Atoms and molecules in intense attosecond fields: beyond the dipole approximation

M Førre¹, J P Hansen¹, L Kochbach¹, S Selstø⁵, T K Kjeldsen³ and L B Madsen³

¹ Department of Physics and Technology, University of Bergen, N-5007 Bergen, Norway
² Atomic Physics, Fysikum, Stockholm University, AlbaNova University Center, SE-106 91 Stockholm, Sweden
³ Lundbeck Foundation Theoretical Center for Quantum System Research, Department of Physics and Astronomy, University of Aarhus, 8000 Aarhus, Denmark

E-mail: morten.forre@ift.uib.no

Abstract. The exact non-dipole minimal-coupling Hamiltonian for an atomic system 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 has been used to study the non-dipole dynamics of atoms and molecules in intense XUV laser pulses. The time-dependent Schrödinger equation is solved without any simplifications.

1. Introduction

There has been much focus on the phenomenon of atomic stabilization in atoms and molecules during the last two decades [1, 2]. Atomic stabilization is a rather counterintuitive phenomenon where an atomic system, that is subjected to a high-frequency laser field, goes through a region of decreasing ionization probability or rate for increasing intensity of the field. The onset of stabilization is characterized by a transition from multiphoton-like ionization to ionization dominated by nonadiabatic shake-off as represented by a lack of overlap between the field-free and field-dressed states and with the emergence of characteristic low-energy electrons [3]. However, in very intense fields the non-dipole magnetic field component of the laser pulse may no longer be neglected [4], and a full quantum-mechanical analysis of the problem, with non-dipole effects included, is desirable [3].

Recently, the exact non-dipole Kramers-Henneberger (KH) form of the light-matter Hamiltonian was derived and used in a study of the role of non-dipole effects in superintense light-atom [3, 5] and light-molecule [6] interactions in the XUV regime. It was shown that high-intensity, high-frequency ionization leads to new non-dipole features, that are manifested by the appearance of a characteristic (non-dipole) lobe in the angular distributions of the ejected photoelectrons.

In this work, we revisit the problem of ionization of matter by superintense, high-frequency laser fields beyond the dipole approximation, and address the question of how the single hydrogen atom, H, and the simplest system exhibiting the chemical bond, H₂⁺, respond to such extreme conditions. We solve the time-dependent Schrödinger equation (TDSE) without any simplifications and obtain the electronic differential ionization spectra following from the ionization.
2. Derivation of the non-dipole Kramers-Henneberger Hamiltonian

The minimal coupling scheme determines the Hamiltonian for a charged particle with mass \( m \) and charge \( q \) in a classical electromagnetic field through the vector potential \( \mathbf{A}(\eta) \) with \( \eta \equiv \omega t - \mathbf{k} \cdot \mathbf{r} \). The scheme implies that the canonical momentum is obtained by \( \mathbf{p} \rightarrow \mathbf{p} - q \mathbf{A} \). Then, the TDSE reads

\[
i\hbar \partial_t \Psi_v(\mathbf{r}, t) = \left[ \frac{1}{2m} (\mathbf{p} - q \mathbf{A})^2 + V(\mathbf{r}) \right] \Psi_v(\mathbf{r}, t),
\]

where the subscript \( v \) refers to the velocity gauge. The wave vector \( \mathbf{k} \) satisfies \( \omega/k = c \). For a transversal wave, the propagation direction is perpendicular to the polarization, \( \mathbf{k} \perp \mathbf{A} \), which corresponds to applying the Coulomb gauge restriction, \( \nabla \cdot \mathbf{A} = 0 \). The Coulomb gauge will be applied throughout this paper. One advantage of the formulation (1) is that the spatial dependence of the field is explicitly accounted for through its presence in the vector potential. However, from a numerical point of view, a disadvantage is that 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.

The TDSE (1) is transformed into a new form by applying a non-dipole Kramers-Henneberger transformation [3]. Let

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

where

\[
\alpha(\eta) \equiv -\frac{q}{\omega m} \int_{\eta_0}^\eta \mathbf{A}(\eta') d\eta'
\]

represents the position relative to the laboratory frame of a classical free electron moving 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

\[
i\hbar \partial_t \Psi_{KH}(\mathbf{r}, t) = H_{KH} \Psi_{KH}(\mathbf{r}, t)
\]

with

\[
H_{KH} = U H_U U^\dagger + i(\partial_t U)U^\dagger.
\]

To evaluate the effect of the unitary translation operators in (4), we use the Baker-Hausdorff lemma [7],

\[
e^{\lambda G} A e^{-\lambda G} = A + \lambda [G, A] + \frac{\lambda^2}{2!} [G, [G, A]] + \ldots
\]

The resulting Hamiltonian, which holds for a general elliptically polarized field, reads [3, 8]

\[
H_{KH} = \frac{p^2}{2m} + V(\mathbf{r} + \alpha) + \frac{q^2}{2m} \mathbf{A}^2 + \frac{1}{2mc^2} \left( \frac{q}{m} \mathbf{A} \cdot \mathbf{p} \right)^2 - \frac{1}{2mc^2} \left[ (\hat{\mathbf{k}} \cdot \mathbf{p}) \left( \frac{q}{m} \mathbf{A} \cdot \mathbf{p} \right) + \left( \frac{q}{m} \mathbf{A} \cdot \mathbf{p} \right) (\hat{\mathbf{k}} \cdot \mathbf{p}) \right]
\]

Within the dipole approximation \( \mathbf{A} \) and \( \alpha \) are space-independent, the last three terms are absent, and (6) reduces to the well-known result [9, 10, 11].

