Carrier envelope phase dependence of electron localization in the multicycle regime

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Abstract. Thus far, the role of the carrier-envelope phase (CEP) of a light pulse has generally been demonstrated in the few-cycle (sub-10 fs) regime. Here, by simulating the molecular dynamics of H\textsubscript{2}\textsuperscript{+} and D\textsubscript{2}\textsuperscript{+}, we report that the electron localization of H\textsubscript{2}\textsuperscript{+} and D\textsubscript{2}\textsuperscript{+} exhibits unexpectedly strong CEP dependence in the multicycle regime. Our results also demonstrate that the electron localization asymmetry in the multicycle regime (15–20 fs) is higher than that in the single-cycle (4 fs) pulse. This counterintuitive phenomenon is discussed by monitoring the motions of both nuclear and electronic wavepackets.

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The development of the carrier-envelope phase (CEP) stabilization technique has enabled precise control of the waveform of a laser pulse. Using such a laser pulse, the laser–matter interaction process can be controlled by changing CEP. Thus far, various processes, such as high harmonic generation (HHG) \cite{1, 2}, above-threshold ionization (ATI) \cite{3}, double ionization \cite{4–6}, molecular dissociation \cite{7, 8}, laser-induced electron emission \cite{9, 10} and so on \cite{11–13}, have been shown to depend sensitively on CEP in the few-cycle regime (<10 fs for the Ti:sapphire laser). An intuitive explanation for this is that, when the pulse duration is comparable with the optical period, the waveform of the electric field becomes sensitive to CEP. Moreover, since the strong-field effects depend on the instantaneous field strength rather than on the intensity envelope, CEP plays an essential role in these processes. In contrast, on increasing the pulse duration, the CEP dependences of electric field and the nonlinear process become weak and gradually disappear in the multicycle regime. Therefore, the effect of CEP has been generally supposed to be essential only for the few-cycle laser pulse except for its signature in quantum trajectory interference \cite{14} and half-cycle cutoff \cite{15–17} of HHG. Note that some gating schemes of single attosecond pulse generation \cite{18–27} also enable us to observe the CEP dependence effect even though a multicycle pulse is used. However, these gating schemes control the HHG within a single optical cycle. Therefore, the underlying physics is equivalent to the effect of a few-cycle pulse.

In this work, we theoretically demonstrated the CEP dependence effect of the dissociation of molecular hydrogen ions in the multicycle regime. A similar dissociation process has been investigated in the few-cycle regime for hydrogen molecules and its isotopes. It was shown that, using a CEP-stabilized 6 fs laser pulse, the electron can be localized to one specific nucleus of hydrogen (H$_2$) and deuterium (D$_2$) molecules \cite{8, 28}. Numerical simulations have also been performed for HD$^+$ \cite{7, 29, 30}, which also exhibits the CEP dependence of electron localization in the sub-10 fs few-cycle pulse. These investigations have attracted further interest in recent years and different schemes have been proposed and demonstrated for electron localization control \cite{31–39}. The general idea of these schemes is to steer the electronic motion with a waveform-controlled few-cycle pulse \cite{31–33} or with the assistance of another visible \cite{35, 36}, near-infrared \cite{37} or extreme-ultraviolet attosecond pulse \cite{38, 39}. In this work, we investigated electron localization control with a multicycle laser pulse. Intuitively, the waveform of the multicycle pulse (15–20 fs) depends less sensitively on the CEP than that of a few-cycle pulse and CEP is expected to play a minor role in the multicycle regime. However, our simulations based on the solution of the time-dependent Schrödinger equation (TDSE) show an unexpectedly strong CEP dependence of the electron localization of H$_2^+$ in a 20 fs laser pulse.

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Using the quantum simulation model, we compared the electronic and molecular dynamics of $\text{H}_2^+$ in the few-cycle and multicycle regimes and discussed the ionization and dissociation processes. It is found that the counterintuitive strong CEP dependence in the multicycle regime can be attributed to the influence of nuclear motion and the different behavior of each vibrational state. Firstly, the electron is finally localized when the molecule is stretched enough to block the electron exchange between the nuclei. Because nuclear motion is slower than that of the electron, the molecule is stretched at the tail of the multicycle pulse, where CEP affects the waveform of the electric field and the electron localization. Secondly, it is shown that multiple vibrational states are dominant in the few-cycle regime. And since different vibrational states exhibit different CEP dependences, the electron localization asymmetry can be significantly smeared out in the few-cycle pulse after averaging over the vibrational state. In contrast, only one vibrational state is dominant in the multicycle pulse and large electron localization asymmetry remains after averaging.

