Nondipole photoelectron momentum shifts in strong-field ionization with mid-infrared laser pulses of long duration

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Abstract

We consider atomic strong-field ionization in the long-pulse limit for linearly polarized infrared laser fields. We show how nondipole effects in the plane formed by the propagation and polarization directions lead to (i) a shift of the origin of the rings describing the energetically allowed final electron momenta for individual above-threshold ionization (ATI) channels and (ii) a redistribution in the continuum population on each ring. In this manner, the photoelectron energy in a given ATI channel depends on the direction of the ejected electron.

Keywords: nondipole, photoelectron, strong-field ionization

(Some figures may appear in colour only in the online journal)

1. Introduction

Nondipole effects show up in strong-field ionization at near- and mid-infrared wavelengths and moderate to high intensities due to radiation pressure and magnetic field effects [1–4]. When the dipole approximation is accurate, the photoelectron momentum distribution (PMD) is symmetric under reflection in the laser polarization plane, i.e. under $k_x \rightarrow -k_x$, where $k_x$ is the momentum along the propagation direction $x$ (atomic units (a.u.) are used throughout). Nondipole effects break this symmetry.

During the last decade, observations of nondipole-induced shifts of the PMD along the laser propagation direction in strong-field ionization experiments [5–10] have fueled interest as recently reviewed [11–13]. The experimental works [5–9, 14] used pulsed lasers of femtosecond duration. The finite pulse durations impeded resolving the individual above-threshold ionization (ATI) channels. The focus of the experiments [5–9] was therefore on the overall shift of the momentum distribution along the laser propagation direction. The shift can be parameterized in terms of well-known quantities such as ionization, $I_p$, and ponderomotive, $U_p$, potentials [15]. A saddle-point analysis in the short-pulse limit based on the strong-field approximation, i.e. neglecting the potential from the remaining ion, gives a shift of $I_p/(3c)$ in the propagation direction [9, 15–20] and a consideration of the radiation pressure gives a shift of $U_p/c$ [3], also in the propagation direction. Note that the Coulomb potential can shift momenta in the direction opposite to that induced by the laser alone [7–9, 21–23]. As discussed, e.g. in reference [14], this Coulomb-induced shift is ellipticity dependent and more pronounced in the case of linearly polarized light than in the case of circularly polarized light.

While the emphasis of the works mentioned above was on nondipole-induced shifts in the short pulse limit based on the strong-field approximation, i.e. neglecting the potential from the remaining ion, this Coulomb-induced shift is ellipticity dependent and more pronounced in the case of linearly polarized light than in the case of circularly polarized light. While the emphasis of the works mentioned above was on nondipole-induced shifts in the short pulse limit, typically the average momentum in the laser propagation direction, nondipole-induced effects on the individual ATI channels have not been considered in detail so far. It is the purpose of this work to provide predictions for the shifts in the long-pulse limit, where the ATI channels can be resolved.

In this manner, we obtain an insight in the importance of nondipole effects on the PMD as a function of the ATI channel, which is a much more differential quantity than the channel-unresolved shifts studied previously. For example we find that the nondipole effects are relatively more important at low than at high ATI energy for strong-field ionization. This contrasts...
the well-known textbook example of single-photon ionization in the perturbative regime, where the electric quadrupole and magnetic dipole effects increase with photon frequency and hence photoelectron energy. In our analysis, we use the strong-field approximation [24] among the Keldysh–Faisal–Reiss approaches [24–26]. Hence, possible shifts associated with the Coulomb potential of the remaining ion are not included in the description. Within this approach, we report nondipole-induced shifts of the center of the energy-conserving rings for a given ATI channel and illustrate redistribution of the continuum population on such a ring due to nondipole terms. This means that the energy of the photoelectron depends on its emission direction in the nondipole case, in contrast to the case of strong-field ionization in the dipole approximation, where the possible energies of a given ATI electron is independent of its propagation direction.

The paper is organized as follows. In section 2, the theory and analytical results are described. In section 3, results are given focusing on the mid-infrared regime. Section 4 concludes.

2. Theory

In this section, we recall the nondipole strong-field-approximation Hamiltonian and the associated Volkov waves [20]. We give expressions for the PMD in the nondipole and dipole approximations. Finally, energy constraints on the final momenta are discussed.

