

# A Schrödinger formulation of the nondipole light-matter interaction consistent with relativity

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An alternative and powerful Schrödinger-like equation for describing beyond dipole laser-matter interactions is derived. It is shown that this particular formulation is numerically very efficient with respect to computational effort and convergence rate of the solutions. Furthermore, and more importantly, its nonrelativistic form turns out to be more compatible with relativity than what seems to be the case with the more common formulations of the nonrelativistic light-matter interaction. Moreover, an extension of this interaction form into the relativistic region preserves, to a large extent, the numerical efficiency.

## I. INTRODUCTION

The question of how the magnetic component of some laser field actually alters the strong-field ionization of atoms and molecules is becoming increasingly important and goes hand in hand with the ongoing development of new extreme light sources. Recent experimental activity in the field has already provided valuable new insight into this emerging area of research [1–9]. The theoretical modeling of the laser-matter interaction in the intense field regime is particularly challenging, primarily due to the fact that the celebrated dipole approximation is generally no longer applicable [10–15]. Furthermore, in the limit of very strong fields the validity of the usual nonrelativistic approximation ultimately breaks down and a relativistic treatment of the laser-matter interaction becomes necessary [16–23].

In the present work we outline a coherent and transparent theoretical model for handling beyond dipole (nondipole) and relativistic corrections effects on an equal footing. This derivation is based on the time-dependent Schrödinger equation and the energy-momentum relation. The resulting interaction Hamiltonian turns out to be very favorable, not only from a computational point of view, but also from the point of view of better understanding the transition between the relativistic and nonrelativistic regimes. The work can be summarized by the formulas (21) and (25) derived in Sec. II, for the nonrelativistic and relativistic interactions, respectively. The usefulness of the new formulation is explicitly demonstrated by studying the ionization dynamics of atomic hydrogen by some short and intense x-ray laser pulse in a regime where the ordinary dipole approximation is inaccurate [24]. It is shown that the here proposed scheme completely outmatches the more standard formulations of the light-matter interaction when it comes to comparing the rate of convergence of the calculations with respect to the number of angular momenta included in the calculations.

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

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## II. THEORY

In the standard nonrelativistic approach, the wave function  $\psi(\mathbf{r}, t)$  of a particle of mass  $m$  and charge  $q$ , evolving in some laser field  $\mathbf{A}$  and (Coulomb) potential  $V$ , is governed by the time-dependent Schrödinger equation (TDSE),

$$i\hbar \frac{\partial}{\partial t} \psi = H \psi, \quad (1)$$

with the minimal coupling formulation of the light-matter interaction Hamiltonian

$$H = \frac{p^2}{2m} + V - \frac{q}{m} \mathbf{A} \cdot \mathbf{p} + \frac{q^2}{2m} A^2. \quad (2)$$

Here the Coulomb gauge restriction  $\nabla \cdot \mathbf{A} = 0$  on the field has been imposed. The vector potential  $\mathbf{A}(\mathbf{r}, t)$  generally depends on both space and time coordinates and satisfies separately the wave equation. From a purely computational point of view, keeping the spatial dependence in the vector potential most often result in an intractable numerical problem. Therefore, and in order to simplify the calculations significantly, the so-called dipole approximation is most often imposed. In this approximation the spatial dependence of the field is not considered, the magnetic field component is neglected, and the vector potential  $\mathbf{A}$  is assumed to depend on time only. One consequence of the approximation is that the last (diamagnetic) term in the Hamiltonian (2) becomes an unimportant time-dependent factor that can be left out. The dipole approximation is usually valid in the limit where the extension of the quantum system in question is much smaller than the wavelength of the incoming light and provided the laser intensity is not so high that the magnetic field component of the field must be included.

In an alternative and less known route, the system Hamiltonian can instead be written as [19, 23, 25, 26]

$$H = \frac{p^2}{2m} + V - \frac{q}{m} \mathbf{A} \cdot \mathbf{p} + \frac{q^2}{4m^2 c} \left\{ A^2, \hat{\mathbf{k}} \cdot \mathbf{p} \right\}, \quad (3)$$

where the unit vector  $\hat{\mathbf{k}}$  indicates the laser propagation direction,  $c$  is the speed of light, and curly brackets denote the anticommutator defined by  $\{a, b\} = ab + ba$ . The anticommutator originates from the fact that the two operators  $A^2$  and  $\hat{\mathbf{k}} \cdot \mathbf{p}$  do not generally commute. The interaction Hamiltonian (3)

may also be obtained as the nonrelativistic limit of the Dirac equation – with the addition of the interaction between the particle’s spin and the external magnetic field [27].