This work is organized as follows. In section 2, the numerical procedure for solving the TDSE is presented. In section 3, the simulation results of the electron localization probability of $\text{H}_2^+$ are shown for different pulse durations. The differences between the molecular dynamics in the few-cycle and multicycle regimes are discussed. Finally, a brief conclusion is given in section 4.

2. Theoretical model

To simulate the dynamics of $\text{H}_2^+$ subjected to a laser pulse, we numerically solved the three-body TDSE with both electronic and nuclear degrees of freedom included. Note that a full description of this problem requires the solution of a six-dimensional TDSE, which is beyond the current capability. Consequently, the reduced-dimensionality model is usually used [7, 29, 30, 34, 40]. Because the time scale of molecular rotation is several hundred fs, it can be neglected in the ultrashort laser pulse. Therefore, nuclear motion is restricted to the polarization direction if the molecule is properly aligned. Moreover, we assumed that the electron also dominantly moves along the polarized direction. Then we obtained a two-dimensional (one molecular plus one electronic degree of freedom) model. This model agrees reasonably with the three-dimensional model [7], even though some quantitative deviations exist. The two-dimensional TDSE can be written as (in atomic units (au)) [40]

$$i \frac{\partial \Psi(z, R, t)}{\partial t} = [H_R(R) + H_e(z) + V_c(z, R) + V(t)]\Psi(z, R, t),$$

where $z$ and $R$ denote the electronic coordinate and internuclear distance, respectively. The corresponding Hamiltonians are

$$H_e = -\frac{1}{2\mu_e} \frac{\partial^2}{\partial z^2},$$

$$H_R = -\frac{1}{2\mu_R} \frac{\partial^2}{\partial R^2},$$

where $\mu_e = 2m_p/(2m_p + 1)$, $\mu_R = m_p/2$ and $m_p$ is the mass of protons. The Coulomb potential is expressed as

$$V_c(z, R) = -\frac{1}{\sqrt{(z - R/2)^2 + \alpha(R)}} - \frac{1}{\sqrt{(z + R/2)^2 + \alpha(R)}} + \frac{1}{R},$$
In our simulation, propagation of a wavefunction as molecular phenomena in the present context. Processes can be well described, which is sufficient to provide a qualitative understanding of electronic and nuclear motions are included in this model. Hence the ionization and dissociation states are shown in table 1. The Born–Oppenheimer potential of \( \text{H}_2^+ \) can be faithfully reproduced. The interaction with the laser field \( E(t) \) can be written as

\[
V(t) = \left(1 + \frac{1}{2m_p + 1}\right) z E(t). \quad (5)
\]

The electric field is polarized along the \( z \)-direction and can be expressed as \( E(t) = E_0 \exp(-2 \ln(2) t^2 / \tau^2) \cos(\omega t + \delta \phi) \), where \( E_0 \), \( \tau \) and \( \omega \) are the amplitude, pulse duration and angular frequency, respectively. \( \delta \phi \) denotes the CEP of the laser pulse. Note that both the electronic and nuclear motions are included in this model. Hence the ionization and dissociation processes can be well described, which is sufficient to provide a qualitative understanding of molecular phenomena in the present context.

To solve the TDSE, we adopted the operator splitting method, which evaluates the propagation of a wavefunction as [7]

\[
\Psi(t + \delta t) = U(t + \delta t, t) \Psi(t) = U_R \left( \frac{\delta t}{2} \right) \left[ \prod_{i=1}^{N} U_V \left( \frac{\delta t}{2N} \right) U_z \left( \frac{\delta t}{2N} \right) U_V \left( \frac{\delta t}{2N} \right) \right] U_R \left( \frac{\delta t}{2} \right). \quad (6)
\]

