

# Paper II

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

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The exact nondipole minimal-coupling Hamiltonian for an atom interacting with an explicitly time- and space-dependent laser field is transformed into the rest frame of a classical free electron in the laser field, i.e., into the Kramers-Henneberger frame. The new form of the Hamiltonian is used to study nondipole effects in the high-intensity, high-frequency regime. Fully three-dimensional nondipole *ab initio* wave packet calculations show that the ionization probability may decrease for increasing field strength. We identify a unique signature for the onset of this dynamical stabilization effect in the photoelectron spectrum.

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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<sup>2</sup> are routinely provided, and intensities 2 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-ultraviolet free-electron laser the dipole approximation cannot be expected to be valid [8]. Thus, motivated by the need to include the full  $\mathbf{k} \cdot \mathbf{r}$  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 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  $\mathbf{A}(\eta)$  with  $\eta \equiv \omega t - \mathbf{k} \cdot \mathbf{r}$ , and  $\mathbf{k}$  the wave number. The scheme implies that the canonical momentum is obtained by  $\mathbf{p} \rightarrow \mathbf{p} - q\mathbf{A}$  and for an electron of charge  $q = -1$  in atomic units, we have  $\mathbf{p} + \mathbf{A}$ , and the time-dependent Schrödinger equation reads

$$i\partial_t \Psi_v(\mathbf{r}, t) = \left[ \frac{1}{2} (\mathbf{p} + \mathbf{A}(\eta))^2 + V(\mathbf{r}) \right] \Psi_v(\mathbf{r}, t), \quad (1)$$

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  $\mathbf{A} \cdot \mathbf{p}$  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  $\mathbf{r}$ , 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  $\mathbf{k} \cdot \mathbf{r}$  to all orders.

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

$$\Psi_{\text{KH}} = U\Psi_v = \exp[i\boldsymbol{\alpha}(\boldsymbol{\eta}) \cdot \mathbf{p}]\Psi_v, \quad (2)$$

where

$$\boldsymbol{\alpha}(\boldsymbol{\eta}) \equiv \frac{1}{\omega} \int_{\boldsymbol{\eta}_i}^{\boldsymbol{\eta}} d\boldsymbol{\eta}' \mathbf{A}(\boldsymbol{\eta}') \quad (3)$$

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 (2), and by using (1) for  $\Psi_v$ , we obtain  $i\partial_t \Psi_{\text{KH}}(\mathbf{r}, t) = H_{\text{KH}} \Psi_{\text{KH}}(\mathbf{r}, t)$  with

$$H_{\text{KH}} = UH_v U^\dagger + i(\partial_t U)U^\dagger. \quad (4)$$

To evaluate the effect of the unitary translation operators in (4), we use the operator identity known as the Baker-Hausdorff lemma [12] and take advantage of the Coulomb gauge restriction  $[\mathbf{p}, \mathbf{A}] = 0$  and  $\mathbf{k} \cdot \mathbf{A} = 0$ . The resulting Hamiltonian reads

$$H_{\text{KH}} = \frac{p^2 + A^2}{2} + V(\mathbf{r} + \boldsymbol{\alpha}) + \frac{k^2}{2} \left( \frac{d\boldsymbol{\alpha}}{d\boldsymbol{\eta}} \cdot \mathbf{p} \right)^2 + \frac{ik^2}{2} \frac{d^2 \boldsymbol{\alpha}}{d\boldsymbol{\eta}^2} \cdot \mathbf{p} + \left( \frac{d\boldsymbol{\alpha}}{d\boldsymbol{\eta}} \cdot \mathbf{p} \right) (\mathbf{k} \cdot \mathbf{p}), \quad (5)$$

