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Probing the structure of multi-center molecules with odd–even high harmonics*

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We study high-order harmonic generation (HHG) from multi-center asymmetric linear molecules numerically and analytically. Our simulations show that odd and even HHG spectra of the asymmetric multi-center system respond differently to the change of the molecular structure. Specifically, when the internuclear distances between these nuclei of the molecule have a small change, the odd spectra usually do not change basically, but the even spectra differ remarkably. Based on this phenomenon, a simple procedure is proposed to probe the positions of these nuclei with odd–even HHG. Our results shed light on attosecond probing of the structure of multi-center molecules using HHG.

Keywords: odd–even harmonics, multi-center asymmetric molecules, molecular structure

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1. Introduction

In the past two decades, high-order harmonic generation (HHG)[1,2] from atoms and molecules has been a hot subject in both experimental and theoretical studies of strong laser–matter interaction. This interest in HHG partly arises from the important applications of HHG in attosecond science,[3,4] including attosecond probing of the electron structure and dynamics.[5,6] The HHG process can be well understood by a semiclassical three-step model:[7] (i) ionization of the active electron by tunneling, (ii) propagation of the electron in the laser field, and (iii) recombination of the freed electron into the bound state with the emission of a high-energy photon.

Previous studies on HHG are mainly focused on atoms and diatomic molecules. For symmetric diatomic molecules, such as H_2^+ [8–11] and N_2,[12–15] due to the central symmetry of the potential, only odd harmonics are emitted. The phenomenon is similar to the atomic case. In particular, the HHG spectrum of symmetric diatomic molecules also shows a striking minimum, which is absent for the atomic spectrum and has been identified as arising from the effect of two-center interference when the rescattering electron returns to and recombines with these two atomic centers of the molecule.[8] This minimum has attracted great interest in recent years.[16–24] It can be used to probe the internuclear distance of the diatomic molecule[25–28] and judge the sign of the dipole phase in the molecular orbital tomography procedure.[12] For asymmetric diatomic molecules, such as HeH^{2+},[29–38] CO,[39–43] and NO,[44] due to symmetry breaking, both odd and even harmonics are emitted. It has been shown that these odd and even harmonics possess different spectral properties[45–48] and carry different information of the target,[31,49–51] and therefore need to be studied separately.

The odd–even HHG from asymmetric diatomic molecules has shown promising applications in ultrafast measurements. For instance, using odd–even HHG, one can probe the electron dynamics of asymmetric molecules,[52–56] image the asymmetric orbital,[43] and probe the nuclear dynamics of asymmetric molecules.[57] The degree of orientation for asymmetric molecules can also be calibrated with odd–even HHG.[58,59]

Recently, attention is increasingly turning to the interaction of the laser field with polyatomic molecules (such as CH_4,[60–62] CH_2Cl_2,[63] C_{60},[64] and SF_6[65]) and solids.[66–72] For symmetric polyatomic molecules such as CO_2, the orbital symmetries can be complex with multiple nodal planes, and multielectron dynamics can become significant with several orbitals close to the highest occupied molecular orbital.[27] For asymmetric polyatomic molecules such as HCCl,[73] OCS,[50] and N_2O,[74] due to symmetry breaking, the situation is more complex and some new effects can emerge. To probe the structure of multi-center asymmetric molecules with odd–even HHG, a detailed theoretical study on this issue is needed.

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In this paper, we study odd–even HHG from multi-center asymmetric linear molecules H_3^{2+} and H_4^{3+} with diverse internuclear distances at different orientation angles numerically and analytically. The time-dependent Schrödinger equation (TDSE) of the simple single-electron systems can be solved exactly at present. Our simulations show that the emission of odd and even harmonics from these asymmetric systems depends differently on the change of the internuclear distance. Specifically, as this distance between two neighboring nuclei has a small change, odd spectra do not change usually, while even spectra differ remarkably. As a result, the positions of interference minima in odd spectra for the multi-center systems are also insensitive to the internuclear distance, different from two-center cases. We show that the phenomena are associated with different HHG mechanisms of odd versus even harmonics. With the sensitivity of even HHG spectra to the internuclear distance, a procedure is proposed to probe this distance using the relative yields of odd versus even harmonics.