In equation (6), \( U_V(\frac{\delta t}{2}) = \exp(-i \delta t (V_z + V(t))/2) \), \( U_R(\frac{\delta t}{2}) = \exp(-i \delta t H_R/2) \simeq (1 + i H_R \delta t/2)^{-1} \times (1 - i H_R \delta t/2) \), \( U_z(\delta t) = \exp(-i \delta t H_z) \) and can be calculated in a similar way. Note that, in this equation, the electronic operator is evaluated \( N \) times more often than the nuclear operator. In our simulation, \( N = 10 \). As shown in [7], such a treatment requires 20–25% fewer operations than the conventional method [41]. We solved the TDSE on grids ranging from 0 to 60 au and from -100 to 100 au for the nuclear and electronic coordinates, respectively. 1024 × 1024 points are adopted and the time step \( \delta t \) is about 0.1 au. The initial states are chosen as the vibrational bound states of \( \text{H}_2^+ \). To obtain the vibrational bound states, we diagonalized the Hamiltonian, which is a large but sparse matrix. The eigenenergies of the 20 lowest vibrational states are shown in table 1. By starting from the initial state, the wavefunction is intergraded in the time range from \(-4 \tau\) to \(4 \tau\). An absorption boundary is applied in the region \( |z| > 90 \text{ au} \), and the ionization probability \( P_i \) is calculated by using the norm of the absorbed wavefunction. As in [7, 41], two dissociation channels can be identified by the regions in configuration space.

| \( v \) | Energy (au) | \( v \) | Energy (au) |
|-----|------------|-----|------------|
| 0   | -0.59995   | 10  | -0.52651   |
| 1   | -0.59106   | 11  | -0.52191   |
| 2   | -0.58175   | 12  | -0.51790   |
| 3   | -0.57257   | 13  | -0.51429   |
| 4   | -0.56504   | 14  | -0.51125   |
| 5   | -0.55732   | 15  | -0.50868   |
| 6   | -0.55008   | 16  | -0.50660   |
| 7   | -0.54336   | 17  | -0.50491   |
| 8   | -0.53717   | 18  | -0.50322   |
| 9   | -0.53163   | 19  | -0.50183   |

where \( \alpha(R) \) is the soft-core parameter. To make the model more realistic, we adjusted this parameter with the internuclear distance so that the 1s\( \sigma \) Born–Oppenheimer potential of \( \text{H}_2^+ \) can be faithfully reproduced.
where the corresponding wavepackets propagate,
\[
P_l : \Omega = | z - R/2 | < 5 \quad \text{and} \quad R > 10, \tag{7}
\]
\[
P_r : \Omega = | z + R/2 | < 5 \quad \text{and} \quad R > 10. \tag{8}
\]

\(P_l\) and \(P_r\) refer to the electron staying in the vicinity of the left and right nuclei, respectively. The dissociation probabilities for these two channels can be calculated as
\[
P = \int_\Omega | \Psi |^2.
\]

Note that the preparation mechanism of a \(H_2^+\) ion beam generally indicates an incoherent Franck–Condon (FC) distribution of vibrational states in experiments [42]. Thus, it is necessary to average the observables over the initial vibrational states weighted by FC factors [29, 43], i.e.
\[
\eta = \sum_v \eta_v F_v, \tag{9}
\]
where \(\eta_v\) denotes the observable calculated for the vibrational state \(v\) and \(F_v\) is the corresponding FC factor (see the inset of figure 5(a)). Then we can evaluate the electron localization asymmetry by defining the parameter
\[
\beta = \frac{P_l - P_r}{P_l + P_r}.
\]

3. Results and discussion

With the above model, we simulated the electron localization of \(H_2^+\) in a laser pulse by varying the duration from 4 to 25 fs. The dashed line and dash-dotted line in figure 1 show the CEP dependence of the localization probabilities around the right \((P_r)\) and left \((P_l)\) nuclei, respectively. The solid line shows the CEP dependence of the electron localization asymmetry \(\beta\). The pulse durations in figures 1(a)–(f) are 4, 8, 10, 15, 20 and 25 fs, respectively. The laser intensity is \(1 \times 10^{14} \text{ W cm}^{-2}\) and the central wavelength is 800 nm. In our calculation, the 20 lowest vibrational states are selected as the initial states and propagated in the laser field independently. FC averaging is performed for these vibrational states using equation (9) to calculate the final electron localization probability. As shown in figure 1, electron localization exhibits a clear CEP dependence for all these pulses. In the few-cycle regime (see figures 1(a) and (b)), the asymmetry of electron localization is only 0.03. On increasing the pulse duration, electron localization still depends sensitively on CEP, and the electron localization asymmetry increases to 0.08 for the 10 and 15 fs pulses. For the 20 fs pulse (see figure 1(e)), the electron localization asymmetry increases to 0.3, which is approximately ten times larger than that in the few-cycle regime. Such a trend is contrary to our intuition, because, compared with that of the few-cycle pulse, the waveform of the multicycle pulse depends less sensitively on CEP and a weaker CEP dependence is expected as in HHG and ATI [1–3]. However, the results in the multicycle regime clearly show a stronger CEP dependence and a larger electron localization asymmetry than those in the few-cycle regime.

