Exact Nondipole Kramers-Henneberger Form of the Light-Atom Hamiltonian: An Application to Atomic Stabilization and Photoelectron Energy Spectra

M. Førre,1 S. Selstø,1 J. P. Hansen,1 and L. B. Madsen2

1Department of Physics and Technology, University of Bergen, N-5007 Bergen, Norway
2Department of Physics and Astronomy, University of Aarhus, 8000 Aarhus, Denmark

The general field of laser-matter interactions is characterized by impressive progress in light-source technology. Light sources with pulses of shorter and shorter duration and ever increasing intensities are being developed. Pulses containing only a few cycles and with a duration of less than 10 fs are now commercially available [1]. Intensities of 10^{14} W/cm^2 are routinely provided, and intensities two orders of magnitude higher, reaching the field strength of the Coulomb interaction in atoms and molecules, are not unusual. Femtosecond laser pulses have been used to produce coherent extreme-ultraviolet pulses of attosecond duration, and the expression “attosecond metrology” [2] was coined for the investigation of matter with such short pulses [3]. Other developments include the large-scale intense free-electron laser projects at DESY (Hamburg, Germany) and SLAC (Stanford, USA). The TESLA test facility in Hamburg has begun operation in the far-ultraviolet regime, and, e.g., a study of the interaction of intense soft X-rays with atom clusters was reported [4]. The clusters absorbed energy much more efficiently than anticipated from existing models, and the physical mechanism responsible for the excess in the absorbed energy is currently subject to some controversy [5].

Typically the laser-atom interaction is described in the dipole approximation where several equivalent formulations exist; the most popular ones being the velocity gauge, the length gauge and the Kramers-Henneberger frame [6]. It is, however, clear that the new light sources alluded to above pave the way for studies of atomic and molecular systems under extreme nonperturbative conditions [7]. In the case of atoms interacting with light from the vacuum-ultra-violet free-electron laser the dipole approximation cannot be expected to be valid [8]. Thus, motivated by the need to include the full \( k \cdot r \) term in the description of the light-matter interaction, we here revisit the question of equivalent formulations of electrodynamics.

We transform the exact nondipole minimal-coupling Hamiltonian for an atom in an explicitly time- and space-dependent laser field into the rest frame of a classical free electron in the laser field. In the dipole approximation, this frame is known as the Kramers-Henneberger frame [9]. Our transformed exact nondipole Hamiltonian takes a simple form and is very useful for the discussion of strong-field dynamics. We apply it to the study of \( H \) in the high-intensity, high-frequency regime, and confirm the phenomenon of atomic stabilization, i.e., the possibility of having a decreasing ionization probability-rate with increasing intensity (for reviews see, e.g., [10]). Most importantly, we point out that the onset of the dynamic stabilization can be directly observed from electron energy spectra. [Atomic units (a.u.) with \( m_e = e = \hbar = 1 \) are used throughout. All derivations are straightforwardly generalized to atoms and molecules involving more electrons.]

The minimal coupling scheme determines the Hamiltonian for a charged particle in an electromagnetic field through the vector potential \( A(\eta) \) with \( \eta = \omega t - k \cdot r \), and \( k \) the wave number. The scheme implies that the canonical momentum is obtained by \( p \to p - qA \) and for an electron of charge \( q = -1 \) in atomic units, we have \( p + A \), and the time-dependent Schrödinger equation reads

\[
i\hbar \frac{\partial}{\partial t} \Psi_v(r, t) = \left[ (p + A(\eta))^2 + V(r) \right] \Psi_v(r, t),
\]

where the subscript \( v \) refers to the velocity gauge. The advantage of this formulation is that the spatial dependence of the field is explicitly accounted for through its presence in the vector potential. A disadvantage is that the interaction is not expressed in terms of the physical \( E- \) and \( B-\) fields. Also numerically, the evaluation of the action of the \( A \cdot p \)-term can be quite involved unless a diagonal representation of \( \Psi_v \) with respect to this operator is applied. Until now only the alternative multipole formulation of Power-Zienau-Woolley [6, 11] has, in principle, kept the spatial dependence to all orders. The multipolar form represents the interaction in terms of the physical fields and the electron coordinate \( r \), but, as the name suggests, it is inherently designed to provide an expansion of the light-matter interaction, and consequently very impractical if one wishes to retain \( k \cdot r \) to all orders.