2. Theoretical methods

2.1. Numerical procedure

The Hamiltonian of the molecular system studied here is $H(t) = \boldsymbol{p}^2/2 + V(\boldsymbol{r}) + \boldsymbol{r}\cdot\boldsymbol{E}(t)$ (in atomic units of $\hbar = e = m_e = 1$). We assume the molecular axis is located in the *xoy* plane and the laser field $\boldsymbol{E}(t)$ is linearly polarized along a direction parallel to the x axis. For linear molecules H_3^{2+} and H_4^{3+}, the soft-core potential $V(\boldsymbol{r})$ used here has the following form: $V(\boldsymbol{r}) = -Z/\sqrt{\xi+(\boldsymbol{r}-\boldsymbol{R}_1)^2} - Z/\sqrt{\xi+(\boldsymbol{r}-\boldsymbol{R}_2)^2} - Z/\sqrt{\xi+(\boldsymbol{r}-\boldsymbol{R}_3)^2}$ for H_3^{2+} and $V(\boldsymbol{r}) = -Z/\sqrt{\xi+(\boldsymbol{r}-\boldsymbol{R}_1)^2} - Z/\sqrt{\xi+(\boldsymbol{r}-\boldsymbol{R}_2)^2} - Z/\sqrt{\xi+(\boldsymbol{r}-\boldsymbol{R}_3)^2} - Z/\sqrt{\xi+(\boldsymbol{r}-\boldsymbol{R}_4)^2}$ for H_4^{3+}. Here $\boldsymbol{R}_1$, $\boldsymbol{R}_2$, $\boldsymbol{R}_3$, and $\boldsymbol{R}_4$ are the vectors between the origin and different nucleus, and Z is the effective charge which is adjusted to obtain the desired ground-state energy. Similarly, in our comparison studies, the soft-core potential for diatomic molecules H_2^+ has the following form of $V(\boldsymbol{r}) = -Z/\sqrt{\xi+(\boldsymbol{r}-\boldsymbol{R}/2)^2} - Z/\sqrt{\xi+(\boldsymbol{r}+\boldsymbol{R}/2)^2}$. Here, $\boldsymbol{R}$ is the vector between these two nuclei of H_2^+. ξ is the smoothing parameter which is used to avoid the Coulomb singularity. For comparison, we have used the value of $\xi = 0.5$ for all cases of multi-center molecules explored in the paper. The main results associated with properties of odd–even HHG from multi-center asymmetric molecules discussed in the paper are not sensitive to the choice of the parameter ξ. $\boldsymbol{E}(t) = \boldsymbol{e}_x E(t)$ with $E(t) = f(t)E\sin\omega_0 t$ is the external electric field, and $\boldsymbol{e}_x$ is the unit vector along the x axis. $f(t)$ is the envelope function. E and ω_0 are the amplitude and the frequency of the external electric field, respectively. In our calculations, we use a ten-cycle laser pulse which is linearly ramped up for three optical cycles and then kept at a constant intensity for four additional cycles and finally linearly ramped down for three optical cycles. Numerically, the TDSE of $i\dot{\psi}(t) = H(t)\psi(t)$ is solved numerically using the spectral method.[75] We work with a grid size of $L_x \times L_y = 409.6 \times 102.4$ a.u. for the x and y axes, respectively. Details of our TDSE calculations can be seen in Ref. [76].

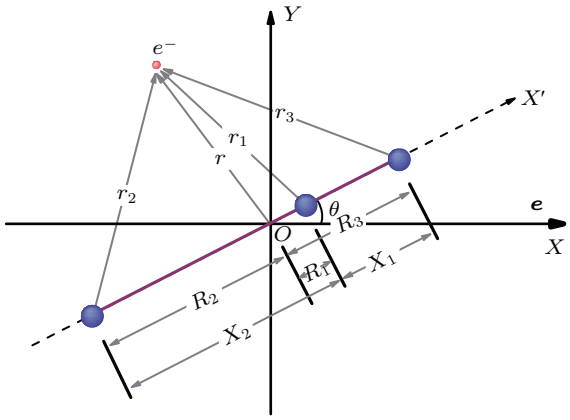


Fig. 1. (color online) A sketch of the molecular geometry and the coordinate system used in the 2D simulations for H_3^{2+}. The polarization direction $\boldsymbol{e}$ of the laser field is along the x axis. The molecular axis x' is located in the *xoy* plane with an angle θ to the x axis and the center of mass of the molecular system agrees with the origin of the coordinate system o. $\boldsymbol{R}_1$, $\boldsymbol{R}_2$, and $\boldsymbol{R}_3$ denote the positions of these three nuclei of H_3^{2+} to the origin, respectively. $\boldsymbol{r}_1$, $\boldsymbol{r}_2$, and $\boldsymbol{r}_3$ denote the positions of the electron to these three nuclei and $\boldsymbol{r}$ denotes that to the origin. X_1 and X_2 are the internuclear distances between two adjacent nuclei.

