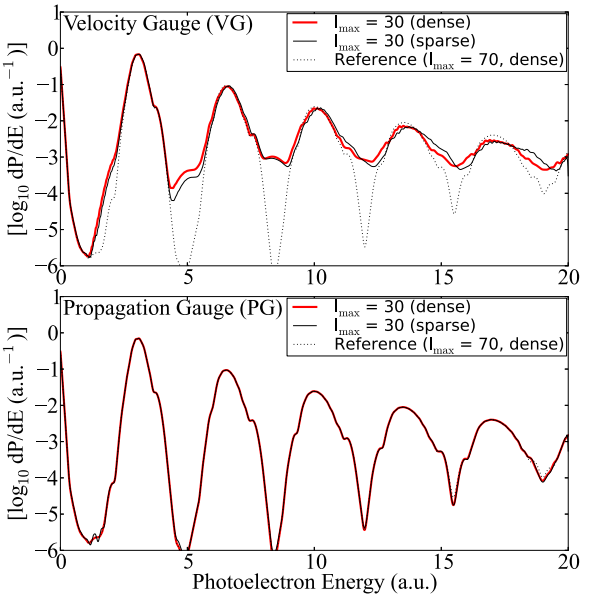

FIG. 3. Energy distributions for the same case as in Fig. 2, but for a fixed value of $l_{max} = 30$. Two different radial simulation domains have been used, one sparse grid with $k_{max} = 1200$ and $R_{max} = 450$ a.u. (thin black line) and one dense grid with $k_{max} = 3200$ and $R_{max} = 800$ a.u. (thick red line). The top and bottom panels depict the results as obtained with the velocity gauge and the propagation gauge, respectively. The dashed curve shows the fully converged calculations with the richest set of basis parameters, i.e., $R_{max} = 800$ a.u., $k_{max} = 3200$, and $l_{max} = 70$ a.u.

electron displacement is prerequisite for obtaining fully converged energy distributions, which in turn serves as an ideal benchmark quantity for testing the numerical performance of the PG with respect to the VG.

Focusing for the moment on the kinetic energy region $E_k \in [2.25\text{ a.u.}, 3.75\text{ a.u}]$, i.e., the net absorption of one photon from the laser field, the results obtained by solving the TDSE in the usual velocity gauge and in the propagation gauge are shown in the middle and bottom panels in Fig. 2. The different curves pertain to different upper thresholds for the (angular) basis expansion (17), where $l_{max} = 10$–20 was used in the propagation gauge (bottom panel) and $l_{max} = 10$–70 was applied in the velocity-gauge (middle panel). From the figure it is clear that the structure in the energy distribution around the one-photon absorption resonance has very different convergence properties in the two respective gauges. Whereas $l_{max} = 15$ is sufficient when using the nondipole Hamiltonian (16), the standard first-order expansion of the VG Hamiltonian, i.e., Eq. (15), requires $l_{max} = 50$ in order to reach the same level of accuracy. This is quite a remarkable difference considering that all possible values of the magnetic quantum numbers m are included for each value of l. If we for simplicity assume a straightforward software implementation where all nonzero matrix elements are stored in the computer memory, we are looking at more than a tenfold increase in basis size, overall memory consumption, and execution run-time for the problem at hand.

In Fig. 3 the photoelectron distribution is shown for two different radial representations. Here the low-accuracy representation (sparse grid) corresponds to a radial box size of $R_{max} = 450$ a.u. and $k_{max} = 1200$ equidistantly distributed B splines, whereas the high-accuracy basis (dense grid) comprises $k_{max} = 3200$ B splines distributed over $R_{max} = 800$ a.u. The top and bottom panels show the logarithm of the energy distributions for the five first multiphoton resonances obtained with the Hamiltonians (15) and (16), respectively. The dashed background curve depicts in both panels the fully converged energy distribution (the reference result) as obtained with the dense grid radial representation and $l_{max} = 70$. Again, the PG variant of the interaction Hamiltonian achieves a higher performance at a considerably cheaper computational cost than its VG counterpart. For the propagation gauge case, both

grids reproduce the reference energy distribution, whereas for the case of the velocity gauge the two different radial representations both predict deviating photoelectron spectra with respect to the reference, especially for the higher kinetic energies. The reason for the faster convergence of the PG with respect to the radial parameters can be understood when comparing the $\hat{\boldsymbol{k}} \cdot \boldsymbol{r}$ and the $\hat{\boldsymbol{k}} \cdot \boldsymbol{p}$ forms of the associated nondipole interaction terms in Eqs. (15) and (16), respectively. The first one, depending on the spatial coordinate, will promote a large potential value for $|\boldsymbol{r}| \gg 0$, indicating that the ionized electron, possibly situated far from the nucleus, will be harshly accelerated and decelerated in the field. As such, at intermediate times during the laser pulse, very large kinetic energies are attained temporarily by the ionized electron in the VG case and high-energy continuum states are populated. These states tend to be less accurately, or even wrongly, represented in a sparse radial discretization, thus influencing the entire intermediate dynamics and preventing the correct final state in the end.

