Phase-locked high-order-harmonic and sub-100-as pulse generation from stretched molecules

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High harmonic generation from diatomic molecules in a linearly polarized intense laser field is investigated and the emission time of the harmonics is discussed with the time-frequency analysis method. It is shown that high harmonic generation from molecules at equilibrium distance is similar to that from atoms. Only the harmonics in the cutoff are synchronized, i.e., well phase-locked, whereas the other harmonics are not phase-locked. For the molecule stretched well beyond its equilibrium distance, the harmonics exhibit distinct time-frequency characteristics. The harmonic spectrum can be extended to \( I_p + 8 U_p \), where \( I_p \) and \( U_p \) are the ionization and ponderomotive potential, and the harmonics with energies below \( I_p + 3.17 U_p \) are not phase-locked and the harmonics with energies beyond \( I_p + 3.17 U_p \) are well phase-locked. Thus a large range of harmonics which are well phase-locked are produced, and a train of clean attosecond (as) pulses with a single 90-as pulse in each half optical cycle can be generated with a multicycle laser pulse. Using a few-cycle laser pulse, an isolated attosecond pulse with a duration of about 95 as is obtained.

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I. INTRODUCTION

Driven by intense laser pulses, atomic and molecular systems can emit lights at frequencies multiple of that of the laser field [1]. This nonlinear process, known as high harmonic generation (HHG), is a subject of great interest [2,3] owing to its potential applications for the coherent extreme ultraviolet (xuv) source [4,5] and the generation of attosecond (as) pulses [6–8]. For atoms, a typical harmonic spectrum shows that the intensity decreases drastically for the first few orders, then remains almost constant for many orders, forming a plateau, and finally drops sharply for the highest orders, which is called the cutoff. This process has been extensively studied [2] and well-understood in terms of the semiclassical three-step model [9] which has been embedded into the quantum model [10]. In the semiclassical description, the electron first tunnels through the potential barrier formed by the combined Coulomb and laser fields, then it oscillates almost freely in the laser field and gains kinetic energy, finally, it can recombine with the parent ion and emit energetic photons. According to this model, the maximum kinetic energy acquired by the electron in the laser field when it recombines with the nucleus is \( 3.17 U_p \), thus the cutoff of the harmonic spectrum occurs at the energy \( I_p + 3.17 U_p \).

The study on HHG from molecules is at the early stage [11–13]. It is found that the HHG from molecules shows the similar spectral pattern to that from atoms. However, molecules have additional degrees of freedom and more complicated symmetries, thus the physical phenomena are much richer. Since the molecule has more than one nuclei, the electron may be detached from one nucleus, then be accelerated by the laser field towards another nucleus, where it is captured and emits an energetic photon. Taking into account this effect, a mechanism to generate harmonics with very high frequencies has been proposed [14]. The numerical results show that the harmonic spectrum can be extended to \( I_p + 8 U_p \) for the molecule with appropriate large internuclear distance [14]. Moreover, by numerically solving the one- and two-dimension time-dependent Schrödinger equations, it is shown that HHG from \( H_2^+ \) and \( H_2 \) exhibits pronounced minima in the harmonic spectra [15]. This phenomenon can be attributed to the interference between the HHG originating from the different nuclei of molecules. Further, it is indicated that the interference effect leads not only to a minimum in the harmonic spectrum, but also to a destruction of even harmonics over a broad bandwidth [12]. Recently, the two-center interference of HHG from \( CO_2 \) and \( O_2 \) has been observed experimentally [16,17] and discussed theoretically [18]. Moreover, it is shown that HHG from molecules depends sensitively on the molecular orientation with respect to the laser polarization direction [19,20]. In addition, the molecular orbit symmetry also plays an important role [13,21], which has been demonstrated by the experimental results [22].