If we disregard terms beyond second order in  $1/c$ , the two formulations (2) and (3) are equivalent and can be used interchangeably, i.e., they would yield the same results in any exact treatment, provided the laser-matter interaction does not introduce relativistic effects. Nonetheless, the formulation (3), also called the *propagation gauge* formulation [25], has proven to be numerically advantageous in handling laser-matter interactions in the intense field limit where the dipole approximation breaks down [25, 27]. The wave function  $\psi'$  in the propagation gauge is related to the original wave function  $\psi$  in Eq. (1) by the gauge transformation [25]

$$\psi' = e^{iX} \psi, \quad (4)$$

with

$$X(\eta) = X(\omega t - \mathbf{k} \cdot \mathbf{r}) = \frac{q^2}{2m\hbar\omega} \int_{-\infty}^{\eta} A^2(\eta') d\eta', \quad (5)$$

where  $\omega$  is the central angular frequency of the laser field and  $\mathbf{k} = \omega/c \hat{\mathbf{k}}$  is the wave vector.

In order to introduce the magnetic field in the standard formulation (2) of the light-matter interaction, it is common to write out the vector potential in terms of a Maclaurin series expansion, i.e., writing

$$\mathbf{A}(\mathbf{r}, t) = \mathbf{A}_0(t) + \frac{1}{c} \hat{\mathbf{k}} \cdot \mathbf{r} \mathbf{E}_0(t) + \dots, \quad (6)$$

where  $\mathbf{A}_0(t)$  now refers to the dipole field and  $\mathbf{E}_0 = -\frac{\partial}{\partial t} \mathbf{A}_0$  represents the corresponding homogeneous electric field. In this approximation the magnetic field component is given by

$$\mathbf{B}(\mathbf{r}, t) = \nabla \times \mathbf{A} = \frac{1}{c} \hat{\mathbf{k}} \times \mathbf{E}_0 + \dots \quad (7)$$

Applying the expanded potential (6), the minimal coupling Hamiltonian (2) is now cast into

$$H = \frac{p^2}{2m} + V - \frac{q}{m} \mathbf{A}_0 \cdot \mathbf{p} + \frac{q^2}{2m} A_0^2 - \frac{q}{mc} \mathbf{E}_0 \cdot (\mathbf{p} - q\mathbf{A}_0) \hat{\mathbf{k}} \cdot \mathbf{r} + \dots \quad (8)$$

The performance of the Hamiltonian (8) in actual numerical calculations will be demonstrated later. We will not go back and elaborate on the corresponding propagation gauge formulation (3). More specifically, we shall demonstrate that including spatial dependence in the  $\mathbf{A} \cdot \mathbf{p}$  term in Eqs. (2) or (3) may be problematic, both from a conceptual and a computational point of view.

For pedagogical reasons, we begin with showing how Eq. (3) can be derived. To this end, we take as starting point the relativistic minimal coupling Hamiltonian for a spinless particle of charge  $q$  and mass  $m$  confined in a (Coulomb) po-

tential  $q\phi = V$  and interacting with some laser field  $\mathbf{A}$ ,

$$H = \sqrt{m^2 c^4 + (\mathbf{p} - q\mathbf{A})^2 c^2} - mc^2 + V = \frac{(\mathbf{p} - q\mathbf{A})^2}{2m} - \frac{(\mathbf{p} - q\mathbf{A})^4}{8m^3 c^2} + \dots + V. \quad (9)$$

Next, we impose the gauge transformation

$$\mathbf{A} \rightarrow \mathbf{A}' = \mathbf{A} + \nabla \xi \quad (10)$$

$$\phi \rightarrow \phi' = \phi - \partial_t \xi \quad (11)$$

on the potentials, with

$$\xi(\eta) = \xi(\omega t - \mathbf{k} \cdot \mathbf{r}) = \frac{q}{2m\omega} \int_{-\infty}^{\eta} A^2(\eta') d\eta'. \quad (12)$$

Note at this point that this gauge transformation on the potentials is fully equivalent to the unitary transformation imposed on the system wave function in Eq. (4). The Hamiltonian (9) is then cast into its propagation gauge form,

$$H = \frac{(\mathbf{p} - q\mathbf{A} + \frac{q^2}{2mc} A^2 \hat{\mathbf{k}})^2}{2m} - \frac{(\mathbf{p} - q\mathbf{A} + \frac{q^2}{2mc} A^2 \hat{\mathbf{k}})^4}{8m^3 c^2} + \dots + V - \frac{q^2}{2m} A^2 = \frac{p^2}{2m} + V - \frac{q}{m} \mathbf{A} \cdot \mathbf{p} + \frac{q^2}{4m^2 c} \{A^2, \hat{\mathbf{k}} \cdot \mathbf{p}\} - \frac{p^4}{8m^3 c^2} + \frac{q}{4m^3 c^2} \{\mathbf{A} \cdot \mathbf{p}, p^2\} - \frac{q^2}{8m^3 c^2} \{A^2, p^2\} + \frac{q^3}{2m^3 c^2} A^2 \mathbf{A} \cdot \mathbf{p} - \frac{q^2}{2m^3 c^2} (\mathbf{A} \cdot \mathbf{p})^2 + \dots, \quad (13)$$

where in the last step only terms of order  $(1/c)^2$  and lower have been written out explicitly. The Hamiltonian (13) in the present form also accounts for relativistic kinematic effects and was recently derived in [19, 23]. Going to the nonrelativistic limit, i.e., omitting terms of order  $(1/c)^2$  and higher, the nonrelativistic propagation gauge Hamiltonian (3) is finally retrieved.