In ab initio calculations with superintense, high-frequency fields, the KH frame outmatches completely both the length gauge and the velocity gauge in computational speed. Therefore, we are now going to use the KH light-matter Hamiltonian to study a model of the non-dipole ionization dynamics of the hydrogen atom in the ground state, \( \text{H}(1s) \), and the hydrogen molecular ion, \( \text{H}_2^+ \), in the \( 1\sigma_g \) and \( 1\sigma_u \) states, respectively. The systems are subject to a linearly polarized laser pulse propagating along the positive \( z \) direction, corresponding to the vector potential,

\[
\mathbf{A}(t-z/c) = \frac{E_0}{\omega} \sin^2 \frac{\pi (t-z/c)}{T} \sin \frac{\omega(t-z/c) + \phi}{T} \hat{x}
\]

Here, \( E_0 \) is the peak electric field amplitude, \( \omega \) is the central frequency, \( T \) is the total pulse duration, \( \phi \) is the carrier-envelope phase, \( c \) is the speed of light, and \( \hat{x} \) indicates the polarization direction. Atomic units (a.u.), with \( m_e = e = \hbar = a_0 = 1 \), are used hereafter.
Figure 1. Total ionization probability vs peak electric field strength for H(1s) exposed to a 5-cycle linearly polarized laser pulse with $\omega = 2$ a.u., in the non-dipole (black solid curve) and dipole (red dashed curve) cases, respectively. The laser pulse has a sine-square profile.

3. Ionization of H(1s) in superintense XUV laser pulses

We consider H(1s) exposed to a 5-cycle laser pulse in the attosecond range with central frequency $\omega = 2$ a.u.. The TDSE is solved numerically based on a split-step operator approximation on a spherical grid as detailed elsewhere [12, 13]. The wave function is expanded on the grid points $[r_i, \Omega_{jk}] = (r_i, \theta_j, \phi_k)$ as

$$\Psi_{KH}(r_i, \Omega_{jk}, t) = \sum_{l,m} \int_{\Omega} f_{l,m}(r_i, t) Y_{l,m}(\Omega_{jk}),$$

(8)

and the initial field-free H(1s) state is obtained from the exact analytical expression. Reflection at the edges $r = r_{max} = 100$ a.u. is avoided by imposing an absorbing boundary. Converged and gauge invariant results were obtained with $l_{max} = 29$, propagation time-step $\Delta t = 0.0075$ a.u., and $\Delta r = 0.2$ a.u.. The grid representation in all three coordinates makes the method very flexible. In particular, the method imposes no symmetry demands on the system under study. Photoelectron probability distributions and total ionization probabilities are calculated from the projection of the final wave function on the continuum states,

$$\frac{\partial P}{\partial \Omega \partial k} = \sum_{l,m} (-i)^l e^{i\sigma_l} Y_{lm}({\Omega}) (\Phi^k_C | \Psi_{KH}(t = T))$$

(9)

Here, $\Psi(t = T)$ is the wave function at the end of the pulse, $\Phi^k_C$ is the radial continuum wave function (normalized on the $k$ scale), $k = \sqrt{2E}$ is the wave number, and $\sigma_l = \arg \Gamma (l + 1 - \frac{1}{2} k)$ is the Coulomb phase shift. We note that in the presence of the non-dipole field the different $m$ components of the wave function in (8) couple.

It is well known that an atomic system interacting with high-intensity, high-frequency fields may go through a region of decreasing ionization for increasing field strength, i.e. it enters the so-called stabilization regime [1]. In such high fields the non-dipole magnetic field component of the laser pulse may no longer be neglected, and it has been pointed out that the magnetic field could have a detrimental effect on the degree of stability [4]. In figure 1 the total ionization probability vs peak...
Figure 2. Calculated dipole (red dashed curve) and non-dipole (black solid curve) differential ionization probabilities $dP/dE$ vs kinetic energy of the ejected electron for three different laser intensities in figure 1, i.e. for $E_0 = 5, 20$ and 45 a.u.. Note that for $E_0 = 5$ a.u. the dipole and non-dipole calculation results cannot be distinguished in the figure.

The electric field strength is shown for the non-dipole (black solid curve) and dipole (red dashed curve) cases, respectively [3]. We find that the dipole approximation remains valid up to field strengths of the order of 10 a.u., and we also find in general only a small effect of the non-dipole terms on stabilization.