HHG produces a light source covered by a broad bandwidth with equidistant frequencies. The superposition of several harmonics will lead to a train of ultrashort pulses with the duration on the attosecond time scale. This process has a clear analogy with the mode-locked laser. By superposing \( N \) harmonics, the pulse duration can decrease to \( 1/(2N) \) of the driving laser period in the Fourier transform limit [23,24]. Similar to the mode-locked laser, the phase-locking, i.e., synchronization of the harmonics is of utmost importance both from a fundamental point of view as well as for the application of the attosecond pulse generation [25]. The phase-locking means that the phase difference \( \Delta \phi \) between consecutive harmonics is constant, so that the emission time \( t_e = \Delta \phi/\Delta \omega \) [26] is equal for these harmonics, i.e., the harmonics are synchronically emitted. This issue has been discussed

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as early as the production of high harmonics [23,24]. With
the development of the three-step model, the phase-locking
of HHG from atoms has been extensively investigated
[25–29]. It is shown that each harmonic is mainly associated
to two electron trajectories (so-called long and short trajec-
tories) [9,10]. Since the two trajectories have very different
return times, the harmonics are not synchronically emitted,
i.e., are poorly phase-locked. As a result, the superposition of
several harmonics leads to a chaotic attosecond pulse train
(APT) including at least two bursts per half optical cycle of
the driving laser field [27]. Due to the applications for inves-
tigating and controlling ultrafast processes on the atom and
electron levels, the generation of clean APT has always been
of keen interest (see Refs. [6–8] and reviews [2,30,31]). To
obtain a clean APT instead of a chaotic sequence of bursts,
the phase-locked harmonics covered by a broad bandwidth
are required, and then much attention has been paid to the
phase-locking of the harmonics [26–29]. It is demonstrated
that the short trajectory contribution can be macroscopically
selected by carefully adjusting the phase-matching condi-
tion of HHG [27]. When the laser pulse is focused slightly before
the gas jet, the short trajectory is selected. Then the harmon-
cics are emitted almost at the same time, i.e., well phase-
locked, and a clean APT with a single attosecond pulse per
half optical cycle can be generated [27,28]. Even though this
scheme has been verified experimentally [8,26], it is shown
that the phase-locking of the harmonics depends critically on
the experimental conditions and the harmonics are not per-
fectedly phase-locked [26]. So far, the shortest attosecond pulse
generated using the HHG from atoms has a duration of about
200 as in experiments. An intense research is afoot to push
the pulse duration to even shorter time [32].

HHG from molecules also leads to the generation of at-
tosecond pulses [33]. However, the phase-locking of HHG
from molecules has not been thoroughly investigated. In
this paper, we focus on the HHG process of diatomic mole-
cules in a linearly polarized intense laser pulse. Using the
time-frequency analysis method, the emission time of the har-
monics is investigated. It is shown that HHG from molecules
with large internuclear distance exhibits strikingly different
time-frequency characteristics in contrast to that from atoms.
The harmonic spectrum can be extended to $I_p + 8U_p$, and the
emission times of the harmonics with energies beyond $I_p + 3.17U_p$ are
nearly equal, so that a large range of harmonics which are well phase-locked are produced. By superposing these phase-locked harmonics, a clean APT with a single 90-as pulse in each half optical cycle is generated. Further-
more, the HHG process from molecules is investigated with the semiclassical three-step model, and the time-frequency characteristics are interpreted. Finally, it is shown that a single attosecond pulse with a duration of 95 as can be gen-
erated with a few-cycle laser pulse.

II. FORMULATION

In the present work, we consider the interaction of a di-
atomic molecule with a linearly polarized intense laser pulse.
The one-dimensional time-dependent Schrödinger equation
for this system is [in atomic units (a.u.)] [14,15]

\[ i \frac{\partial \psi(x,t)}{\partial t} = \hat{H} \psi(x,t) = \left[ -\frac{1}{2} \frac{\partial^2}{\partial x^2} + V(x,R) - E(t)x \right] \psi(x,t). \]  

(1)