2.1. Nondipole strong-field-approximation Hamiltonian

Our approach was discussed in detail elsewhere [20], and recently applied to the related process of laser-assisted electron scattering [27], so the presentation here is brief. For linearly polarized light of low frequency and high intensity, it is accurate to consider the approximate nondipole strong-field-approximation Hamiltonian for an electron in an electromagnetic field [20]. In this approach, the leading-order nondipole magnetic field effects are included along the dipole-induced motion. The nondipole strong-field-approximation Hamiltonian reads in the velocity gauge (VG) [20]

\[
H_{\text{SFA,VG}}^{\text{ND,VG}} = H_{\text{ND,VG}}^{\text{SFA}} - e V(r).
\]  

(1)

Here \( V(r) \) denotes the single-active-electron potential and

\[
H_{\text{ND,VG}}^{\text{SFA}} = \frac{(p + \mathbf{A}'(t))^2}{2} - e \mathbf{A}(0)(t) \cdot \mathbf{r} - \frac{A^{(0)}(t)^2}{2c} \hat{\mathbf{x}}
\]  

(2)

describes the laser-electron (superscript L-e) interaction, with

\[
\mathbf{A}'(t) = \mathbf{A}^{(0)}(t) \hat{\mathbf{z}} + \frac{\mathbf{A}^{(0)}(t)^2}{2c} \hat{\mathbf{x}}
\]  

(3)

the vector potential with linear polarization along \( \hat{\mathbf{z}} \) and propagation along \( \hat{\mathbf{x}} \). We model \( \mathbf{A}^{(0)}(t) \) by the expression

\[
\mathbf{A}^{(0)}(t) = \hat{\mathbf{z}} A_0 \sin(\omega t).
\]

The subscripts ND and VG in equations (1) and (2) refer to the inclusion of nondipole terms and the usage of the VG, respectively. The superscript SFA refers to the strong-field approximation. The superscript \( (0) \) on the vector potential reminds us that the vector potential only depends on time. We note that the Hamiltonian in equation (2) has been considered in detail in the high-intensity, high-frequency regime [28–35]. In the dipole approximation the term proportional to \( 1/c \) in equation (3) is absent and equation (2) reduces to the dipole Hamiltonian.

2.2. Nondipole strong-field-approximation Volkov wave function

One verifies by substitution that

\[
\psi_{\text{ND,VG}}^{\text{VK}}(r, t) = \frac{1}{(2\pi)^{3/2}} e^{i\mathbf{k} \cdot \mathbf{r} - i\int_0^t dt' \mathbf{A}'(t')^2/2} \psi_{\text{VG}}(r, t)
\]

(4)

is a solution to the time-dependent Schrödinger equation,

\[
i\partial_t \psi_{\text{ND,VG}}^{\text{VK}}(r, t) = H_{\text{SFA,L}}^{\text{ND,VG}}(t) \psi_{\text{ND,VG}}^{\text{VK}}(r, t),
\]

with the Hamiltonian from equation (2). The wave function in equation (4) is the VG nondipole strong-field-approximation Volkov wave function [20]. In the long-pulse limit and to leading-order in \( 1/c \), it may be expressed as

\[
\psi_{\text{ND,VG}}^{\text{VK}}(r, t) = \frac{1}{(2\pi)^{3/2}} \exp(i\mathbf{k} \cdot \mathbf{r}) \times \sum_{n = -\infty}^{\infty} e^{-i(n^2/2 + U_p + \omega_0) \mathbf{r}} J_n(\alpha_0 \cdot \mathbf{k}, \frac{U_p}{\omega_0}).
\]

(5)

Here, we defined a modified ponderomotive potential

\[
U_p' = \left(1 + \frac{k_v}{c}\right) U_p
\]

(6)

with \( U_p = A_0^2/4 \) the ponderomotive potential in the dipole approximation. We note that the energy in equation (6) depends on the direction of asymptotic wavenumber of the electron through its projection on the laser propagation direction, \( k_v \). The implications of such a dependence was discussed in the context of a relativistic treatment in reference [1]. The expression for \( U_p' \) in equation (6) agrees to first order in \( 1/c \) with the result in equation (21) of reference [23]. The dependence on \( k_v \) in equation (6) reflects that the mean energy of the electron moving in the electromagnetic field described by the nondipole strong-field Hamiltonian depends on the magnitude of its momentum in the laser propagation direction due to the magnetic field effects [20]. In equation (5) the quiver radius is defined as \( \alpha_0 = -A_0/\omega \) and the symbol \( J_n(u, v) \) denotes a generalized Bessel function of integer order [24]. The introduction of \( J_n(u, v) \), through their generating function, facilitates the convenient form of the time-dependent phase in equation (5) at the cost of the infinite sum.