Now, neglecting the spatial dependence of the vector potential, i.e., letting  $\mathbf{A}(\mathbf{r}, t) \rightarrow \mathbf{A}_0(t)$  in Eq. (3), the Hamiltonian is converted into

$$H = \frac{p^2}{2m} + V - \frac{q}{m} \mathbf{A}_0(t) \cdot \mathbf{p} + \frac{q^2}{2m^2 c} A_0^2(t) \hat{\mathbf{k}} \cdot \mathbf{p}, \quad (14)$$

a form which is reminiscent of the interaction used in [28–31]. Already at this point one may notice one clear advantage of the propagation gauge formulation of the light-matter interaction in that the Hamiltonian (14) actually accounts for magnetic field effects through the last term in the interaction [25], albeit no spatial dependence of the field has been retained. This stands in stark contrast to the standard minimal coupling formulation (2) for which the vector potential must be explicitly space-dependent in order to account for the magnetic field, cf., Eqs. (6–8). The Hamiltonian (14) has yet another advantage in that in the limit of vanishing potential

$V$ , the momentum  $\mathbf{p}$  becomes a constant of the motion, i.e., a conserved quantity. This is the main reason why the propagation gauge formulation in general tends to be more favorable from a purely numerical point of view, in particular in the limit of very strong perturbations.

As mentioned previously, including spatial dependence in the vector potential in the  $\mathbf{A} \cdot \mathbf{p}$  operator term in the interaction Hamiltonian (2) or (3) may cause some additional trouble. Therefore, we are now about to introduce an additional unitary transformation to the light-matter formulation that will prove to be extremely useful in investigating intense field effects beyond the dipole approximation. To this end, we start out by writing out the Hamiltonian (3) on the following trivially extended form:

$$H = \frac{p^2}{2m} + V(\mathbf{r}) - \frac{q}{m} \mathbf{A}_0 \cdot \mathbf{p} - \frac{q}{m} (\mathbf{A} - \mathbf{A}_0) \cdot \mathbf{p} + \frac{q^2}{4m^2c} \left\{ A^2, \hat{\mathbf{k}} \cdot \mathbf{p} \right\}, \quad (15)$$

where  $\mathbf{A}_0 = \mathbf{A}_0(t)$  and  $\mathbf{A} = \mathbf{A}(\mathbf{r}, t)$  as defined by Eq. (6). Next, the goal is to get rid of the  $(\mathbf{A} - \mathbf{A}_0) \cdot \mathbf{p}$  term from the formulation. This may be achieved by introducing the unitary transformation

$$\psi'' = U \psi' \quad (16)$$

to the propagation gauge wave function  $\psi'$  in Eq. (4), with

$$U = \exp \left[ \frac{i}{\hbar} \boldsymbol{\alpha}(\mathbf{r}, t) \cdot \mathbf{p} \right] \quad (17)$$

$$\boldsymbol{\alpha} = -\frac{q}{m} \int_{-\infty}^t (\mathbf{A} - \mathbf{A}_0) dt'. \quad (18)$$

The transformation will give rise to some new interaction terms which can be calculated using the Baker-Campbell-Hausdorff formula:

$$e^{ia} b e^{-ia} = b + \frac{i}{1!} [a, b] + \frac{i^2}{2!} [a, [a, b]] + \frac{i^3}{3!} [a, [a, [a, b]]] + \dots \quad (19)$$

The Hamiltonian from this new point of view becomes

$$\begin{aligned} H' &= U H U^\dagger + i\hbar \dot{U} U^\dagger \\ &= \frac{p^2}{2m} + V(\mathbf{r} + \boldsymbol{\alpha}) - \frac{q}{m} \mathbf{A}_0 \cdot \mathbf{p} + \frac{q^2}{4m^2c} \left\{ A^2, \hat{\mathbf{k}} \cdot \mathbf{p} \right\} \\ &\quad - \frac{q}{2m^2c} \left\{ \hat{\mathbf{k}} \cdot \mathbf{p}, \mathbf{A} \cdot \mathbf{p} \right\} - \frac{q^3}{2m^3c^2} A^2 \mathbf{A} \cdot \mathbf{p} + \frac{q^2}{2m^3c^2} (\mathbf{A} \cdot \mathbf{p})^2. \end{aligned} \quad (20)$$

The transformed Hamiltonian (20) contains in total three new terms that originate from the  $(\mathbf{A} - \mathbf{A}_0) \cdot \mathbf{p}$  operator in Eq. (15). In addition, the potential has been shifted, i.e.,  $V(\mathbf{r}) \rightarrow V(\mathbf{r} + \boldsymbol{\alpha})$ .