In this equation, single active electron approximation is used
(the so-called $H_n^+$ model [15]), $V(x,R)$ is the Coulomb
potential, and $E(t)$ is the electric field of the laser pulse.
The Coulomb potential for the interaction of electron with the
nuclei is represented by the “soft-core” potential which can be
written as [14,15]

\[ V(x,R) = -\frac{1}{\sqrt{1 + (x - R/2)^2}} - \frac{1}{\sqrt{1 + (x + R/2)^2}}, \]  

(2)

where $R$ is the internuclear distance, and the two nuclei are
fixed at $\pm R/2$, respectively. The laser field polarized in the $\hat{x}$
direction is

\[ \mathbf{E}(t) = E(t) \hat{x} = E_0 \delta(t) \sin(\omega_0 t) \hat{x}, \]  

(3)

where $E_0$, $f(t)$, and $\omega_0$ are the amplitude, envelope, and
angular frequency of the laser pulse, respectively. The time-
dependent Schrödinger equation can be solved with the
Crank-Nicolson method [3], starting from the field-free
ground state which is obtained by propagation in imaginary
time, and finally the wave function $\psi(x,t)$ can be obtained.
The space range is from ~1000 a.u. to 1000 a.u., which is
almost two orders of magnitude higher than the quivering
amplitude of the electron in the laser field. To prevent the
reflection of the wave function from boundary, the wave
function is multiplied by a $\cos^{10}$ mask function that varies
from 1 to 0 over a width of 250 a.u. at the outer boundary.
The spatial and temporal step are 0.1 and 0.07 a.u., respec-
tively. According to the Ehrenfest theorem [34], the time-
dependent dipole acceleration $a(t)$ is

\[ a(t) = -\langle \psi(x,t) | \frac{\partial V(x,R)}{\partial x} - E(t) | \psi(x,t) \rangle. \]  

(4)

The harmonic spectrum can be obtained from the Fourier
transform of $a(t)$

\[ S_q(\omega) \sim |a_q|^2, \]  

(5)

\[ a_q = \int a(t) e^{-iq\omega_0 t} dt, \]  

(6)

where $q$ is the harmonic order. By superposing several har-
monics, an ultrashort pulse is generated and the temporal
shape of this pulse is

\[ I(t) = \left| \sum_q a_q e^{i\omega_0 t} \right|^2. \]  

(7)

III. RESULTS AND DISCUSSIONS

First, we consider the HHG from the molecule at equilib-
rium distance. The harmonic spectrum is shown in Fig. 1(a).
Here the laser intensity is $5 \times 10^{14}$ W/cm$^2$, wavelength is
800 nm, and the ponderomotive potential $U_p = E_0^2/(4\omega_0^2)$ is
1.1 a.u. As in the previous works [12,15], we assume that the laser pulse is turned on and off linearly over three optical cycles, and so for a total of ten optical cycles. The internuclear distance \( R \) is 2 a.u., and the corresponding ionization potential \( I_p \) is calculated to be 1.25 a.u. As shown in Fig. 1(a), the harmonic intensity decreases for the first five harmonics, then remains almost constant for many harmonics, and finally drops sharply from the 83rd harmonic. Moreover, it is shown that the even order harmonics are suppressed and a minimum occurs at the 23rd harmonic. All these results are consistent with the previous ones [12,15], which can be attributed to the interference between the HHG from the two different nuclei of the molecule. To further understand the HHG process, we investigate the emission time of the harmonics with the time-frequency analysis method [35], and the results are shown in Fig. 1(b). As shown in this figure, there are two dominant quantum trajectories (so-called short and long trajectories) with different emission times contributing to the same harmonic in each half optical cycle. As increasing the harmonic order, the emission time for the short trajectory increases, that for the long trajectory decreases, and the emission time is almost equal for the harmonics in the cutoff. Thus for the harmonics in the plateau, the emission times of these two trajectories are much different, and the harmonics are not synchronized, i.e., poorly phase-locked. For the harmonics in the cutoff, the short and long trajectories are superposed, and the harmonics are synchronized, i.e., phase-locked. All these characteristics are similar to those of the HHG from atoms.