2.3. PMD in the long-pulse limit

In the S-matrix formulation [24], the leading-order transition for multiphoton ionization reads in the nondipole case

\[
(S - 1)_{ij} = -i \int_{-\infty}^{\infty} dt \langle \psi_{\text{ND,VG}}^{\text{VK}} | A' \cdot \mathbf{p} + \frac{A^2}{2} | \phi_0 \rangle
\]

(7)
where \(|\psi_{NDVG}^{(k)}\rangle\) is given by equation (5), \(A'\) is defined in equation (3) and \(|\phi_0\rangle\) is the field-free initial state, which includes the phase \(e^{-i\omega t}\) describing the time evolution of the ground state with energy \(E_0 = -I_p\). The superscript B indicates the leading-order Born expression for the \(S\)-matrix. We assume that the pulse is switched on and off adiabatically at very early and late times and performing the time integral in equation (7) results in

\[
(S - 1)^B_{fi} = -2\pi i \sum_n T_f(k_n) \frac{k_n^2}{2} \delta\left(\frac{k_n^2}{2} - n\omega + U'_p + I_p\right),
\]

with an energy conserving delta function giving the final kinetic energy as

\[
\frac{k^2}{2} = n\omega - U'_p - I_p
\]

for \(n > n_0\) and \(n_0\) the smallest number that makes the right-hand side of equation (9) positive. The \(T\)-matrix element in equation (8) to first order in \(1/c\) reads

\[
T_f(k_n) = (-i)^n(U'_p - n\omega)J_{-n}\left(\alpha_0 \cdot k_n, \frac{U'_p}{2\omega}\right) \tilde{\phi}(k_n),
\]

with \(\tilde{\phi}(k_n) = (2\pi)^{-3/2} \int dr \exp(-i k_n \cdot r)\phi_0(r)\) the Fourier transform of the initial orbital evaluated at the momentum \(k_n\). The multiphoton ionization rate is related to the norm square of the transition matrix [44]. Equation (9) gives the energy in the different ATI channels. The energy, \(k^2_n/2\), for a given ATI channel, \(n\), depends on the component of the momentum, \(k_n\), along the laser propagation direction \(\hat{x}\) due to the presence of \(U'_p\) in equation (9). We model the nondipole PMD by the norm square of the \(T\)-matrix element, i.e.

\[
\frac{dP}{dk_n} = \left|(U'_p - n\omega)J_{-n}\left(\alpha_0 \cdot k_n, \frac{U'_p}{2\omega}\right) \tilde{\phi}(k_n)\right|^2.
\]

In the case of the dipole approximation, the corresponding quantity reads

\[
\frac{dP}{dk_{n,D}} = \left|(U_p - n\omega)J_{-n}\left(\alpha_0 \cdot k_{n,D}, \frac{U_p}{2\omega}\right) \phi_0(k_n)\right|^2,
\]

where the subscript \(D\) denotes that equation (12) is only valid in the dipole approximation. We see from equations (11)–(12) that the main differences between PMDs in the nondipole and the dipole approximations are the presence of \(U'_p\) instead of \(U_p\), and \(k_{n,D}\) instead of \(k_{n,D}\). We discuss the differences associated with the magnitudes of the momenta in section 2.4.

In closing this section, we note that the energy conserving relation of equation (9) follows directly from the time integral in the present approach. An alternative perspective that would lead to the same energy conserving delta function would be to consider the condition for constructive interference between electrons ejected in different periods, see, for example, references [36–38].

### 2.4. Constraints on PMD from energy conservation

In the dipole approximation, the magnitude of the final momenta for \(n\)-photon absorption satisfy

\[
\frac{k_{n,D}^2}{2} = n\omega - U_p - I_p.
\]

Equation (13) shows that the possible final momenta form a circle with its center in the origin and radius given by \(k_{n,D} = \sqrt{2(n\omega - U_p - I_p)}\), an ATI ring. If we consider the \((k_x, k_z)\) plane \((k_y = 0)\), where the \(x\) direction is the propagation direction and the \(z\) direction is the polarization direction of the laser, the final momenta read

\[
k_{n,D,x} = k_{n,D} \cos(\theta),
\]

\[
k_{n,D,z} = k_{n,D} \sin(\theta),
\]

where \(\theta\) is measured from the \(x\)-axis.