To confirm the unexpected strong CEP dependence in the multicycle pulse, we simulated the electron localization at different laser intensities for the 15, 20 and 25 fs pulses. The results are shown in figure 2 for intensities of (a) \(5 \times 10^{13}\), (b) \(8 \times 10^{13}\) and (c) \(2 \times 10^{14} \text{ W cm}^{-2}\). We can see a clear CEP dependence of electron localization for the intensities of \(8 \times 10^{13}\) and \(2 \times 10^{14} \text{ W cm}^{-2}\), where the electron localization asymmetry is still very high (\(\sim 0.1\) and 0.2). At a low intensity \((5 \times 10^{13} \text{ W cm}^{-2})\), the electron localization asymmetry decreases to 0.02 for the 15 and 20 fs pulses and is very low for the 25 fs pulse. Note that the total dissociation probability is also very low at this low intensity. However, an evident CEP dependence can still be observed.
Figure 1. (a) Green dashed and blue dash-dotted lines: probabilities of the electron localized on the right ($P_r$) and left ($P_l$) nuclei as a function of CEP $\phi_0$, respectively. Magenta solid line: electron localization asymmetry defined by $(P_l - P_r)/(P_l + P_r)$. The laser intensity is $1 \times 10^{14}$ W cm$^{-2}$ and the central wavelength is 800 nm. The pulse durations are (a) 4, (b) 8, (c) 10, (d) 15, (e) 20 and (f) 25 fs.

Figure 2. Electron localization asymmetry as a function of CEP for the 15 fs (blue solid line), 20 fs (red crosses) and 25 fs (magenta dashed line) multicycle pulses. The laser intensities are (a) $5 \times 10^{13}$, (b) $8 \times 10^{13}$ and (c) $2 \times 10^{14}$ W cm$^{-2}$.

for the 15 and 20 fs pulses. These results indicate that indeed the strong CEP dependence exists in the multicycle pulse for laser intensities from $5 \times 10^{13}$ to $2 \times 10^{14}$ W cm$^{-2}$.

Next, we presented the results for pulses with different wavelengths of 400 and 1000 nm in figure 3. For the 400 nm pulse, due to the large number of optical cycles and the small ponderomotive energy, the asymmetry becomes relatively weaker than that for the 1000 nm pulse. Nonetheless, the pronounced CEP dependence is still observed for both the 400 and 1000 nm pulses. Furthermore, we also simulated the dissociation and ionization for the isotopic
Figure 3. Electron localization asymmetry as a function of CEP for pulses with different wavelengths of (a) 400 and (b) 1000 nm. The laser intensity is $8 \times 10^{13}$ W cm$^{-2}$ and the pulse duration is 15 fs.

Figure 4. CEP-dependent electron localization asymmetry in the dissociation of $D_2^+$. The laser parameters are the same as in figure 1(d).

molecular ion $D_2^+$ with the same method outlined in section 2. The electron localization asymmetry is shown in figure 4. Compared to the results of $H_2^+$ under the identical laser parameters in figure 1(d), one can see that the asymmetry amplitude of $D_2^+$ is weaker but still evident. These results demonstrate that the CEP dependence in the multicycle regime is a general phenomenon that exists in a wide range of laser intensities and wavelengths and also for its isotope.