Following, we consider the attosecond pulse generation from the molecule at equilibrium distance. The temporal shape of the attosecond pulses generated by superposing five harmonics in the plateau is shown in Fig. 2(a). The parameters are the same as those in Fig. 1. As shown in Fig. 2(a) two dominant pulses corresponding to the short and long quantum trajectories are generated in each half optical cycle. Since the harmonics are poorly phase-locked, the APT will be more chaotic if more harmonics are superposed. This poor quality of the APT limits its application. Figure 2(b) shows the temporal shape of the attosecond pulses generated by superposing five harmonics in the cutoff. It can be seen that the harmonics in the cutoff are well phase-locked, then a clean APT with a single pulse in each half optical cycle is generated. The duration of the attosecond pulse is 340 as, which is greater than the calculated result 267 as in the Fourier transform limit. It is because the harmonic intensity decreases drastically in the cutoff, so that only one or two harmonics effectively contribute to the attosecond pulse, thus the phase-locking of the harmonics in the cutoff is not of much interest [27].

Nowadays, the molecule can be stretched well beyond the equilibrium distance [36], and the HHG from stretched mol-
fascinating result shown in Fig. 3 to the HHG from the molecule at equilibrium distance, a non-phonically emitted, i.e., not phase-locked. However, in contrast to the emission time of the harmonics. Figure 3 illustrates the harmonic spectrum of the HHG from the stretched molecule. The laser intensity is $2.5 \times 10^{14}$ W/cm$^2$, and the ionization potential $I_p$ is calculated to be 0.68 a.u. In contrast to Fig. 1(a), the HHG from the stretched molecule exhibits two cutoffs in the spectrum, one at the 43rd harmonic, corresponding to the energy $I_p + 3.17U_p$, and the other one at the 91st harmonic, corresponding to the energy $I_p + 8U_p$. These results are consistent with the previous ones [14]. Further, we investigate the emission time of the harmonics. Figure 3(b) illustrates the time-frequency characteristics of the HHG from the stretched molecule. It can be seen from this figure that the harmonics with energies below $I_p + 3.17U_p$ are not synchronically emitted, i.e., not phase-locked. However, in contrast to the HHG from the molecule at equilibrium distance, a fascinating result shown in Fig. 3(b) is that there is only one dominant quantum trajectory contributing to the harmonics with energies beyond $I_p + 3.17U_p$ in each half optical cycle. As a result, the harmonics with energies beyond $I_p + 3.17U_p$ are emitted at almost the same time, so that these harmonics are well phase-locked. In addition, these phase-locked harmonics cover a broad bandwidth, which is beneficial for the generation of ultrashort attosecond pulses.

The temporal shape of the attosecond pulse generation from the molecule stretched well beyond its equilibrium distance is shown in Fig. 4. The parameters are the same with those in Fig. 3. Figure 4(a) illustrates the APT generated by superposing five harmonics with energies below $I_p + 3.17U_p$. As shown in this figure, these harmonics are poorly phase-locked and so a chaotic APT is generated. The solid line in Fig. 4(b) illustrates the APT generated by superposing five harmonics with energies beyond $I_p + 3.17U_p$. In contrast to the results shown in Fig. 4(a), a clean APT with a single attosecond pulse in each half optical cycle is obtained as shown in Fig. 4(b). The duration of each attosecond pulse is about 270 as, which is close to the pulse duration in the Fourier transform limit. Further, from Fig. 3(b), we can see a fascinating characteristic that the harmonics with energies beyond $I_p + 3.17U_p$ are well phase-locked. These phase-locked harmonics cover a bandwidth greater than 40 eV, implying the generation of attosecond pulse with a duration less than 100 as. This is clearly demonstrated by the results shown in Fig. 4(b), in which the dashed line shows the temporal shape of the attosecond pulses generated by superposing 15 harmonics with energies beyond $I_p + 3.17U_p$. As shown in this figure, a clean APT with a single attosecond pulse in each half optical cycle is obtained, and the pulse duration is reduced to about 90 as. Note that these results survive for a range of internuclear distances. Sub-100 attosecond pulses can be produced from the molecule with an internuclear distance in the range from 70 to 100 a.u.
FIG. 5. The dependence of the degree of phase-locking $\tilde{\eta}$ on the harmonic order for the molecule with small ($R=2$ a.u.) and large ($R=80$ a.u.) internuclear distance, respectively.

