Interplay between pulse parameters and alignment of $\text{H}_2^+$ in strong laser fields

M Abu-samha$^1$ and L B Madsen$^2$

$^1$ College of Sciences and Humanities, Fahad bin Sultan University, Tabuk 71454, KSA
$^2$ Department of Physics and Astronomy, Aarhus University, 8000 Aarhus C, Denmark

E-mail: mabusamha@fbsu.edu.sa

Abstract. We present an \textit{ab initio} study of strong field ionization of aligned $\text{H}_2^+$ by general elliptically polarized laser pulses containing few optical cycles. In particular, we address the effects of laser pulse parameters (light ellipticity and carrier-envelope phase) and molecular alignment on photoelectron momentum distributions (PMDs) of $\text{H}_2^+$.

1. Introduction

Strong field experiments and theory exploit observables such as photoelectron momentum distributions (PMDs), in investigations of atomic-scale electron dynamics, including attosecond streaking [1], strong-field circular dichroism [2,3] and attosecond ionization dynamics or attoclock experiments [4–9].

In strong-field ionization of molecules [10–13], the nature of the probed targets has effects on the PMDs; for example, interferences due to the multicenter character, coupling of electronic and nuclear motions, and multielectron effects. Strong-field studies of $\text{H}_2^+$ using general elliptically polarized laser [14–17] addressed role of the Coulomb potential, coupling of the nuclear and electronic motions, and light-induced subcycle dynamics and how these factors impact above-threshold ionization and PMDs. In Ref. [16], PMDs for $\text{H}_2^+$ and $\text{D}_2^+$ suggest that vibronic coupling is negligible, which validates solution of the time-dependent Schrödinger equation (TDSE) at frozen geometric structure [17]. In Ref. [17], the TDSE calculations were performed in two dimensions and the resulting PMDs revealed an offset angle which is a function of both intensity and wavelength.

In a recent three-dimensional TDSE study of aligned Ar 3p$_x$ and $\text{H}_2^+$ in circularly polarized fields [18], the PMDs corresponding to molecular hydrogen showed a substantial alignment effect due to the anisotropy of the molecular potential and possible coupling to low-lying excited states. In the present work, we expand on the results published in Ref. [18]. Namely, we study interplay between the laser pulse parameters and the molecular alignment, and how it affects PMDs. In particular, we address effects of light ellipticity and carrier-envelope phase on PMDs and lateral PMD (LPMDs) of aligned $\text{H}_2^+$.

2. Computational details

The laser pulse is described by $\vec{F}(t) = -\partial_t \vec{A}(t)$, where $\vec{A}(t)$ is the vector potential, given as $\vec{A}(t) = A_0 f(t) \left( \cos(\omega t + \varphi) \cos(\epsilon/2) \hat{i} + \sin(\omega t + \varphi) \sin(\epsilon/2) \hat{j} \right)$, with $A_0$ the amplitude, $\omega$ the
carrier angular frequency, $\varphi$ the carrier-envelope phase (CEP) value, $\epsilon$ the light ellipticity, and $f(t) = \sin^2(\omega t/2N)$ the envelope for an $N$-cycle pulse. In the present work, $\omega = 0.057$ a.u. corresponding to 800 nm wavelength, and $N = 2$ optical cycles. Atomic units (a.u.) are used.

The TDSE calculations were carried out for $\epsilon = \pi/2$ (corresponding to circular polarization) and for $\epsilon = 1.35$ rad, following the experiment of Ref. [7]. To address the effect of CEP, the calculations were performed for CEP values $\varphi = -\pi/2$ and 0 in the circular polarization limit.

In TDSE calculations, the time-dependent wavefunction is expanded in spherical harmonics for the angular degrees of freedom and the TDSE is solved in the velocity gauge with a grid representation for the reduced radial wave functions [19]. We use an equidistant grid with 4096 points that extends up to 400 a.u. and a time-step of $\Delta t = 0.005$ a.u. in our split-step Crank-Nicholson propagator. PMDs are computed by projecting the wavepacket at the end of the laser pulse on Coulomb scattering states [18, 20].

3. Results and discussion

We explore the PMDs originating from the $\sigma_g$ ground state of the $\text{H}_2^+$ molecule at internuclear separation of 5 a.u., and at alignment angles $\beta = 0^\circ$, 30$^\circ$, 60$^\circ$, and 90$^\circ$. The alignment angle $\beta$ is defined as the angle between the molecular frame and the lab frame $z$-axes. In our TDSE calculations, the molecular axis is defined along the molecular frame $z$-axis, whereas the laser polarization is defined in the $xy$-plane of the lab frame. For $\beta = 0^\circ$, the molecular axis is perpendicular to the plane of laser polarization. For $\beta = 90^\circ$, on the other hand, the molecular axis and the lab frame $x$-axis are aligned.