To obtain deeper insight into the sensitive CEP dependence in the multicycle pulse, we compared the ionizations and electron localizations of $H_2^+$ in the few-cycle and multicycle regimes. Figure 5(a) shows the probabilities of ionization ($P_i$) and electron localization ($P_l + P_r$) for different vibrational states for the 4 fs pulse. Note that the first vibrational state is $v = 0$. We can see that the contributions of the five lowest vibrational states ($v \leq 4$) to ionization and electron localization are very low and thus can be neglected. The ionization and electron localization probabilities gradually increase for higher vibrational states, reaching about 0.3 when $v = 8$. As described in section 2, the final ionization and electron localization probabilities
should be averaged over all vibrational states. The corresponding weights, i.e. FC factors, are shown in the inset. Figure 5(b) shows the electron localization probability as a function of vibrational state after including the weight of the FC factor. We can see that, besides the five lowest vibrational states, contributions from states higher than the 12th ($v > 11$) are also significantly suppressed. This is due to the small FC factor related to the high vibrational states. Therefore, the vibrational states $v = 5$ and 7–10 dominantly contribute to electron localization. Figure 5(c) shows the CEP dependence of electron localization asymmetry for $v = 5, 7, 8$...
Figure 6. Same as figure 5, but for a pulse duration of 8 fs.

and 9. As shown in this figure, the electron localization asymmetry of each vibrational state exhibits evident CEP dependence, e.g. the asymmetries $\beta$ are approximately 0.04 and 0.08 for $v = 7$ and 8, respectively. However, these state exhibit different behaviors of CEP dependence. For instance, when CEP $\phi_0 = 0.2\pi$, the electron localization asymmetry $\beta < 0$ for $v = 7$, i.e. the electron is dominantly localized at the right nucleus. However, the localization asymmetry $\beta > 0$ for $v = 8$, which is opposite that for $v = 7$. This behavior significantly suppresses the strength of the CEP dependence, thus resulting in a weak CEP dependence of localization asymmetry after FC averaging. A similar situation occurs in the case of the 8 fs pulse. The ionization ($P_i$), electron localization ($P_l + P_r$) and FC-averaged localization probabilities are respectively shown in figures 6(a) and (b) as functions of the vibrational state. Note that only the 15 lowest vibrational states are shown because the contribution of the higher states is too small after FC averaging. We can see that, for the 8 fs laser pulse, the vibrational states $v = 5, 6, 7$ and 8 dominantly contribute to electron localization. The CEP dependences of
Figure 7. Same as figure 5, but for a pulse duration of 15 fs.

electron localization for these states are shown in figure 6(c). Similar to the situation in the 4 fs pulse, these vibrational states exhibit sensitive but different behaviors of CEP dependence. Consequently, the FC-averaged electron localization asymmetry decreases to 0.03 even though each individual vibrational state exhibits a very high asymmetry (e.g. $\beta > 0.2$ for $v = 7, 8$).

Figures 7 and 8 show the ionization and electron localization probabilities for different vibrational states in the cases of 15 and 20 fs pulses, respectively. We can see that the ionization and electron localization probabilities for the four lowest vibrational states are still very low in the multicycle pulse. But for $v \geq 5$, the localization probability decreases and the ionization probability becomes very high due to the low bound energy and long pulse duration. Consequently, the high vibrational states ($v \geq 5$) play an unimportant role in electron localization in the multicycle regime. As shown in figure 8(b), only the fifth vibrational state dominantly contributes to electron localization for the 20 fs pulse. Therefore, the FC averaging does not smear out the CEP dependence effect and a large electron localization asymmetry...
Figure 8. Same as figure 5, but for a pulse duration of 20 fs.

still remains as shown in figure 8(c). The situation in the 15 fs pulse lies between those in the 8 and 20 fs pulses. As shown in figures 7(b) and (c), three vibrational states (v = 4, 5 and 6) are dominant in the 15 fs pulse, and the electron localization asymmetry is larger than 0.25 for all these states. Substantial electron localization asymmetry (≃ 0.1) is still observed after FC averaging.