At this point, the attentive reader may already have made the important observation that the last two terms in Eq. (20) are also present in the general relativistic Hamiltonian (13) – but with opposite signs. This means that when introducing the unitary transformation (16-18) to the relativistically extended formulation (13), all such terms happen to cancel

exactly against each other, and therefore none of them should appear separately in any consistent model of the light-matter interaction, neither in the relativistic nor in the nonrelativistic limit. To this end, the formally correct (nonrelativistic) Hamiltonian takes the simpler form

$$\begin{aligned} H &= \frac{p^2}{2m} + V(\mathbf{r} + \boldsymbol{\alpha}) - \frac{q}{m} \mathbf{A}_0 \cdot \mathbf{p} + \frac{q^2}{4m^2c} \left\{ A^2, \hat{\mathbf{k}} \cdot \mathbf{p} \right\} \\ &\quad - \frac{q}{2m^2c} \left\{ \hat{\mathbf{k}} \cdot \mathbf{p}, \mathbf{A} \cdot \mathbf{p} \right\}. \end{aligned} \quad (21)$$

We have here finally arrived at a relatively compact expression for the light-matter interaction. In this formulation the pure dipole interaction is represented by a separate term. In addition, the Hamiltonian contains three terms that are attributed to radiation of the electromagnetic field beyond the dipole approximation, i.e., the usual propagation gauge term proportional  $A^2$ , a new term proportional to  $\mathbf{A}$  arising from the original  $\mathbf{A} \cdot \mathbf{p}$  operator in Eq. (3), and finally, as a side effect of the transformation, the shifted potential  $V(\mathbf{r} + \boldsymbol{\alpha})$ . This modified potential may be expanded in ascending powers of  $1/c$ , i.e.,

$$V(\mathbf{r} + \boldsymbol{\alpha}) = V(\mathbf{r}) + \frac{q}{mc} \hat{\mathbf{k}} \cdot \mathbf{r} \nabla V(\mathbf{r}) \cdot \mathbf{A}_0 + \dots \quad (22)$$

We are here primarily occupied with  $1/c$  corrections to the light-matter interaction, and as such we keep only the leading order correction to the Coulomb potential as well as substitute  $\mathbf{A}$  with  $\mathbf{A}_0$  in the remaining two beyond dipole interaction terms. Then, the Hamiltonians (20) and (21) are cast into

$$\begin{aligned} H &= \frac{p^2}{2m} + V - \frac{q}{m} \left[ 1 + \frac{\hat{\mathbf{k}} \cdot \mathbf{p}}{mc} \right] \mathbf{A}_0(t) \cdot \mathbf{p} + \frac{q^2}{2m^2c} A_0^2(t) \hat{\mathbf{k}} \cdot \mathbf{p} \\ &\quad + \frac{q}{mc} \hat{\mathbf{k}} \cdot \mathbf{r} \nabla V \cdot \mathbf{A}_0(t) - \frac{q^3}{2m^3c^2} A_0^2 \mathbf{A} \cdot \mathbf{p} + \frac{q^2}{2m^3c^2} (\mathbf{A}_0 \cdot \mathbf{p})^2, \end{aligned} \quad (23)$$

and

$$\begin{aligned} H &= \frac{p^2}{2m} + V - \frac{q}{m} \left[ 1 + \frac{\hat{\mathbf{k}} \cdot \mathbf{p}}{mc} \right] \mathbf{A}_0(t) \cdot \mathbf{p} + \frac{q^2}{2m^2c} A_0^2(t) \hat{\mathbf{k}} \cdot \mathbf{p} \\ &\quad + \frac{q}{mc} \hat{\mathbf{k}} \cdot \mathbf{r} \nabla V \cdot \mathbf{A}_0(t), \end{aligned} \quad (24)$$

respectively, and where  $V = V(\mathbf{r})$  now refers to the unshifted potential. Note that the anticommutation rules in Eqs. (20) and (21) become unnecessary as  $\mathbf{A} \rightarrow \mathbf{A}_0$  since the operators now commute, and they are therefore omitted in Eqs. (23) and (24).

The Hamiltonian (24), which contains all beyond dipole interaction terms up to and including order  $1/c$  corrections, constitutes the main result of the present work. The alternative formulation (23) is reminiscent of the light-matter interaction derived recently by Brennecke and Lein [32] and used in explaining experimental data on magnetic field effects in the strong-field ionization of atoms [6]. In the nonrelativistic limit, and provided relativistic corrections of order  $(1/c)^2$  and higher are unimportant to the dynamics, the two formu-

lations (23) and (24) would yield similar but not identical results. Any discrepancies could then be attributed to the two extra terms appearing in Eq. (23) – both of which are of relativistic order and happen to cancel exactly when the fully relativistic interaction (13) is considered.