In conclusion, we have from the velocity-gauge minimal-coupling Hamiltonian derived an equivalent form of the light-matter interaction. Motivated by the current development of superintense and high-frequency light sources used to study atomic and molecular systems, we present a formalism tailored for describing quantum-mechanical systems interacting with fields beyond the dipole approximation. From this point of view, the motion along the laser propagation direction due to the combined contribution of the electric and magnetic (nondipole) fields is explicitly accounted for by a velocity-gauge-like operator. As such, we suggest that the present formalism can be viewed as a generalized velocity gauge where the kinematics in the laser propagation direction is inherently included on an equal footing with the motion in the polarization direction and we call the resulting gauge the propagation gauge. The propagation-gauge Hamiltonian has a relatively simple form and one of its highly advantageous properties is that the long-wavelength approximation can be imposed directly without making any assumptions about the importance of magnetic-field effects, which stands in direct contrast to the original velocity-gauge formulation. Furthermore, by solving the time-dependent Schrödinger equation for atomic hydrogen exposed to a superintense XUV laser pulse of femtosecond duration, we demonstrate that the propagation-gauge form of the interaction Hamiltonian is highly tractable from a numerical perspective due to its faster convergence properties, as compared to the original velocity-gauge form of the Hamiltonian.

This work was supported by the Bergen Research Foundation (Norway) and the Norwegian Metacenter for Computational Science (Notur). All calculations were performed on the Cray XE6 (Hexagon) supercomputer installation at Parallab, University of Bergen.

[1] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Photons and Atoms. Introduction to Quantum Electrodynamics* (Wiley, New York, 1997).
[2] F. Mandl and G. Shaw, *Quantum Field Theory* (Wiley, New York, 2010).
[3] A. D. Bandrauk, F. Fillion-Gourdeau, and E. Lorin, J. Phys. B **46**, 153001 (2013).
[4] A. Bugacov, M. Pont, and R. Shakeshaft, Phys. Rev. A **48**, R4027 (1993).
[5] N. J. Kylstra, R. A. Worthington, A. Patel, P. L. Knight, J. R. Vázquez de Aldana, and L. Roso, Phys. Rev. Lett. **85**, 1835 (2000).
[6] 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).
[7] M. Førre, S. Selstø, J. P. Hansen, and L. B. Madsen, Phys. Rev. Lett. **95**, 043601 (2005).
[8] M. Førre, J. P. Hansen, L. Kocbach, S. Selstø, and L. B. Madsen, Phys. Rev. Lett. **97**, 043601 (2006).
[9] K. J. Meharg, J. S. Parker, and K. T. Taylor, J. Phys. B **38**, 237 (2005).
[10] M. Førre and A. S. Simonsen, Phys. Rev. A **90**, 053411 (2014).
[11] A. S. Simonsen and M. Førre, Phys. Rev. A **92**, 013405 (2015).
[12] Y. I. Salamin, S. X. Hu, K. Z. Hatsagortsyan, and C. H. Keitel, Phys. Rep. **427**, 41 (2006).
[13] S. Selstø, E. Lindroth, and J. Bengtsson, Phys. Rev. A **79**, 043418 (2009).
[14] A. Di Piazza, C. Müller, K. Z. Hatsagortsyan, and C. H. Keitel, Rev. Mod. Phys. **84**, 1177 (2012).
[15] H. Bachau, M. Dondera, and V. Florescu, Phys. Rev. Lett. **112**, 073001 (2014).
[16] C. I. Moore, A. Ting, S. J. McNaught, J. Qiu, H. R. Burris, and P. Sprangle, Phys. Rev. Lett. **82**, 1688 (1999).
[17] 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).
[18] M. Førre, S. Selstø, J. P. Hansen, T. K. Kjeldsen, and L. B. Madsen, Phys. Rev. A **76**, 033415 (2007).
[19] M. Førre, Phys. Rev. A **74**, 065401 (2006).
[20] D. Dimitrovski, M. Førre, and L. B. Madsen, Phys. Rev. A **80**, 053412 (2009).
[21] M. Y. Emelin and M. Y. Ryabikin, Phys. Rev. A **89**, 013418 (2014).
[22] M. Y. Ryabikin and A. M. Sergeev, Opt. Express **7**, 417 (2000).
[23] J. J. Sakurai, *Modern Quantum Mechanics* (Academic-Wesley, Reading, 1994).
[24] A. V. Kim, M. Y. Ryabikin, and A. M. Sergeev, Phys. Usp. **42**, 54 (1999).
[25] Y. V. Vanne and A. Saenz, Phys. Rev. A **79**, 023421 (2009).