The results shown in figures 5–8 indicate that the CEP dependences of different vibrational states and their contributions play a crucial role in the electron localization of H₂⁺. The FC averaging effect significantly smears out the CEP dependence of electron localization in the few-cycle regime. On the other hand, one should note that the electron localization asymmetry in the multicycle regime is higher than that in the few-cycle regime. For instance, as shown in figures 8(c) and 5(c), the electron localization asymmetry is as high as approximately 0.6 for v = 4 in the 20 fs pulse case. In contrast, it is only 0.08 for the 4 fs pulse. To understand the underlying physics, we compared the molecular dynamics in the few-cycle
Figure 9. (a) Evolution of the molecular wavepacket $\int dz \left| \Psi(z, R, t) \right|^2$. The blue solid and green dashed lines denote the probabilities of the electron localized on the left and right nuclei, respectively. (b) Evolution of the electronic wavepacket $\int dR \left| \Psi(z, R, t) \right|^2$. The green solid line denotes the vector potential of the driving pulse. The color bar indicates the probability density of the wavepacket on the logarithmic scale. The laser intensity is $1 \times 10^{14}$ W cm$^{-2}$, the pulse duration is 4 fs and the CEP is 0. Panels (c) and (d) are the same as (a) and (b), but for a pulse duration of 20 fs.

and multicycle regimes. Figures 9(a) and (b) show the evolution of the molecular wavepacket $\int dz \left| \Psi(z, R, t) \right|^2$ and the electronic wavepacket $\int dR \left| \Psi(z, R, t) \right|^2$ for the 4 fs laser pulse. The solid and dashed lines in figure 9(a) denote the probabilities of the electron localized on the left and right nuclei, respectively. The solid line in figure 9(b) denotes the vector potential of the driving pulse. Figures 9(c) and (d) are the same as (a) and (b), but for the pulse duration of 20 fs. The laser intensity is $1 \times 10^{14}$ W cm$^{-2}$ and the CEP is 0 in both cases. The initial state is chosen as the most dominant vibrational state, i.e. the ninth and fifth vibrational states for the 4 and 20 fs pulses (see figures 5(b) and 8(b)), respectively. We can see from figure 9(a) that the molecular wavepacket expands very slowly when $t < 2T_0$, where $T_0 = 2.7$ fs is the optical period of the driving pulse. The internuclear distance is less than 5 au, and thus the Coulomb barrier between the two nuclei is very low. Consequently, as shown in figure 9(b), the electron wavepacket motion, which is much faster than the molecular motion because of the smaller mass of electrons, follows the driving pulse and shifts its localization in each half cycle. On increasing the time, the molecular wavepacket gradually expands, and the internuclear distance increases to 6 au at $t = 5–7 T_0$. The internuclear barrier becomes very high, inhibiting the oscillation of
the electron wavepacket [8, 44]. Therefore, we can see two branches of the electron wavepacket when $t > 5 T_0$ (see figure 9(b)). One branch is localized at the left nucleus and the other one is localized at the right nucleus. The corresponding electron localization probabilities $P_l$ and $P_r$ increase rapidly from 5 to $7 T_0$ and gradually saturate at 0.13 and 0.11 (see figure 9(a)). However, as shown in figure 9(b), the laser field is very weak at $t = 5 T_0$ because of the short duration of the few-cycle pulse. The tail field is too weak to efficiently control electron localization. Therefore, the electron localization asymmetry is very low for the 4 fs pulse.

We now turn to the molecular dynamics in the multicycle pulse. As shown in figures 9(c) and (d), dissociation occurs dominantly at $t = 5–7 T_0$ and electron localization shifts between the two nuclei when $t < 6 T_0$, which is similar to that for the 4 fs pulse. From $t = 7 T_0$, the electron localization probabilities $P_l$ and $P_r$ rapidly increase and stabilize at 0.1 and 0.14, respectively, when $t > 12 T_0$. Comparing figures 9(d) with 9(b), we can see that the tail field of the multicycle pulse at $t = 5–7 T_0$ is much higher than that of the few-cycle pulse. For the multicycle pulse, the tail field is still strong enough to efficiently control electron localization. Therefore, a higher localization asymmetry can be obtained.