Although relativistic effects are not the main concern of the present work, for completeness we also present the relativistically extended light-matter interaction correct to order  $1/c^2$ , as obtained by imposing the unitary transformation (16-18) to the general Hamiltonian (13). Comparing Eqs. (13) and (20) this is simply achieved by including all the surviving  $1/c^2$  corrections. The final result becomes

$$H = \frac{p^2}{2m} - \frac{p^4}{8m^3c^2} + V(\mathbf{r} + \boldsymbol{\alpha}) - \frac{q}{m} \mathbf{A}_0 \cdot \mathbf{p} + \frac{q^2}{4m^2c} \{A^2, \hat{\mathbf{k}} \cdot \mathbf{p}\} - \frac{q}{2m^2c} \{\hat{\mathbf{k}} \cdot \mathbf{p}, \mathbf{A} \cdot \mathbf{p}\} + \frac{q}{4m^3c^2} \{\mathbf{A} \cdot \mathbf{p}, p^2\} - \frac{q^2}{8m^3c^2} \{A^2, p^2\}, \quad (25)$$

which is valid up to but not including order  $(1/c)^3$  corrections. Notice at this point that the two extra terms appearing in Eqs. (20) and (23) are not part of this equation, merely confirming that the alternative formulations (21) and (24) comply better with the theory of relativity. Note further that only relativistic corrections to the kinetic energy have been taken into account in Eq. (25) and that the usual spin-orbit and Darwin interaction terms have not been considered. It is, however, relatively straightforward to add these into the theory if needed. The result (25) is equivalent but not identical to the results derived recently in [19, 23], the most important distinction being that the transformation (16-18) was not imposed in the previous works.

### III. RESULTS AND DISCUSSION

We have now come to the point where we explicitly want to demonstrate the capability of the alternative formulation of the light-matter interaction, i.e., the power of the nonrelativistic Hamiltonian (24) in comparison with the standard formulation (8). However, in order to compare their respective results on an equal footing, the two terms which were of relativistic order and correctly left out from the formulation in the transition from Eq. (20) to (21) had to be reintroduced, i.e., the Hamiltonian (23) is used for this particular analysis.

For simplicity, we here consider the x-ray regime and a hydrogen  $1s$  electron exposed to a 1.36 keV laser pulse which was powered on and off over a period of 15 cycles. The laser pulse is modeled in terms of the vector potential

$$\mathbf{A}_0(t) = \frac{E_0}{\omega} f(t) \sin \omega t \hat{\mathbf{u}}_p, \quad (26)$$

where  $E_0$  is the electric-field strength at peak intensity, and  $\hat{\mathbf{u}}_p$  is a unit vector pointing in the laser polarization direction. Furthermore, the function  $f(t)$  determines the temporal shape of the pulse and is here taken to be a sine-squared function,

i.e.,

$$f(t) = \begin{cases} \sin^2\left(\frac{\pi t}{T}\right), & 0 < t < T \\ 0, & \text{otherwise,} \end{cases} \quad (27)$$

where  $T$  indicates the total duration of the pulse.

The TDSE in Eq. (1) is then solved numerically using spherical coordinates and the spectral method, with the wave function  $\psi$  expanded in a sufficiently large set of both quasi-continuum (scattering) and bound eigenstates of the field-free hydrogen atom as obtained by diagonalizing its system Hamiltonian in a B-spline basis [33]. More details on the numerical implementation are given in [34]. Accurate numerical results were obtained with a finite radial grid extending to  $r = |\mathbf{r}| = 66$  a.u., and with the maximum attainable kinetic energy of the electron in the restricted basis set truncated at 610 a.u. Moreover, the number of angular momentum pairs  $(l, m)$  included in the expansion of the wave function was varied until convergence was achieved. Note that when going beyond the dipole approximation, the azimuthal symmetry of the problem is broken, which means that values of the magnetic quantum number  $m$  running from  $-l$  to  $+l$  must be taken into consideration. In our study we aimed at comparing the rate of convergence of the calculations with respect to the number of angular momenta included.

The atom is assumed exposed a laser pulse of peak intensity  $1.26 \times 10^{22}$  W/cm<sup>2</sup>, which corresponds to the value  $E_0 = 600$  a.u. in Eq. (26). Furthermore, the value of  $\omega = 50$  in atomic units. At this high laser intensity and short wavelength of the light, the dipole approximation is no longer valid and magnetic effects become important [23, 24]. Nonetheless, the intensity is not so high that relativistic effects are prominent.

We are here primarily interested in the energy distribution of the emitted photoelectron, which is calculated by means of the equation

$$\frac{dP}{dE} = \frac{1}{k} \frac{dP}{dk} = \frac{1}{k} \sum_{lm} |\langle \Phi_{klm}^C(\mathbf{r}) | \psi(\mathbf{r}, t = T) \rangle|^2, \quad (28)$$

where atomic units have been used,  $\psi(\mathbf{r}, t = T)$  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. The results are shown in Fig. 1.