Here, we transform the Schrödinger equation into a new form by applying a nondipole Kramers-Henneberger transformation. Let

\[
\Psi_{KH} = U \Psi_v = \exp[i \alpha(\eta) \cdot p] \Psi_v,
\]

PACS numbers: 42.50.Hz,32.80.Rm,32.80.Fb.
earlier and found to have a detrimental effect on the stability [14]. However, expected to be within experimental reach in the near future [13]. With the development of new light sources, dynamic stabilization of ground state atoms is, how- ever, expected to be within experimental reach in the near future.

Nondipole terms were investigated in approximate ways earlier and found to have a detrimental effect on the stabilization [13]. The relative role of the different nondipole terms in [13] is illustrated in Fig. 1 for a two-dimensional model atom [16]. The ground state was exposed to a laser pulse propagating in the $x$ direction and of linear polarization $\mathbf{u}_p$ along the $z$-axis corresponding to the vector potential $\mathbf{A}(\eta) = \frac{E_0}{\omega} f(\eta) \sin(\eta) \mathbf{u}_p$, with $f(\eta)$ the envelope, $E_0$ the electric field amplitude and $\phi$ a phase that ensures that the vector potential represents a physical field [17]. The wave function was propagated on a Cartesian grid by means of the split-step operator technique [18]. A 5-cycle laser pulse with central frequency $\omega = 1$ a.u. (46 nm) corresponding to the pulse duration $T = 700$ as, and with carrier-envelope $f(\eta) = \sin^2\left(\frac{\eta}{2\pi}\right)$, was employed. The intensity range was set to $0 < I_0 < 1.4 \times 10^{19}$ W/cm$^2$. The population not accounted for in Fig. 1 is left in excited states. The effects of the last three terms in the Hamiltonian as well as the spatial dependence of the quiver potential $\alpha(r,t)$ cannot be resolved on the scale of Fig. 1. Hence, the nondipole effect observed for field strengths greater than 7 a.u. are exclusively related to the spatial dependence of the $A^2$-term.

As a first application of the new form of the Hamiltonian we consider the interaction with high-intensity, high-frequency fields. In this so-called stabilization regime [14], atoms may go through a region of decreasing ionization for increasing field strength. Stabilization was experimentally observed with Rydberg atoms [13]. With the development of new light sources, dynamic stabilization of ground state atoms is, however, expected to be within experimental reach in the near future [14].

Nondipole terms were investigated in approximate ways earlier and found to have a detrimental effect on the stabilization [12]. The relative role of the different nondipole terms in [13] is illustrated in Fig. 1 for a two-dimensional model atom [16]. The ground state was exposed to a laser pulse propagating in the $x$ direction and of linear polarization $\mathbf{u}_p$ along the $z$-axis corresponding to the vector potential $\mathbf{A}(\eta) = \frac{E_0}{\omega} f(\eta) \sin(\eta) \mathbf{u}_p$, with $f(\eta)$ the envelope, $E_0$ the electric field amplitude and $\phi$ a phase that ensures that the vector potential represents a physical field [17]. The wave function was propagated on a Cartesian grid by means of the split-step operator technique [18]. A 5-cycle laser pulse with central frequency $\omega = 1$ a.u. (46 nm) corresponding to the pulse duration $T = 700$ as, and with carrier-envelope $f(\eta) = \sin^2\left(\frac{\eta}{2\pi}\right)$, was employed. The intensity range was set to $0 < I_0 < 1.4 \times 10^{19}$ W/cm$^2$. The population not accounted for in Fig. 1 is left in excited states. The effects of the last three terms in the Hamiltonian as well as the spatial dependence of the quiver potential $\alpha(r,t)$ cannot be resolved on the scale of Fig. 1. Hence, the nondipole effect observed for field strengths greater than 7 a.u. are exclusively related to the spatial dependence of the $A^2$-term.

![FIG. 1: Ionization and ground state probability for a two-dimensional model atom ](image)

which holds for a general elliptically polarized field. Within the dipole approximation $A$ and $\alpha$ are space-independent, the last three terms are absent, and [13] reduces to the well-known result [9]. However, expected to be within experimental reach in the near future [13]. With the development of new light sources, dynamic stabilization of ground state atoms is, however, expected to be within experimental reach in the near future [14].