The sketches of the coordinate systems of H_3^{2+} and H_4^{3+} used in our two-dimensional (2D) simulations are presented in Figs. 1 and 2. Once the TDSE wave function $\psi(t)$ is obtained, the coherent part of the HHG spectrum parallel to the laser polarization $\boldsymbol{e}$ can be evaluated using

$$F(\omega) = \int \langle\psi(t)|\boldsymbol{e}\cdot\nabla V|\psi(t)\rangle\, e^{i\omega t}\, dt, \tag{1}$$

where ω is the emitted-proton frequency.

For comparison, the ground states of the linear molecules with different molecular parameters studied here have the same ionization potential of $I_p = 1.52$ a.u.

2.2. Analytical description

In Ref. [31], it has been shown that for asymmetric diatomic molecules, there is a close relation between odd–even HHG spectra and odd–even bound-continuum-transition dipoles. This relation reflects the main HHG route for the emission of one odd or even harmonic from asymmetric molecules.[43] The main odd (even) HHG route is associated with the rescattering electron ionizing from the gerade component of the asymmetric orbital and recombining with the gerade (ungerade) component of the asymmetric orbital. A detailed discussion on the odd–even HHG routes can be found in Refs. [32] and [54]. We now explore the relation for multi-center cases. For comparison, the diatomic cases are also discussed here.

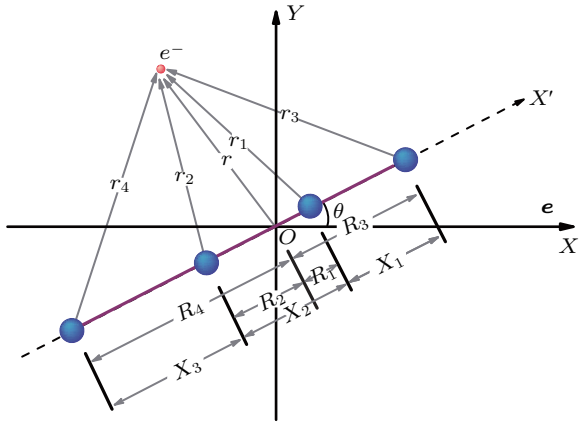


Fig. 2. (color online) Same as Fig. 1 but for H_4^{3+}. $\boldsymbol{R}_1$, $\boldsymbol{R}_2$, $\boldsymbol{R}_3$, and $\boldsymbol{R}_4$ denote the positions of these four nuclei of H_4^{3+} to the origin, respectively. $\boldsymbol{r}_1$, $\boldsymbol{r}_2$, $\boldsymbol{r}_3$, and $\boldsymbol{r}_4$ denote the positions of the electron to these four nuclei and $\boldsymbol{r}$ denotes that to the origin. X_1, X_2, and X_3 are the internuclear distances between two adjacent nuclei.

We assume that the wave functions of the ground states $|0\rangle$ for H_2^+, H_3^{2+} and H_4^{3+} molecules have the forms of $\phi_0^2(\boldsymbol{r}) = N_f(e^{-\kappa r_1} + e^{-\kappa r_2})$, $\phi_0^3(\boldsymbol{r}) = N_f(e^{-\kappa r_1} + e^{-\kappa r_2} + e^{-\kappa r_3})$, and $\phi_0^4(\boldsymbol{r}) = N_f(e^{-\kappa r_1} + e^{-\kappa r_2} + e^{-\kappa r_3} + e^{-\kappa r_4})$, respectively. Here, $\kappa = \sqrt{2I_p}$ and I_p is the ionization potential of the ground state. N_f is the normalization factor and r_n with $n = 1, 2\ldots$ denotes the distance of the electron to the n-th nucleus of the multi-center system. With approximating the continuum state $|\boldsymbol{p}\rangle$ using the plane wave, the odd bound-continuum-transition dipole $D_{odd} \equiv D_{odd}(\omega,\theta) = \langle 0|\boldsymbol{e}_\theta \cdot \boldsymbol{r}|i\sin(\boldsymbol{p}\cdot\boldsymbol{r})\rangle$ at the angle θ (i.e., the angle between the molecular axis and the laser polarization), which is mainly responsible for the emission of odd harmonics along the laser polarization $\boldsymbol{e}_\theta$, can be written as[31,32]

$$D^n_{odd}(\omega,\theta) \propto G^n_{odd}(\omega,\theta)\int dr[e^{-\kappa r}\boldsymbol{e}_\theta\cdot\boldsymbol{r}\sin(\boldsymbol{p}_k\cdot\boldsymbol{r})], \quad (2)$$