The top panel of Fig. 1 shows the energy distribution of the emitted photoelectron as obtained by solving the TDSE with the Hamiltonian (23), and for three different choices for the truncation of the angular momentum quantum number  $l$  in the expansion of the wave function. Specifically, we have set  $l_{\max} = 3, 5$  and  $10$ , respectively. In total eight multiphoton peaks are depicted in the spectrum and only very small differences between the three calculated results are expressed. This finding merely reflects the fact that numerical calculations with the light-matter formulation (23) converge very fast with respect to the total number of angular momenta included in the simulations.

The middle panel of Fig. 1 provides the corresponding result as obtained solving the TDSE with the Hamiltonian (8), but now for relatively high values of the  $l$  truncation. In this



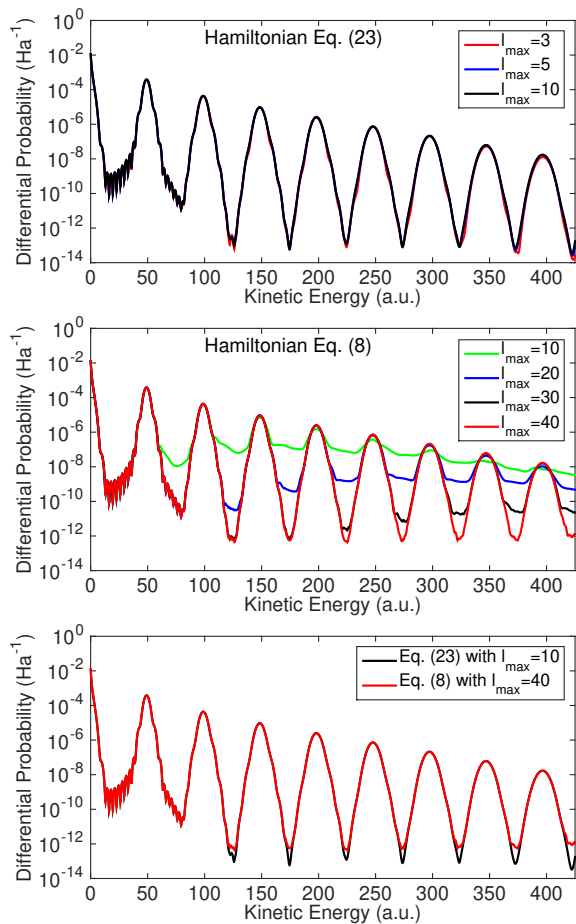


FIG. 1: Kinetic energy spectrum of the emitted photoelectron, as obtained by Eq. (28), for a 15 cycle laser pulse with  $\omega = 50$  a.u. and  $E_0 = 600$  a.u. Top panel: the beyond dipole result calculated using the Hamiltonian (23) and with  $l_{\max} = 3$  (red line), 5 (blue line) and 10 (black line), respectively. Middle panel: the same result obtained with the Hamiltonian (8) and with  $l_{\max} = 10$  (green line), 20 (blue line), 30 (black line) and 40 (red line), respectively. Bottom panel: a comparison of the results of Eqs. (8) and (23) with  $l_{\max} = 40$  (red line) and 10 (black line), respectively.

case  $l_{\max} = 10, 20, 30$  and 40, respectively. As it turns out, and in clear contrast to the upper panel result, the calculation now converges very slowly with respect to increasing number of angular momenta included in the basis expansion, and full convergence is difficult to achieve in practice. This is clearly manifested in the bottom panel of Fig. 1 showing a comparison of the result obtained by Eq. (23) and with  $l_{\max} = 10$ , and Eq. (8) with  $l_{\max} = 40$ , merely demonstrating that not even the  $l_{\max} = 40$  calculation seems to be fully converged for the higher electron energies.

Having settled the numerical superiority of the light-matter interaction formulation (23), which also apply to the formulation (24), next we present a comparison of the results obtained with the two Hamiltonians (23) and (24). The intent is to demonstrate the inaccuracy of the former. To this end, we consider the 6-photon resonance peak in Fig. 1, and the