Equation (9) constrains the final momenta in the nondipole strong-field-approximation approach for the \(n\)th photon absorption channel. We rewrite this expression, specializing to the \((k_x, k_z)\) plane, as

\[
(k_{n,x} + U_p/c)^2 + k_{n,z}^2 = k_{n,D}^2 + (U_p/c)^2.
\]

Equation (16) shows that the final momenta in the nondipole long-pulse limit are confined to a circle with center coordinates

\[
(k_{n,0}, k_{z,0}) = (-U_p/c, 0),
\]

and with a radius

\[
k'_l = \sqrt{k_{n,D}^2 + (U_p/c)^2},
\]

with the superscript \(r\), indicating that \(k'_l\) is a radius of the shifted absorption channel and not a final momentum of the electron. Accordingly, in terms of the overall character of the energy-conserving ATI rings, the nondipole effect shifts the origin of the distribution by an amount \(U_p/c\) opposite the laser propagation direction. As a result the magnitude of the nondipole momentum becomes angle-dependent. To determine the angular dependency one can parameterize the nondipole momenta in the polarization and propagation plane in terms on the angle measured from the origin and with respect to the propagation direction, it can be done by assuming the form

\[
k_{n,x} = k_0 (\theta) \cos(\theta),
\]

\[
k_{n,z} = k_0 (\theta) \sin(\theta),
\]

where \(k_0(\theta)\) is found by assuming the above form of the momenta components and inserting it into equation (16). The angle-dependent magnitude of the momentum is given as

\[
k_0(\theta) = \sqrt{k_{n,D}^2 + (U_p/c)^2}.
\]

Since the applied nondipole strong-field-approximation description provides a nondipole correction to order \(1/c\) [20], the expression for the angle-dependent magnitude of the momentum is only valid to leading order in \(1/c\)

\[
k_0(\theta) = k_{n,D} - U_p \cos(\theta)/c,
\]
with the dipole momentum magnitude $k_{n,D}$ given below equation (13). We see that the shifts in the magnitude of the momenta, as described by equation (21), are consistent with the shift of the origin of the nondipole momentum distributions.

The relative importance of the nondipole shift decreases with the order of the photon-absorption or ATI channel, and hence the energy of the photoelectron. Namely, to leading order in $1/c$ we obtain using equations (21) and (13) that the ratio between the magnitude of the final angle-dependent momentum including nondipole effects, $k_n(\theta)$, and the magnitude of the final angle-independent momentum in the dipole approximation, $k_{n,D}$, is given by

$$\frac{k_n(\theta)}{k_{n,D}} = 1 - \frac{U_p \cos(\theta)}{ck_{n,D}}. \quad (22)$$

For large photon absorption number, $n$, the $1/c$ nondipole correction term decreases with $n$ as $1/\sqrt{n}$.

### 3. Results and discussion

To illustrate the nondipole effects, we consider a laser with a wavelength of 3400 nm and an intensity of $6 \times 10^{13} \text{ W cm}^{-2}$. These values are similar to those used in an experiment [6], where nondipole shifts along the laser propagation direction were reported and are in a regime where the nondipole strong-field-approximation Hamiltonian is accurate [20]. In the experiment a pulsed laser was used and the different photon absorption channels were not resolved. We focus on the long-pulse limit where the individual photon absorption channels can be resolved. The laser is linearly polarized in the $z$ direction and propagates in the $x$ direction. Ground state atomic hydrogen is used as target. All results are obtained from equations (11) and (12) and the energy relations of section 2. These equations show that the main findings reported below regarding nondipole-induced shifts are qualitatively independent of the atomic target. Quantitatively, results for different atoms would differ due to the difference in the value for the Fourier transform of the initial state orbital.