To systematically investigate the temporal distortion and phase-locking of the APT, we introduce the parameter $\eta$ to measure the degree of phase-locking which is defined by [26,28]

$$\eta = \frac{\int_{\tau_r}^{\bar{\tau}_r} dt I(t)}{\int_{\tau_r/2}^{\tau_r/2} dt I(t)},$$

where $I(t)$ is the intensity of the attosecond pulse, and $\tau_r = T_0/(2N)$. We then normalize $\eta$ to $\bar{\eta} = (\eta - 2\tau_r/T_0)/(0.775 - 2\tau_r/T_0)$, where 0.775 is the ratio obtained in the case of perfect phase-locking, i.e., Fourier transform limit. $\bar{\eta}$ measures the proportion of the xuv energy emitted within $\tau_r$ during a half optical cycle. It means that if the harmonics are perfectly phase-locked $\bar{\eta} = 1$, and $\bar{\eta} = 0$ if $I(t)$ is a constant. The dependence of the degree of phase-locking $\bar{\eta}$ on the harmonic order is shown in Fig. 5. The closed and open circles correspond to the HHG from the molecules with small (the parameters are the same as those in Fig. 1) and large (the parameters are the same as those in Fig. 3) internuclear distances. For the molecule at equilibrium distance, it can be clearly seen that the degree of phase-locking $\bar{\eta}$ is less than 0.5 for the harmonics below $80\omega_0$ and increases rapidly to 0.7 for the higher order harmonics. This result indicates that only the harmonics in the cutoff are well phase-locked and the other harmonics are poorly phase-locked. The situation is strikingly different for the stretched molecule. As show in Fig. 5, the degree of phase-locking $\tilde{\eta}$ is less than 0.5 for the harmonics below $45\omega_0$ (close to the energy $I_p + 3.17U_p$) and increases rapidly to 0.7 for the harmonics greater than $45\omega_0$. Thus a large range of harmonics with energies beyond $I_p + 3.17U_p$ are well phase-locked. All these characteristics are consistent with those shown in Fig. 3. Note that $\bar{\eta}$ does not reach exactly to 1. This means that the harmonics are not perfectly phase-locked, which also can be seen from Fig. 3(b).

As shown in Fig. 3(b), the emission times of the harmonics with energies beyond $I_p + 3.17U_p$ are not exactly equal. Thus an intrinsic chirp is existing for these harmonics. With proper material, the intrinsic chirp can be complemented, so that the attosecond pulse duration can be decreased to the Fourier transform limit [26,32].

To understand the underlying physics of the time-frequency characteristics shown in the above figures, we discuss the HHG process in molecules using the semiclassical three step model. For the diatomic molecule, there are two approaches for HHG. One is that the electron recombines with its parent nucleus where it is ionized, and the other one is that the electron is ionized from one nucleus but recombines with the neighboring nucleus. We can use the semiclassical three step model [9] to discuss these processes. Simply, we only investigate the electron motion in a half optical cycle. The electron motion is described by the equation

$$\ddot{x}(t) = -E_0 \sin(\omega_0 t).$$

Assuming that the electron is ionized from the nucleus with zero velocity at the time $t_i$, we have

$$\dot{x}(t) = \frac{E_0}{\omega_0}(\cos \omega_0 t - \cos \omega_0 t_i).$$

For the first approach, the electron recombines with the parent nucleus, and the recombination condition reads

$$x(t) = \frac{E_0}{\omega_0}[\sin \omega_0 t - \sin \omega_0 t_i - \omega_0(t - t_i)\cos \omega_0 t_i] = 0,$$

where $E_0/\omega_0^2$ is the quivering amplitude of the electron in the laser field. This recombination condition is the same with that of HHG from atoms. From Eq. (11), the recombination time which corresponds to the emission time of the harmonics can be solved. For the second approach, we assume that the electron is ionized from the nucleus fixed at $-R/2$, and recombines with the nucleus fixed at $R/2$, then the recombination condition reads

$$x(t) = \frac{E_0}{\omega_0}[\sin \omega_0 t - \sin \omega_0 t_i - \omega_0(t - t_i)\cos \omega_0 t_i] - R = 0.$$