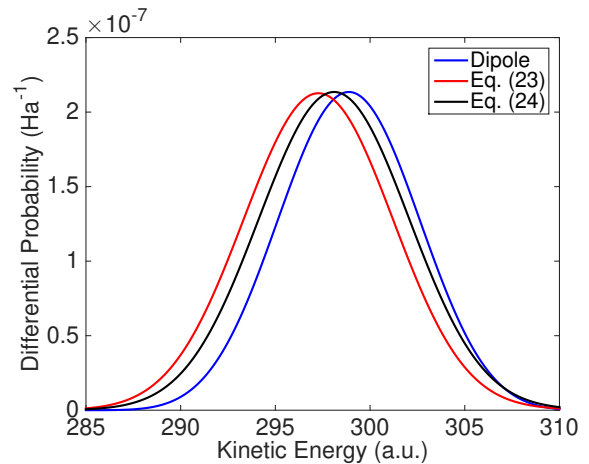


FIG. 2: A zoom of the 6-photon peak of the kinetic energy spectrum of the emitted photoelectron for a 15 cycle laser pulse with  $\omega = 50$  a.u. and  $E_0 = 600$  a.u. The value of  $l_{\max}$  was set to 10 in the calculations. Red line: the result obtained with Eq. (23) [or Eq. (8)]. Black line: the result obtained with Eq. (24). Blue line: the corresponding dipole approximation result.

result of the two respective calculations are shown in Fig. 2 together with the corresponding dipole approximation result. As it turns out, both the interactions (23) and (24) predict a shift of the resonance position to lower electron energies with respect to the dipole result, effectively giving rise to a redshift of the multiphoton ionization spectrum. Furthermore, the former predicts a larger redshift than the latter.

As a matter of fact, the observed discrepancy between the results in Fig. 2 and between the data of Eqs. (23) and (24) in particular, provides some evidence that relativistic effects may play a certain role in the excitation dynamics. Recently it was shown that such effects generally tend to produce a blueshift of the corresponding spectrum [23]. Figure 3 depicts the relativistic result obtained with Eq. (25), and where the substitution  $\mathbf{A} \rightarrow \mathbf{A}_0$  as well as the expansion (22) have been made in the system Hamiltonian. The corresponding nonrelativistic results given by Eqs. (23) and (24) are also shown for comparison. As it turns out, relativistic effects are responsible for a tiny shift of the position of the resonance to higher energies, i.e., a blueshift. Furthermore, the net ionization yield is somewhat reduced as compared to its nonrelativistic counterparts. Noticing that Eq. (23) predicts an even greater redshift than Eq. (24), cf., red and black curve in Fig. 3, it is clear that the formulation (24) is more in line with the relativistic treatment. This merely suggests that the extra terms present in Eqs. (20) and (23) are indeed superfluous, at best, and should be omitted in the general context.

#### IV. CONCLUSION

In this work, we have demonstrated that introducing spatial dependence in the vector potential in the  $\mathbf{A} \cdot \mathbf{p}$  term in the standard nonrelativistic minimal-coupling formulation (2) can be

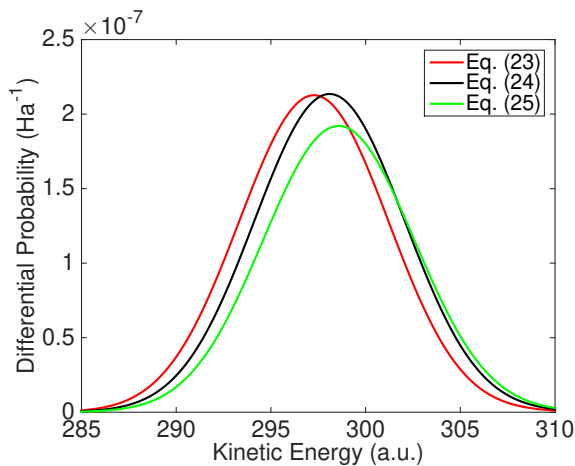


FIG. 3: Same as Fig. 2, but a comparison of the relativistic and non-relativistic results. Red line: the nonrelativistic result obtained with Eq. (23) [or Eq. (8)]. Black line: the nonrelativistic result obtained with Eq. (24). Green line: the corresponding relativistic result obtained with Eq. (25) in the limit  $\mathbf{A} \rightarrow \mathbf{A}_0$  and where the shifted potential (22) is expanded to first order in  $1/c$ .

problematic, in particular in the limit of intense laser fields, as this may lead to predictions that are in disagreement with relativity – even in field regimes where a nonrelativistic treatment is expected to be valid. With the intent to resolve this issue, we have here derived an alternative and relatively compact expression for the light-matter interaction Hamiltonian which in combination with the time dependent Schrödinger equation effectively takes into account both beyond dipole and relativistic effects. The corresponding nondipole nonrelativistic and relativistic formulations are given by Eqs. (21) and (25), respectively, in the present work. The formulation (21) is, although not relativistic in itself, formulated in concordance with relativity – with the consequence that superfluous interaction terms which otherwise would have appeared are consistently omitted. We have further demonstrated that these new interaction forms are computationally much more efficient than the more standard formulations in describing atomic excitation and ionization dynamics in superintense laser fields.