To gain more detailed insight into the molecular dynamics, we have calculated the kinetic energy release (KER) distribution of the asymmetry. Here, the KER distributions, $S_l(r_k)$, for two dissociation channels are obtained by Fourier analysis of the wavefunctions in the regions defined by equations (7) and (8), respectively. Then the absolute asymmetry as a function of the CEP and the kinetic energy in figure 10 is given by $A(E_k, \phi_0) = S_l(E_k, \phi_0) - S_r(E_k, \phi_0)$. As shown in figure 10, we can clearly see the pronounced CEP dependence of the asymmetry at KERs in the range between 0.5 and 2 eV. As discussed in [8, 28], this KER energy corresponds to the coupling between the 1s$\sigma_g$ and 2p$\sigma_u$ states. Therefore, one can simply understand the above results in terms of the potential curve of the 1s$\sigma_g$ and 2p$\sigma_u$ states. Note that the molecular dynamics of H$_2^+$ and its isotopes have been well explained by this two-level model [8, 28]. Figure 11 shows the potential curve of these two states. Recall that the electronic wavefunction is symmetric and anti-symmetric for the 1s$\sigma_g$ and 2p$\sigma_u$ states, respectively (see the inset of figure 11), and the electron wavepacket that localizes at the left and right nuclei can be defined as $\Psi_{l,r} = (\Psi_{\sigma_g} \pm \Psi_{\sigma_u})/\sqrt{2}$ [8]. In other words, the electron localization asymmetry can be

Figure 10. Absolute asymmetry as a function of the CEP and the kinetic energy for the 15 fs, 800 nm driving pulse with an intensity of $8 \times 10^{13}$ W cm$^{-2}$. 
attributed to the interference between the even and odd parity states [30]. Of course, a high-contrast interference and a large electron localization asymmetry occur under the condition that the probability densities of the 1sσg and 2pσu states are equal. Initially, the wavepacket is on the 1sσg states, so that Ψσu = 0. At small internuclear distances, the energy gap between the 1sσg and 2pσu states is very large (≃0.38 au if R = 2 au), and the wavepacket has to be coupled to the upper states via multiphoton processes (7 photons if R = 2 au). In contrast, the energy gap becomes very small with increasing internuclear distance. Then the required number of photons becomes smaller (e.g. 3 photons if R = 3.7 au), and the coupling between the 1sσg and 2pσu states becomes much easier. When R > 6 au, the wavepacket can be pumped to the upper states by absorbing only one photon. In the few-cycle pulse, the molecular wavepacket moves much more slowly than the decay of the electric field. When the internuclear distance is stretched to 6 au, the electric field already becomes very weak and cannot efficiently pump the wavepacket to the upper state, thus resulting in weak interference and low electron localization asymmetry. In contrast, in the multicycle regime, the leading pulse can stretch the internuclear distance and the tail pulse remains very strong when R > 6 au. Therefore, a substantial wavepacket can be coupled to the 2pσu state, resulting in high-contrast interference and large localization asymmetry.

The above discussions can explain the large localization asymmetry in the multicycle pulse. There remains a question as to why the localization asymmetry depends very sensitively on CEP in the multicycle regime. Intuitively, the electric field of a 20 fs pulse, which is shown in figure 12 for φ0 = 0 and π/2, depends less sensitively on CEP. To explain this question, we consider the change of the electric field when changing CEP. For clarity, we expand the electric fields at the center (t = 0) and tail (t = 7 T0), which are shown in figures 12(b) and (c), respectively. We can see from figure 12(b) that the highest peak P1 changes to P1′ if CEP changes from 0 to π/2; however, the change of the amplitude is very small. We introduce the parameter ξ = (E_{P1} - E_{P1}′)/(E_{P1} + E_{P1}′), where E_{P1} and E_{P1}′ are the amplitudes of P1 and P1′, to...
Figure 12. (a) Electric field of 20 fs light pulses with CEPs of 0 (solid line) and \( \pi/2 \) (dashed line). The laser intensity is \( 1 \times 10^{14} \text{ W cm}^{-2} \). The electric fields at the center and tail are enlarged and shown in (b) and (c), respectively.

evaluate this change. At the center of the 20 fs pulse, \( \xi = 0.00094 \). This means that the change of the electric field is less than 0.1\% if CEP changes from 0 to \( \pi/2 \) at the center of the 20 fs pulse, which is consistent with our intuitive idea. On the other hand, as shown in figure 12(c), the electric field at \( t = 7 T_0 \) exhibits a large change when changing CEP. From the peak values of \( P_2 \) and \( P_2' \), \( \xi \) is calculated to be 0.044. Note that for the 4 fs pulse, \( \xi = 0.019 \) at the center of the laser pulse if CEP changes from 0 to \( \pi/2 \). In other words, the electric field at the tail of the 20 fs pulse depends more sensitively on CEP than that at the center of the single-cycle pulse. Recall that electron localization occurs at the tail of the laser pulse as discussed above. Therefore, the large change of the tail electric field of the multicycle pulse can lead to a large change of electron localization when changing CEP. This explains why an unexpectedly high sensitive CEP dependence is obtained in the multicycle regime.