Moreover, there is existing another process that the electron is ionized from the nucleus fixed at $R/2$ and recombines with the nucleus fixed at $-R/2$. This process is similar to the former one, and is not discussed here. The electron energy when it recombines with the nucleus is $E_k = x^2/2$. Combining Eqs. (10) and (12), it can be shown that the electron kinetic energy $E_k$ can reach $8U_p$ when $R = m\pi E_0/(\omega_0^2)$ with $m$ being an odd integer [14]. However, for molecules at equilibrium distance, the typical internuclear distance is only several atom units, e.g., 2 a.u. in our case, which can be neglected in contrast to the electron quivering amplitude. Thus Eq. (12) can be rewritten as
which leads to the similar recombination condition to Eq. (11). Therefore the above two approaches for HHG from the molecule at equilibrium distance exhibit similar characteristics to that from atoms, which is consistent with the previous discussion [14]. Combining Eqs. (10) and (11), the dependence of $E_k$ on the ionization time $t_i$ and emission time $t_e$ for the HHG from the molecule at equilibrium distance can be obtained, which is illustrated in Fig. 6(a). As shown in this figure, there are two ionization and emission times corresponding to the same kinetic energy. The first couple with earlier ionization but later emission times correspond to the long trajectory, and the other couple with later ionization but earlier emission times correspond to the short trajectory. Increasing the kinetic energy, the emission time for the short trajectory increases, that for the long trajectory decreases, and at last these two emission times become equal. This result indicates that the harmonics in the plateau are not synchronically emitted, i.e., poorly phase-locked, and the harmonics in the cutoff are synchronized. It is consistent with the time-frequency characteristics shown in Fig. 1. For the molecule stretched well beyond its equilibrium distance, the internuclear distance is comparable to the electron quivering amplitude. Then the recombination conditions are much different for the above two approaches, and the harmonics will exhibit strikingly different characteristics. The first approach is the same with that of atoms, which has been illustrated in Fig. 6(a). Following, we focus on the HHG in the second approach. The dependence of $E_k$ on the ionization time $t_i$ and emission time $t_e$ can be obtained from Eqs. (10) and (12), which is shown in Fig. 6(b). The internuclear distance $R$ is 80 a.u. ($=\pi E_0/\omega_0^2$) and the laser parameters are the same with those in Fig. 3. As shown in this figure, the maximum kinetic energy reaches to $8U_p$, thus the harmonic spectrum is extended to $I_p + 8U_p$ as shown in Fig. 3. Moreover, it is shown in Fig. 6(b) that there are two ionization and emission times corresponding to the same kinetic energy. Analogically, we call them the short and long trajectories. However, the emission times of these two trajectories are nearly equal. In other words, the harmonics are emitted almost at the same time, i.e., are well-synchronized. Taking into account the above two approaches, we can conclude that the HHG from the stretched molecule can be extended to $I_p + 8U_p$. The harmonics with energies below $I_p + 3.17U_p$ are not synchronized, and the harmonics with energies beyond $I_p + 3.17U_p$ are well-synchronized. In addition, it is shown in Fig. 6(b) that the emission times for the different kinetic energies are not exactly equal. This means that the harmonics exhibit an intrinsic chirp. These results interpret the time-frequency characteristics shown in Figs. 1–5 from the semiclassical point of view.