which holds for a general elliptically polarized field. Within the dipole approximation  $\mathbf{A}$  and  $\boldsymbol{\alpha}$  are space independent, the last three terms are absent, and (5) reduces to the well-known result [9]. In the nondipole case, the importance of these terms is readily understood, e.g., in terms of their effect on a continuum wave function. The two terms proportional to  $k^2$  are of the order of  $E_0^2 v^2 / (\omega^2 c^2)$  and  $E_0 v / c^2$ , respectively, whereas the last term is of order  $E_0 v^2 / (\omega c)$ . We thus see that the effect of the dominant term on a wave function is reduced by a factor  $\sim E_0 / (\omega c)$  compared to the  $p^2$  term. The factor  $E_0 / \omega$  is precisely the quiver velocity of the electron  $v_{\text{quiver}}$ , so we expect that the last three terms may be neglected as long as  $v_{\text{quiver}} / c \ll 1$ . Whenever this condition is fulfilled, the nonrelativistic approach is automatically justified as well. As it turns out, for the field parameters considered here, the effect of the nondipole terms is effectively given by the spatial dependence of the vector potential in 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 [10], 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 [15,16]. The relative role of the different nondipole terms in (5) is now discussed 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  $\mathbf{A}(\boldsymbol{\eta}) = \frac{E_0}{\omega} f(\boldsymbol{\eta}) \sin(\boldsymbol{\eta}) \mathbf{u}_p$  with  $f(\boldsymbol{\eta})$  the envelope and  $E_0$  the electric field amplitude. The wave function was propagated on a Cartesian grid by means of the split-step operator technique [17]. A 5-cycle laser pulse with central frequency  $\omega = 1$  a.u. (46 nm) corresponding to the pulse duration  $T = 760$  as, and with carrier-envelope  $f(\boldsymbol{\eta}) = \sin^2(\frac{\pi \boldsymbol{\eta}}{\omega T})$ , was employed. The intensity range was set to  $0 < I_0 < 1.4 \times 10^{19}$  W/cm<sup>2</sup>. Total ionization and ground state probability versus laser intensity is displayed in Fig. 1. The population not accounted for in the figure is left in excited states. The total effect of the last three terms in the Hamiltonian (5), as well as the spatial dependence of the quiver amplitude  $\boldsymbol{\alpha}(\mathbf{r}, t)$ , is so small that it cannot be resolved on the scale of Fig. 1.

We have, accordingly, justified that for the parameters under concern, it is a very accurate approach to apply the Hamiltonian (5), neglecting the last three additional kinetic energy terms arising from the transformation (2), to a fully three-dimensional study of ionization of a real ground state atom by intense short wave light field beyond the dipole approximation. We consider  $H(1s)$  exposed to 5-cycle laser pulses in the attosecond range with central frequencies  $\omega = 1$  a.u. and  $\omega = 2$  a.u.. The time-dependent Schrödinger equation is solved numerically based on a split-step operator approximation on a spherical grid as detailed elsewhere [18]. The wave function is expanded on the grid points  $[(r_i, \Omega_{jk}) = (r_i, \theta_j, \phi_k)]$  as

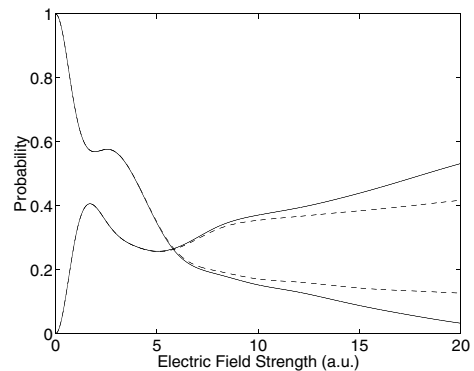


FIG. 1. Ionization and ground state probability for a two-dimensional model atom [16] in the nondipole (solid curve) and dipole (dashed curve) descriptions vs electric field strength for a 5 cycle pulse with  $\omega = 1$  a.u.

$$\Psi(r_i, \Omega_{jk}, t) = \sum_{l,m}^{l_{\max}, m_{\max}} f_{l,m}(r_i, t) Y_{l,m}(\Omega_{jk}), \quad (6)$$

and the initial field-free  $H(1s)$  state is obtained from the exact analytical expression. Reflection at the edges  $r = r_{\max} = 200$  a.u. is avoided by imposing an absorbing boundary. For convergence, we include harmonics up to  $l_{\max} = 29$ , check for gauge invariance, use propagation time step  $\Delta t = 0.01$  a.u., and set  $\Delta r = 0.2$  a.u.. Doubling  $r_{\max}$  only led to minor changes in the results. This confirms that in the present frequency regime Rydberg states are not dynamically involved [19]. Photoelectron probability distributions are calculated by projecting the wave function onto the field-free (discretized) continuum states. We note that the presence of non-dipole 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 non-dipole 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.