In figure 1(top panels), we present PMDs for $\text{H}_2^+$ in circularly polarized laser pulses containing 2 optical cycles, with a laser peak intensity 0.2 PW/cm$^2$ and CEP value of $\varphi = -\pi/2$. At $\beta = 0^\circ$, the PMDs are similar to those obtained for atomic targets, in particular, the PMDs show an offset angle relative to the predictions of strong field approximation, in which the molecular potential is neglected [21]. At $\beta = 30^\circ$, new features can be seen in the PMDs. These features evolve with molecular alignment as can be seen for results at $\beta = 60^\circ$ and 90$^\circ$. These results make an interesting comparison to the atomic case, for which the angular distribution of photoelectrons is insensitive to orbital alignment [18].
Figure 2. (a) Lateral PMDs for $H_2^+$ at alignment angle $\beta = 0^\circ$ (solid line), $30^\circ$ (dots), $60^\circ$ (dashed), and $90^\circ$ (crosses). In (b) and (c), we present LPMD for $H_2^+$ at alignment angle $\beta = 30^\circ$ and $\varphi$ values of (b) $\pi/2$ and (c) $\pi/2$.

Next, we investigate effects of CEP on PMDs of $H_2^+$. In figure 1(bottom panels), we present PMDs for $H_2^+$ for $\varphi = 0$, and otherwise same laser parameters as results in figure 1 (top panels). At $\beta = 0^\circ$, changing the CEP results in a simple overall clockwise rotation of the PMD. At $\beta = 30^\circ$, $60^\circ$, and $90^\circ$, changing the CEP leads to different angular distribution of photoelectrons. For alignment angles different from $\beta = 0^\circ$, the molecular potential is strongly anisotropic in the frame of laser polarization. Hence, the PMDs reflect the anisotropic molecular potential felt by the ionized electron.

The results in figure 1 are highly relevant in view of the ongoing discussion on time-delay in tunneling ionization, see e.g., Refs. [4, 5, 22, 23]. In the attoclock scheme, the shift in the peak of the photoelectron momentum distribution compared to that expected from semi-classical modelling has been used to address the question of a possible time-delay [4, 5, 22, 23]. The results in figure 1 show that the momentum distribution is highly affected by the molecular orientation, and hence the short-range part of the molecular potential. For some alignments, we do not even see a clear peak (figure 1, $\beta = 60^\circ$). This shows that not only Coulomb effects and polarization effects as in [7], but also the short-ranged part of the molecular potential, need to be considered in the interpretation of attoclock experiments.

In figure 2(a), we present lateral PMDs (LPMD) for $H_2^+$ for alignment angles $\beta = 0^\circ$, $30^\circ$, $60^\circ$ and $90^\circ$. The lateral PMDs show strong dependence on molecular alignment. For $\beta = 0^\circ$ and $90^\circ$, the lateral PMDs peak about $k_z = 0$, and the LPMD are reflection symmetric across $k_z = 0$. We interpret this feature in light of coupling to excited states, in particular the $\sigma_u$ state. The population of $\sigma_u$ is found to be zero for alignment angles $\beta = 0^\circ$ and $90^\circ$, analysis performed at the end of the laser pulse. At alignment angles of $30^\circ$ and $60^\circ$, however, the lateral PMDs are asymmetric across $k_z = 0$.

From figure 2, we find the LPMD to be particularly interesting for $H_2^+$ at alignment angle $\beta = 30^\circ$. In this case, the lateral PMD shows a bimodal structure along the $k_z$ axis, with larger ionization probability in the positive $k_z$ direction. The asymmetry of ionization probability along $k_z$ is due to interplay between laser carrier envelope phase and anisotropic molecular potential. We have checked that a change in the CEP by $\pi$ shifts the distribution by $180^\circ$ in $\varphi$ as can be seen in figure 2, which leads to a reflection of the distribution with respect to $k_z = 0$, in accordance with the change in the effective molecular potential seen by the outgoing electron upon that CEP change. Simultaneously, at $\beta = 30^\circ$, there is a relatively stronger coupling to the $\sigma_u$ state, and the resulting double-peak structure in the lateral PMD may be consistent with ionization out of this state.

To address the effect of light ellipticity on PMDs, TDSE calculations were carried out for $H_2^+$ in an elliptically polarized laser with light ellipticity $\epsilon = 1.35$ rad. The results are shown in figure 3 for different molecular alignment. One can clearly see that the PMDs show strong dependency on alignment angle, regardless of light ellipticity. This is due to the anisotropic
nature of the $H_2^+$ potential. For $\beta = 0^\circ$, $30^\circ$, and $60^\circ$, changing the ellipticity have minor effects on PMDs. However, at $\beta = 90^\circ$, changing the ellipticity has substantial effect on the PMDs. In this case, the peak maximum shifts by a considerable angle.

4. Conclusions
The present study indicates that it is not straightforward to interpret the effects of laser pulse parameters on PMDs of aligned $H_2^+$, due to the anisotropic nature of the short-range molecular potential and possible coupling to low-lying excited states.

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