To evaluate the effect of the unitary translation operators in [13], we use the operator identity known as the Baker-Hausdorff lemma [12] and take advantage of the Coulomb gauge restriction $[p,A] = 0$ and $k \cdot A = 0$. The resulting Hamiltonian reads

$$H_{KH} = \frac{p^2 + A^2}{2} + V(r + \alpha) + \frac{k^2}{2} \left( \frac{d\alpha}{d\eta} \cdot p \right)^2 + \frac{ik^2}{2} \frac{d^2\alpha}{d\eta^2} \cdot p + \left( \frac{d\alpha}{d\eta} \cdot p \right) (k \cdot p),$$

where

$$\alpha(\eta) \equiv \frac{1}{\omega} \int_{\eta}^{\eta_0} d\eta' A(\eta')$$

represents the quiver motion relative to the laboratory frame of a classical free electron in the field. The Hamiltonian corresponding to the new point of view is obtained by taking the time-derivative on both sides of [13], and by using [13] for $\Psi$, we obtain

$$i\partial_t \Psi_{KH}(r,t) = H_{KH} \Psi_{KH}(r,t)$$

with

$$H_{KH} = U H_0 U^\dagger + i (\partial_t U) U^\dagger.$$
and the initial field-free H(1s) state is obtained from the exact analytical expression. Reflection at the edges $r = r_{\text{max}} = 200$ a.u. is avoided by imposing an absorbing boundary. For convergence, we include harmonics up to $l_{\text{max}} = 29$, check for gauge invariance, use propagation time-step $\Delta t = 0.01$ a.u., and set $\Delta r = 0.2$ a.u.. Photoelectron probability distributions are calculated by projecting the wave function onto the field-free (discretized) continuum states. We note that the presence of nondipole terms will lead to a population of different $m$-values in (6).

In Fig. 2 total ionization- and ground state probabilities are shown for the fully three-dimensional case in the nondipole and dipole limits for two different frequencies. We observe that the dipole approximation remains valid up to field strengths of the order of 10 a.u., and we find in general only a small effect of the nondipole terms on stabilization.

We now turn to the central question of how stabilization can most efficiently be experimentally detected. Measurement of absolute probabilities will require control of all parameters of the experiment: atom density, pulse characteristics, repetition rates, electron counts etc.. We therefore suggest measuring the energy-differential photoelectron spectrum. Figure

FIG. 2: Upper panel: As Fig. 1 but for the fully three-dimensional case with the system initially prepared in the H(1s) state. Lower panel: corresponding results for $\omega = 2$ a.u.

FIG. 3: Upper panel: Ionization probability density $dP/dE$ for H(1s) vs. electric field strength and energy of the ionized electron for a squared shaped 5-cycle pulse (380 as) with $\omega = 2$ a.u. (23 nm) by fully three-dimensional calculations. Middle panel: Results from the time-averaged potential. Lower panel: Total ionization probability in the dipole approximation (full curve) and for the time-averaged potential (dashed curve).
shows the ionization probability density $dP/dE$ vs. electric field strength and energy $E$ of the ionized electron with the full interaction potential (upper panel) and with the time-averaged Kramers-Henneberger potential (middle panel) [10],

$$V_0(\alpha_0; \mathbf{r}) = \frac{1}{T} \int_0^T V(\mathbf{r} + \alpha dt, (7)$$

where $\alpha_0 \equiv E_0/\omega^2$ is the quiver amplitude. The dipole and nondipole results are practically identical, and only the dipole results are shown in Fig. 3. For lower field strengths a regular pattern of multiphoton resonances corresponding to absorption of $1\omega, 2\omega$ or $3\omega$ from the field is present. However, the multiphoton ionization process weakens at higher intensities as the stabilization sets in. Simultaneously, there is a steady growth in the portion of low-energy photoelectrons in the spectrum which can be assigned to $V_0$ of (7). That $V_0$ is responsible for the growth in the low-energy spectrum is readily seen by comparison of the upper and middle panels. The processes leading to ionization effectively divide into two competing classes: The multiphoton ionization superimposed on a monotonically increasing ‘background’ ionization process due to $V_0$ solely. This is explicitly illustrated in the lower panel of Fig. 3 where the total ionization probability vs. electric field strength is shown [20]. Multiphoton ionization dominates at lower field strengths, whereas the picture is the opposite at higher values of $E_0$. The ionization due to the $V_0$ potential reflects to what extent the laser pulse is turned-on and off adiabatically, and in a ‘sudden approximation’ picture it represents the lack of overlap between the field-free and the field-dressed states. Common in both photoelectron spectra is the presence of peaks in the probability density which cannot be attributed to multiples of $\omega$. Instead, they are a result of the non-adiabatic turn-on and turn-off of the field and can be associated with the higher-order Fourier components of the pulse.