with

$$\begin{aligned} G^2_{odd}(\omega,\theta) &= \cos(p_k R/2\cos\theta),\\ G^3_{odd}(\omega,\theta) &= \cos(p_k R_1\cos\theta)+\cos(p_k R_2\cos\theta)\\ &\quad +\cos(p_k R_3\cos\theta) \end{aligned}$$

and

$$\begin{aligned} G^4_{odd}(\omega,\theta) &= \cos(p_k R_1\cos\theta)+\cos(p_k R_2\cos\theta)\\ &\quad +\cos(p_k R_3\cos\theta)+\cos(p_k R_4\cos\theta) \end{aligned}$$

for the cases of H_2^+, H_3^{2+}, and H_4^{3+} molecules, respectively. Similarly, the even bound-continuum-transition dipole $D_{even} \equiv D_{even}(\omega,\theta) = \langle 0|\boldsymbol{e}_\theta\cdot\boldsymbol{r}|\cos(\boldsymbol{p}\cdot\boldsymbol{r})\rangle$ which is related to the emission of even harmonics along the laser polarization can be written as

$$D^n_{even}(\omega,\theta) \propto G^n_{even}(\omega,\theta)\int dr[e^{-\kappa r}\boldsymbol{e}_\theta\cdot\boldsymbol{r}\sin(\boldsymbol{p}_k\cdot\boldsymbol{r})], \quad (3)$$

with

$$\begin{aligned} G^3_{even}(\omega,\theta) &= \sin(p_k R_1\cos\theta)+\sin(p_k R_2\cos\theta)\\ &\quad +\sin(p_k R_3\cos\theta) \end{aligned}$$

and

$$\begin{aligned} G^4_{even}(\omega,\theta) &= \sin(p_k R_1\cos\theta)+\sin(p_k R_2\cos\theta)\\ &\quad +\sin(p_k R_3\cos\theta)+\sin(p_k R_4\cos\theta) \end{aligned}$$

for the cases of H_3^{2+} and H_4^{3+} molecules, respectively. Here, $\boldsymbol{p}_k$ is the effective momentum of the continuum state $|\boldsymbol{p}\rangle \propto |e^{i\boldsymbol{p}_k\cdot\boldsymbol{r}}\rangle$, with $p_k = |\boldsymbol{p}_k| = [2(I_p + E_p)]^{1/2}$ that considers the Coulomb acceleration.[12] $E_p = \omega - I_p$ is the energy of the continuum state $|\boldsymbol{p}\rangle$.

Note, due to the selection rule in the integral of the dipole, the odd dipole D_{odd} is associated with the gerade component $|0_g\rangle$ of the ground state $|0\rangle = |0_g\rangle + |0_u\rangle$ and the even dipole D_{odd} is associated with the ungerade one $|0_u\rangle$. The qualitative relationship between the corresponding dipoles $D_{odd(even)}$ and the HHG power spectra $S_{odd(even)}$ along the laser polarization is[12,43]

$$\begin{aligned} S_{odd}(\omega,\theta) &\sim |a(\omega)D_{odd}(\omega,\theta)|^2, \quad \omega = (2n+1)\omega_0,\\ S_{even}(\omega,\theta) &\sim |a(\omega)D_{even}(\omega,\theta)|^2, \quad \omega = 2n\omega_0. \end{aligned} \quad (4)$$

Here, $a(\omega)$ is the spectral amplitude of the continuum electron which is not sensitive to the angle θ.[12] In the following, we will compare odd–even HHG spectra to the corresponding odd–even dipoles.

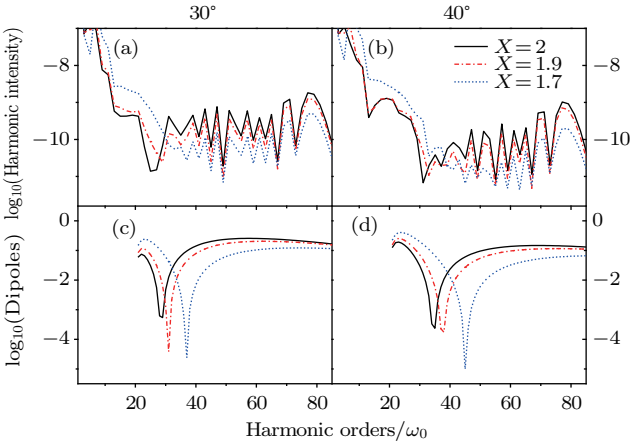


Fig. 3. (color online) Comparisons of odd HHG spectra of Eqs. 1(a) and 1(b) and dipoles $|D^2_{odd}(\omega,\theta)|^2/\omega^4$ of Eqs. 2(c) and 2(d) for 2D linear model molecules H_2^+ with the same ionization potential of $I_p = 1.52$ a.u. but diverse internuclear distances of $X = 2$ a.u. (solid black curves), $X = 1.9$ a.u. (dash-dotted red curves), and $X = 1.7$ a.u. (dotted blue curves) at $\theta = 30°$ (the left column) and $\theta = 40°$ (right). The laser parameters used are $I = 1\times10^{15}$ W/cm^2 and $\lambda = 600$ nm.