Figure 2 shows the calculated dipole (red dashed curve) and non-dipole (black solid curve) ionization probability densities $dP/dE$ vs kinetic energy of the ejected electron for three different field strength, $E_0 = 5, 20$ and 45 a.u.. For the lowest field strength ($E_0 = 5$ a.u.), the dipole and non-dipole results practically coincide, and 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 spectra. The dipole and non-dipole results are rather similar, even for the highest peak intensity, but it seems like the magnetic component of the laser pulse tends to wash out the remaining interference oscillations in the dipole spectra. Furthermore, the fraction of low-energy electrons is greater in the non-dipole case than in the dipole case.

Figure 3 shows the dipole (left) and non-dipole (right) angular distributions of the ejected electron for the three cases in figure 2. The data are averaged over the carrier-envelope phase $\phi$. The horizontal axis ($x$ axis) indicates the laser polarization direction, and the pulse propagates in the positive $z$ direction (upwards). The difference between the dipole and non-dipole cases is striking: the common $p$-like shape in the weak field (dipole) limit (upper panel) develops gradually into a characteristic three-lobe shape in the super-strong field (non-dipole) regime (lower panel) [5]. Essentially the same features as were reported for the ionization of $H(4s)$ at optical frequencies [14]. This opens the possibility that the effect could be measured in the near future by applying a low-frequency laser pulse.

The appearance of the extra (non-dipole) lobe in the distributions can be explained from a newly proposed non-dipole, high-frequency ionization mechanism [5]. The mechanism has a direct classical counterpart and is understood from a classical collision model, where the electron escapes from the nucleus as a result of an intimate interplay between the electromagnetic and Coulomb forces on the electronic orbit during the pulse. A free electron subject to the combined electric and magnetic fields in the pulse is displaced in the laser propagation direction due to the Lorentz force acting on it. For the very intense non-dipole field considered here the bound electron behaves as being a free particle in the field, and at some point its trajectory is practically confined to the upper hemisphere (positive $z$-coordinate). However, in this displaced position the electron goes through successive encounters with the nucleus that may ultimately be sufficient to release the electron with a dominating momentum component in the negative $z$ direction.
4. Ionization of H$_2^+$ ($1\sigma_g$) and H$_2^+$ ($1\sigma_u$) in superintense XUV laser pulses

Figure 4 shows the corresponding probabilities for electron ejection in different directions of H$_2^+$ after interaction with a linearly polarized 6-cycle laser pulse with central frequency $\omega = 3$ a.u. and peak electric field amplitude of $E_0 = 50$ a.u. [6]. The initial state is in (a) the symmetric ground state ($1\sigma_g$) and in (b) the lowest anti-symmetric state ($1\sigma_u$). In this case, the angular differential ionization probabilities are calculated from the radial probability current flowing through a spherical boundary placed at some large distance $R_b \approx 70a_0$, i.e.,

$$\frac{\partial P}{\partial \Omega} = \int_{t_0}^{T_f} \text{Im} \left( r^2 \Psi^* \frac{\partial \Psi}{\partial r} \right)_{r=R_b} \, dt,$$

with $t_0$ the instant of time just after the end of the pulse and $R_b$ large enough to ensure that no flux has passed it during the pulse. The final time $T_f$ in (10) is such that all the outgoing flux has had time to pass $R_b$. The total size of the sphere used for the flux analysis is $150a_0$, and an absorbing boundary prevents unphysical reflections.

The angular distributions display a multi-lobe structure with a dominating electron ejection in the direction opposite to the pulse propagation direction in the non-dipole field (see [6] for further details on superintense laser-molecule dynamics in linearly- and circularly polarized laser fields). One striking difference between the gerade and ungerade situation is that there is no electron ejection in the plane perpendicular to the internuclear axis for the ungerade case. Even the non-dipole lobe is cut into two pieces because of the nodal plane. This stands in complete contrast to the case with the gerade initial state where the electron is likely to escape in all directions defined by this plane. The whole difference can be attributed to the underlying anti-symmetry of the initial $1\sigma_u$ state and to two-center interference effects [6].

5. Conclusions

In conclusion, we presented a new formulation of the interaction between atoms and light maintaining full spatial dependence of the fields. By full three-dimensional wavepacket simulations we have solved the time-dependent Schrödinger equation beyond the dipole approximation, for the interaction
Figure 4. Dipole (left) and non-dipole (right) electron angular distributions of $H_2^+ (1\sigma_g)$ (b) and $H_2^+ (1\sigma_u)$ (c) after interaction with a linearly polarized 6-cycle pulse with $\omega = 3$ a.u. and peak electric field amplitude of $E_0 = 50$ a.u.. The laser pulse has a sine-square profile and is propagating in the upward direction. The polarization axis is indicated by the dashed line. The data are averaged over the carrier-envelope phase. The upper panel (a) shows the orientation of the molecule with respect to the pulse.

of $H(1s)$, $H_2^+ (1\sigma_g)$ and $H_2^+ (1\sigma_u)$ with very intense, high-frequency, attosecond fields, and obtained the photoelectron probability distributions. The main finding is a characteristic non-dipole lobe in the angular distributions of the ejected electron. The presence of this lobe, both in the atomic and the molecular case, only emphasizes its general importance in superintense light-matter interactions.

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