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- [1] M. Ilchen, G. Hartmann, E. V. Gryzlova, A. Achner, E. Allaria, A. Beckmann, M. Braune, J. Buck, C. Callegari, R. N. Coffee, et al., *Nat. Commun.* **9**, 4659 (2018).
- [2] 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).
- [3] A. Ludwig, J. Maurer, B. W. Mayer, C. R. Phillips, L. Gallmann, and U. Keller, *Phys. Rev. Lett.* **113**, 243001 (2014).
- [4] H. Zimmermann, S. Meise, A. Khujakulov, A. Magaña, A. Saenz, and U. Eichmann, *Phys. Rev. Lett.* **120**, 123202 (2018).
- [5] S. Eilzer, H. Zimmermann, and U. Eichmann, *Phys. Rev. Lett.* **112**, 113001 (2014).
- [6] A. Hartung, S. Eckart, S. Brennecke, J. Rist, D. Trabert, K. Fehre, M. Richter, H. Sann, S. Zeller, K. Henrichs, et al., *Nat. Phys.* **15**, 1222 (2019).
- [7] B. Willenberg, J. Maurer, B. W. Mayer, and U. Keller, *Nat. Commun.* **10**, 5548 (2019).
- [8] J. Maurer, B. Willenberg, J. Daněk, B. W. Mayer, C. R. Phillips, L. Gallmann, M. Klaiber, K. Z. Hatsagortsyan, C. H. Keitel, and U. Keller, *Phys. Rev. A* **97**, 013404 (2018).
- [9] O. Hemmers, R. Guillemin, E. P. Kanter, B. Krässig, D. W. Lindle, S. H. Southworth, R. Wehlitz, J. Baker, A. Hudson, M. Lotrakul, et al., *Phys. Rev. Lett.* **91**, 053002 (2003).
- [10] 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).
- [11] M. Førre, J. P. Hansen, L. Kocbach, S. Selstø, and L. B. Madsen, *Phys. Rev. Lett.* **97**, 043601 (2006).
- [12] H. Bachau, M. Dondera, and V. Florescu, *Phys. Rev. Lett.* **112**, 073001 (2014).
- [13] M. Førre, S. Selstø, J. P. Hansen, and L. B. Madsen, *Phys. Rev. Lett.* **95**, 043601 (2005).
- [14] S. Chelkowski, A. D. Bandrauk, and P. B. Corkum, *Phys. Rev. Lett.* **113**, 263005 (2014).
- [15] S. Chelkowski, A. D. Bandrauk, and P. B. Corkum, *Phys. Rev. A* **92**, 051401(R) (2015).
- [16] S. Selstø, E. Lindroth, and J. Bengtsson, *Phys. Rev. A* **79**, 043418 (2009).
- [17] A. Di Piazza, C. Müller, K. Z. Hatsagortsyan, and C. H. Keitel, *Rev. Mod. Phys.* **84**, 1177 (2012).
- [18] Y. I. Salamin, S. X. Hu, K. Z. Hatsagortsyan, and C. H. Keitel, *Phys. Rep.* **427**, 41 (2006).
- [19] T. Kjellsson Lindblom, M. Førre, E. Lindroth, and S. Selstø, *Phys. Rev. Lett.* **121**, 253202 (2018).
- [20] H. Bauke, H. G. Hetzheim, G. R. Mocken, M. Ruf, and C. H. Keitel, *Phys. Rev. A* **83**, 063414 (2011).
- [21] T. Kjellsson, S. Selstø, and E. Lindroth, *Phys. Rev. A* **95**, 043403 (2017).
- [22] M. S. Pindzola, J. A. Ludlow, and J. Colgan, *Phys. Rev. A* **81**, 063431 (2010).
- [23] M. Førre, *Phys. Rev. A* **99**, 053410 (2019).
- [24] T. E. Moe and M. Førre, *Phys. Rev. A* **97**, 013415 (2018).
- [25] M. Førre and A. S. Simonsen, *Phys. Rev. A* **93**, 013423 (2016).
- [26] A. S. Simonsen and M. Førre, *Phys. Rev. A* **93**, 063425 (2016).
- [27] T. Kjellsson, M. Førre, A. S. Simonsen, S. Selstø, and E. Lindroth, *Phys. Rev. A* **96**, 023426 (2017).
- [28] M. Y. Ryabikin and A. M. Sergeev, *Opt. Express* **7**, 417 (2000).
- [29] 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).
- [30] A. V. Kim, M. Y. Ryabikin, and A. M. Sergeev, *Phys. Usp.* **42**, 54 (1999).
- [31] M. Y. Emelin and M. Y. Ryabikin, *Phys. Rev. A* **89**, 013418 (2014).
- [32] S. Brennecke and M. Lein, *J. Phys. B: At. Mol. Opt. Phys.* **51**, 094005 (2018).
- [33] H. Bachau, E. Cormier, P. Decleva, J. E. Hansen, and F. Martín, *Rep. Prog. Phys.* **64**, 1815 (2001).
- [34] M. Førre and A. S. Simonsen, *Phys. Rev. A* **90**, 053411 (2014).