The generation of an isolated attosecond pulse has also been of great interest. Irradiating the gas of krypton with a few-cycle laser pulse, an isolated pulse with a duration of 650 occurred as was observed for the first time by Hentschel et al. [6]. In contrast to the HHG from atoms, HHG from stretched molecules covers a broader bandwidth, and the superposition of these harmonics can lead to an attosecond pulse with shorter duration. Figure 7(a) shows the spectrum of the HHG from a molecule stretched well beyond the equilibrium distance. The internuclear distance $R$ is 100 a.u., and the laser intensity is $5 \times 10^{14}$ W/cm². A sin² shaped pulse is adopted, and then the electric field is expressed $E(t) = E_0 \sin(\pi t / T_p) \cos[\omega_0(t - T/2)]$ where $T = 5 T_0$, the corresponding duration is about 5 fs full width at half maximum. As shown in Fig. 7(a), the harmonic spectrum is extended to $165 \omega_0$, which is close to the energy $I_p + 8U_p$. Figure 7(b) illustrates the time-frequency characteristics of the harmonics shown in Fig. 7(a). As shown in Fig. 7(b), the harmonics below $80 \omega_0$ (close to the energy $I_p + 3.17U_p$) are not synchronized, and the other harmonics beyond $80 \omega_0$ are well-synchronized. All these characteristics are similar to those in the multicycle pulse case. Moreover, the harmonics greater than $150 \omega_0$ are only emitted near $t = 2.5 T_0$ when the laser intensity reaches its maximum. While at other time, the laser intensity of the few-cycle pulse decreases rapidly, and then the corresponding harmonic energy is less than $150 \omega_0$. As a result, there is only one quantum trajectory contributing to the harmonics at the end of the plateau in the few-cycle laser pulse case. Thus these harmonics are continuous as shown in

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**FIG. 6.** The dependence of kinetic energy $E_k$ on the ionization time $t_i$ and emission time $t_e$ in the case (a) the electron recombines with its parent nucleus where it is ionized and (b) the electron is ionized from one nucleus and recombines with the neighboring nucleus. The parameters are the same as those in Fig. 3.

\[
x(t) = \frac{E_0}{\omega_0^2} [\sin \omega_0 t - \sin \omega_0 t_i - \omega_0(t - t_i) \cos \omega_0 t_i] - R = \frac{E_0}{\omega_0^2} [\sin \omega_0 t - \sin \omega_0 t_i - \omega_0(t - t_i) \cos \omega_0 t_i] = 0,
\]

(13)
the intensity of the two satellite pulses is so low that they can be ignored. Note that all the above results are a single molecule effect. Propagation effect may play an important influence on the harmonics. At appropriate conditions, the electron trajectories might be sifted by propagation effects, so that the time confinement of attosecond pulses can be improved. These effects will be investigated in detail in the future.

IV. CONCLUSIONS

HHG from diatomic molecules in a linearly polarized intense laser field is investigated and the time-frequency characteristics of the harmonics are discussed. It is shown that there are two approaches for HHG from diatomic molecules. One is that the electron recombines with its parent nucleus where it is ionized, and the other is that the electron is ionized from one nucleus but recombines with the neighboring nucleus. For the molecule at equilibrium distance, these two approaches exhibit similar characteristics as those from atoms. Only the harmonics in the cutoff are phase-locked, and there are two dominant electron trajectories with much different emission times in each half optical cycle contributing to the harmonics in the plateau, then these harmonics are not synchronized, i.e., poorly phase-locked. For the molecule stretched well beyond the equilibrium distance, these two approaches exhibit strikingly different characteristics. The HHG by the first approach is the same with that from atoms. For the second approach, the harmonic spectrum can be extended to $I_p + 8U_p$, and the fascinating characteristic is that the emission time is nearly equal, then the harmonics are well-synchronized, i.e., phase-locked. Thus for the HHG from stretched molecules, the harmonics with energies below $I_p + 3.17U_p$ are not phase-locked, and the harmonics with energies beyond $I_p + 3.17U_p$ are well phase-locked. As a result, a large range of harmonics which are well phase-locked are produced, and a clean APT with a single 90-as pulse in each half optical cycle is obtained with a multicycle laser pulse. Furthermore, the results indicate that an isolated attosecond pulse with the duration of 95 as can be generated with a few-cycle laser pulse.

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