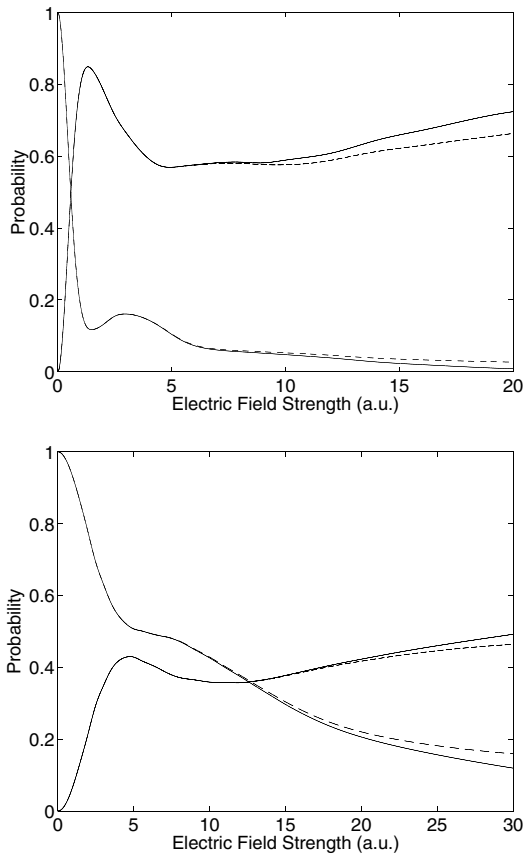


FIG. 2. Upper panel: As Fig. 1, but for the fully three-dimensional case with the system initially prepared in the  $H(1s)$  state. Lower panel: Corresponding results for  $\omega = 2$  a.u..

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 3 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],

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

where  $\alpha_0 \equiv E_0/\omega^2$  is the quiver amplitude. The dipole and nondipole results are practically identical, and only the

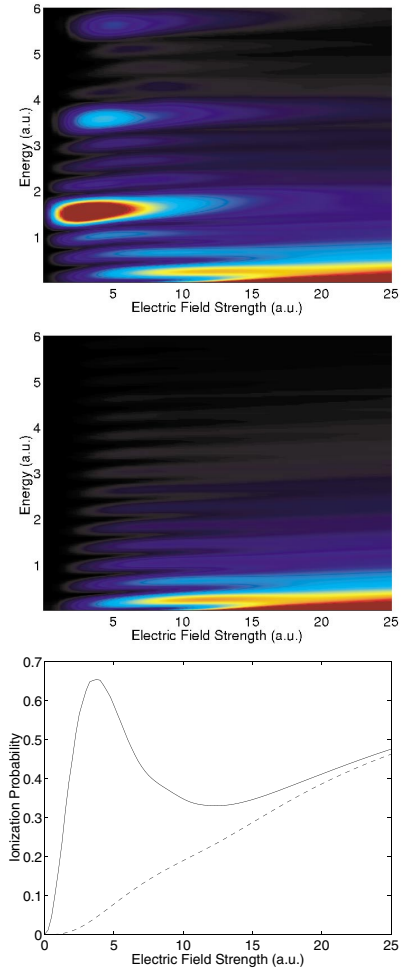


FIG. 3 (color). Upper panel: Ionization probability density  $dP/dE$  for  $H(1s)$  vs electric field strength and energy of the ionized electron for a squared shaped 5-cycle pulse (380 as) with  $\omega = 2$  a.u. (23 nm) by fully three-dimensional calculations. Middle panel: Results from the time-averaged potential (7). Lower panel: Total ionization probability in the dipole approximation (solid curve) and for the time-averaged potential (7) (dashed curve).

dipole results are shown in Fig. 3. In addition, the characteristic features observed in the figure are insensitive to the actual shape of the pulse. 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 solely to  $V_0$ . 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 nonadiabatic turn-on and turn-off of the field and can be associated with the higher-order Fourier components of the pulse.

In summary, we have presented a new formulation of the interaction between atoms and light maintaining full spatial dependence of the fields. We have 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 have considered the phenomenon of dynamic stabilization in intense high-frequency fields. We have shown by full three-dimensional wave packet simulations that the nondipole terms do not destroy the stabilization effect, and most importantly that the photoelectron spectra in the stabilization regime shows 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.

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- [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.