The rapid development of the CEP stabilization technique has facilitated the experimental observation of the CEP dependence in the multicycle regime. However, it should be noted that the fluctuation and distribution of the laser intensity over the focal volume may frustrate the experiential demonstration. This is because the CEP dependence of electron localization differs at different intensities and the change of laser intensity can smear out the electron localization asymmetry, especially when the \( \text{H}_2^+ \) target beam is larger than the focal spot \([45]\). Recently, Schultze et al \([46]\) have developed a new technique for measuring spatially resolved ion yield. By using this technique, the focal volume effect can be overcome and a high electron localization asymmetry can be detected as shown above. However, in conventional measurement, the ion yield is integrated over the entire focal volume. To investigate this effect,
we averaged the electron localization probability over the intensity distribution as [29]

$$P = \int_{I_m}^{I_0} \frac{P(I)}{I} \, dI,$$

where $P(I)$ is the FC-averaged electron localization probability at the laser intensity $I$. This integration is performed over the intensity from 0.3 to $1.05 \times 10^{14}$ W cm$^{-2}$. We have confirmed that the electron localization probability for $I < 0.3 \times 10^{14}$ W cm$^{-2}$ can be neglected. As shown in figures 5(b) and 8(b), the contributions from $v = 4$ to 8 are dominant. To reduce computational effort, the other vibrational states are not included in the FC averaging. Figure 13 shows the CEP dependence of the localization asymmetry after averaging over laser intensity. We can see that the electron localization asymmetry with an amplitude of 0.03 can still be obtained. This result also indicates that the fluctuation in laser intensity will not prevent us from observing the electron localization asymmetry.

4. Conclusions

In summary, the electron localization of $\text{H}_2^+$ is investigated in the few-cycle and multicycle regimes. By using a numerical model of TDSE, we show that electron localization exhibits an unexpectedly high CEP dependence in the multicycle regime. Also, the localization asymmetry in the multicycle regime is higher than that in the few-cycle regime. Our simulation shows that this electron localization asymmetry is still observable even when including the focal volume effect. By comparing the molecular dynamics in the few-cycle and multicycle regimes, we find that different vibrational states exhibit different CEP dependences of electron localization and that multiple vibrational states are dominant in the few-cycle regime. Therefore, the FC averaging markedly smears out the electron localization asymmetry in the 4 fs pulse. In contrast, only one vibrational state is dominant in the 20 fs pulse and large electron localization asymmetry remains after FC averaging. On the other hand, we find that electron localization occurs at the tail of the laser pulse, which is still very strong in the multicycle regime, hence resulting in effective control of electron localization. However, the tail of the few-cycle pulse
is too weak to efficiently control the electron localization. These two effects induce a larger electron localization asymmetry and a strong CEP dependence in the multicycle regime.

On the other hand, it is worth noting that electron localization control indicates the potential to control the molecular bound and chemical reactions, which has been an exciting goal in chemistry and has attracted much attention in recent years. Most of the previous studies works concentrate on coherent control using a few-cycle pulse [8, 28–31]. Possibly, because of the instinctive idea, coherent control using a multicycle pulse was ignored. Our results indicate that molecular motion also plays an important role in electron localization. Efficient control may require a good balance between the control of electron motion and molecular motion. Note that the nucleus moves more slowly than the electron. Therefore, properly shaped pulses with a long duration (e.g. 20 fs) may be preferable for electron localization control of H₂⁺, H₂ and their isotopes. Recent investigations have also shed more light on this idea [32, 47]. Moreover, our result has indicated that the preparation of the initial state also plays an important role in electron localization. The dissociation from a single vibrational state prevents the effect of FC averaging and can maintain a large electron localization asymmetry. Some methods have been proposed to control the vibrational state of D₂⁺ by applying the laser-induced dipole force at the proper time within one optical period [37, 48, 49]. By combining this technique with the use of a CEP-stabilized ultrashort pulse, efficient control of electron localization can be expected.

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