In summary, we presented a new formulation of the interaction between atoms and light maintaining full spatial dependence of the fields. We analyzed the terms in the interaction Hamiltonian and argued, supported by numerical evidence, that certain terms can be neglected. For the present field parameters, the main nondipole effects come from $A(\eta)^2$. As an application, we considered the phenomenon of dynamic stabilization in intense high-frequency fields. We showed by full three-dimensional wavepacket simulations that the nondipole terms do not destroy the stabilization effect, and most importantly that the photoelectron spectra in the stabilization regime show very characteristic features: After onset of stabilization all ionized electrons have very low kinetic energy. Thus, by simply measuring the energy of the released electrons stabilization can be detected.

It is a pleasure to thank Thomas K. Kjeldsen for useful discussions and for critically reading the manuscript. The present research was supported by the Norwegian Research Council through the NANOMAT program and the Nordic Research Board NordForsk and by the Danish Natural Science Research Council.

[1] T. Brabec, and F. Krausz, Rev. Mod. Phys. 72, 545 (2000).
[2] M. Hentschel, R. Kienberger, Ch. Spielmann, G. A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher, and F. Krausz, Nature (London) 414, 509 (2001).
[3] R. Kienberger, E. Goulielmakis, M. Uiberacher, A. Baltuska, V. Yakovlev, F. Bammer, A. Scrinzi, Th. Westerwalbesloeh, U. Kleineberg, U. Heinzmann, M. Drescher, and F. Krausz, Nature (London) 427, 817 (2004).
[4] H. Wabnitz, L. Bittner, A. R. B. de Castro, R. Döhrmann, P. Gürtler, T. Laarmann, W. Laasch, J. Schulz, A. Swiderski, K. von Haeften, T. Möller, B. Faatz, A. Fateev, J. Feldhaus, C. Gerth, U. Hahn, E. Saldin, E. Schneidmiller, K. Sytchev, K. Tiedke, R. Treusch, and M. Yurkov, Nature (London) 420, 482 (2002).
[5] R. Santra and C. H. Greene, Phys. Rev. Lett. 91, 233401 (2003); C. Siedschlag, and J. M. Rost, Phys. Rev. Lett. 93, 043402 (2004).
[6] C. Cohen-Tannoudji, J. Dupont-Roc, G. Grynberg, Introduction to Quantum Electrodynamics (Wiley, New York, 1997); R. Loudon, The quantum theory of light (Claredon press, Oxford, 1983).
[7] K. J. Meharg, J. S. Parker, and K. T. Taylor, J. Phys. B 38, 237 (2005).
[8] A. V. Lugovskoy, and I. Bray, J. Phys. B 37, 3427 (2004).
[9] W. Pauli and M. Fierz, Nuovo Cimento 15, 167 (1938); H. A. Kramers, Collected scientific papers (Amsterdam: North-Holland) p 866 (1956); W. C. Henneberger, Phys. Rev. Lett. 21, 838 (1968).
[10] M. Gavrila, J. Phys. B 35, 147 (2002); A. M. Popov, O. V. Tikhonova, and E. A. Volkova, J. Phys. B 36, 125 (2003).
[11] E. A. Power, and S. Zienau, Phys. Rev. A 48, 427 (1995); R. G. Wooley, Proc. Roy. Soc. A 321, 557 (1971).
[12] For Hermitian operators $A$ and $G$ and a real parameter $\lambda$ this lemma states that $\exp(iG\lambda)A\exp(-iG\lambda) = A + i\lambda[G,A] + (i^2\lambda^2/2!)[G,[G,A]] + \ldots + (i^n\lambda^n/n!)[G,[G,\ldots,[G,A]],\ldots] + \ldots$. See, e.g., J. J. Sakurai, Modern Quantum Mechanics (Addison-Wesley, Reading, 1994).
[13] M. P. de Boer, J. H. Hoogenraad, R. B. Vrijen, L. D. Noordam, and H. G. Muller, Phys. Rev. Lett. 71, 3263 (1993).
[14] M. Dondera, H. G. Muller, and M. Gavrila, Phys. Rev. A 65, 031405 (2002).
[15] A. Bugacov, M. Pont, and R. Shakeshaft, Phys. Rev. A 48, 4027 (1993).
[16] 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).
[17] L. B. Madsen, Phys. Rev. A 65, 053417 (2002).
[18] M. D. Feit, A. J. Fleck, Jr., and A. Steiger, J. Comput. Phys. 47, 412 (1982).
[19] J. P. Hansen, T. Svetrik, and L. B. Madsen, Phys. Rev. A 68, 031401(R) (2003); T. Birkeland, M. Førre, J. P. Hansen, and S. Selstø, J. Phys. B 37, 4205 (2004).
[20] Note that the difference between the full curve in the lower panel of Fig. 3 and the ionization curve in Fig. 2 is due to the different pulse forms used.