Figure 1 shows the PMD in the plane spanned by the propagation and polarization directions of the laser for the first ATI peak, i.e. for the photon absorption channel with $n = n_0$, see equation (9). Figure 1(a) shows the distribution in the dipole approximation, equation (12), while figure 1(b) shows the distribution obtained with the nondipole approach, equation (11). Finally, figure 1(c) shows the energetically allowed final momenta with the dipole (equations (14) and (15)) and nondipole (equations (19) and (20)) approaches. The intersection of the two full, black lines indicates the center of the first ATI ring in the nondipole strong-field-approximation approach and the intersection of the full, black horizontal line and the dashed, red line indicates the center of the ATI ring in the dipole approximation thus highlighting the shift of the two PMD’s. A comparison between the three panels illustrates the main characteristics of the nondipole effects in the long-pulse limit: a shift of the center of the energy-conserving rings as given by equation (17), i.e. a shift of the center along the negative $k_x$ axis by $U_p/c$ in the nondipole case compared to the dipole case. The other main feature is a nondipole-induced redistribution of the continuum population along the allowed energy-conserving momentum values. This latter feature is highlighted in figure 2, which shows the angular distributions obtained from the momenta in figure 1 for each direction $\theta$ with respect to the laser propagation direction, $x$. The figure gives the results of the nondipole (full, black) and dipole (dashed, red) approaches. The insert is identical to figure 1(c). We see
Figure 2. Angular distribution for the absorption channel $n = n_0 = 216$ corresponding to $n\omega = 1.216U_p$ for the nondipole (full, black curve) and dipole (dashed, red curve) cases. The angle $\theta$ is measured from the $k_x$ axis with respect to the origin. The vertical dashed line marks the polarization direction $\theta = 90^\circ$. The insert shows the possible final momenta for the absorption channel as in figure 1(c).

Figure 3. As figure 2, but for absorption channel $n = n_0 + 83 = 299$, corresponding to $n\omega = 1.683U_p$.

that the angular distribution obtained within the dipole approximation is symmetric about the line of the laser polarization, dashed line at $\theta = 90^\circ$. The nondipole terms break this symmetry. We notice, for example, the nondipole-induced changes to the pair of dipole peaks closest to the polarization direction, i.e. closest to $\theta = 90^\circ$. The nondipole terms induce an increase in the signal in the forward propagation direction ($\theta < 90^\circ$) and a decrease in the signal in the direction opposite to the propagation direction ($\theta > 90^\circ$). In a simple picture, we may think about this change as an effect of the radiation pressure. The cycle-averaged momentum kick in the $x$ direction is obtained from equation (20) of reference [20] and is given by $U_p/e$ (see also references [3, 4] and references therein). A comparison of results like the ones in figures 1 and 2 for increasing values of $n$ (not shown) confirms that the nondipole-induced shifts decrease in relative magnitude for increasing ATI channel, i.e. as the photon absorption channel number $n$ increases. As an example, we show in figure 3 the result for $n = n_0 + 83 = 299$, still within the range of the direct ATI electrons with energy less than $< 2U_p$, but with much smaller nondipole-induced shifts in the peak positions although with some nondipole effects affecting the relative heights of the different peaks. The insert highlights the decrease in relative difference of the possible final nondipole and dipole momenta with increasing photon-absorption number, $n$, by comparing the insert with figure 1(c). Therefore illustrating the decrease of the nondipole shift with increasing $n$ as described by equation (22).

Finally, we illustrate that the ATI-channel-resolved nondipole-induced changes of the photoelectron distributions are also present in spectra that are integrated over ATI channels. Here, as an alternative to the shifting-of-the-peaks effect of figures 2 and 3, which is also present in the total angular distribution, we show the distribution summed over the contributing ATI channels as a function of the momentum $k_x$ in the laser propagation direction. We see from figure 4(a), that the distribution, as expected, is symmetric around $k_x = 0$ in the case of the electric dipole approximation. In the nondipole approach, an asymmetric distribution is predicted as shown in figure 4(b). We observe that the present nondipole approach predicts that most of the strong-field ejected electrons, ionize into states with positive momenta along the laser propagation direction.

It is important to emphasize that the present strong-field-approximation approach neglects the influence of the Coulomb potential of the parent ion. For linear polarization, where the
classical trajectory of the liberated electron may be driven back to the parent ion, this Coulomb effect will shift the low-energy electrons in the direction opposite to what is predicted here [7–9, 14, 21–23]. In the case of circularly polarized light, the rescattering dynamics is less pronounced and the effect of the Coulomb potential of the residual ion is relatively less important. Indeed, after the initial submission of this work, a report on nondipole-induced shifts similar to the ones considered here but for circular polarization appeared [38]. In that work the time-dependent Schrödinger equation was solved for a single-electron model of atomic helium. The authors confirmed the shift of the center of the ATI rings reported here and the associated direction-dependent shifts of the continuum photoelectron energy and relationalized their findings in terms of a strong-field approximation model.

4. Conclusion

In the present work, we considered nondipole effects in the PMD following atomic strong-field ionization by an intense laser field with a wavelength in the mid-infrared regime. We focused on the long-pulse limit, where the individual ATI laser field with a wavelength in the mid-infrared regime. We showed, using the focused ionization of hydrogen atoms and molecules Chin. Phys. B 29 013302

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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