3. Results and discussion

3.1. Comparisons of HHG spectra and dipoles

In Fig. 3, we plot the odd harmonic spectra and relevant dipoles from two-center symmetric molecules H_2^+ with the laser intensity $I = 1\times10^{15}$ W/cm^2 and the wavelength $\lambda = 600$ nm for different internuclear distances R at different

orientation angles θ. First of all, from Fig. 3, we can observe that as the internuclear distance changes, the odd spectra at an angle θ differ remarkably from each other. Secondly, at a certain angle, the relative yields of odd harmonics for different internuclear distances in the HHG plateau region are well predicted by relevant dipoles, as described by Eq. (4). Thirdly, the HHG spectra and the corresponding dipoles show a striking minimum arising from two-center interference effects[8] in the HHG plateau region. The position of this minimum shifts toward higher harmonic orders remarkably as the internuclear distance decreases. As the position of the minimum is very sensitive to the internuclear distance, it is possible to judge the positions of the nuclei through this minimum in the spectra. This ultrafast probing procedure with HHG is meaningful especially for a chemical reaction where the positions of the nuclei of molecules can change rapidly in the attosecond scale. However, the situation is different for multi-center linear molecules, as shown in Figs. 4 and 5.

In Figs. 4 and 5, we show the odd spectra and relevant dipoles for linear H_3^{2+} and H_4^{3+} at different internuclear distances and different angles. For comparison, the whole internuclear distances of the model systems are fixed at a certain value with $X_1+X_2=4$ a.u. for H_3^{2+} and $X_1+X_2+X_3=6$ a.u. for H_4^{3+}. The laser parameters used in Fig. 4 for H_3^{2+} are the same as in Fig. 3. For H_4^{3+}, the energy gap between the ground state and the first excited state is smaller and the system is easier to ionize than the case of H_3^{2+}. So in Fig. 5, we have used the laser parameters of $I=6\times10^{14}$ W/cm^2 and $\lambda=1200$ nm.

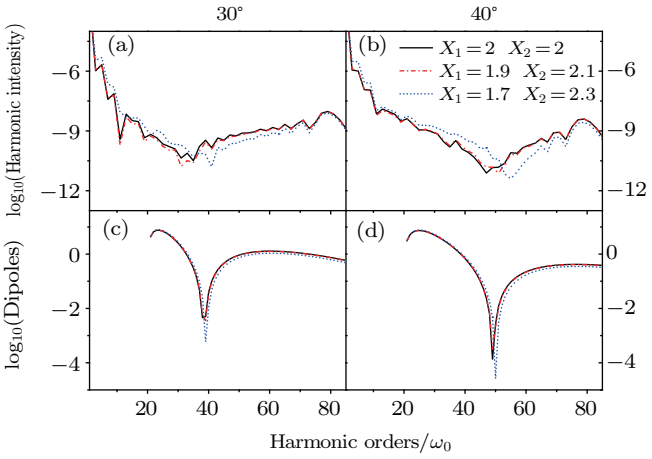


Fig. 4. (color online) Comparisons of odd HHG spectra of Eqs. 1(a) and 1(b) and dipoles $|D^3_{\rm odd}(\omega,\theta)|^2/\omega^4$ of Eqs. 2(c) and 2(d) for 2D linear model molecules H_3^{2+} with the same ionization potential of $I_{\rm p}=1.52$ a.u., but diverse internuclear distances of $X_1=2$ a.u. and $X_2=2$ a.u. (solid black curves), $X_1=1.9$ a.u. and $X_2=2.1$ a.u. (dash-dotted red curves), and $X_1=1.7$ a.u. and $X_2=2.3$ a.u. (dotted blue curves) at $\theta=30°$ (left) and $\theta=40°$ (right). The laser parameters used are $I=1\times10^{15}$ W/cm^2 and $\lambda=600$ nm.

One can observe that the odd spectra in the first row of Fig. 4 also show a striking minimum. However, as the internuclear distance has a small change, the position of the minimum as well as the relative yields of harmonics do not change basically. These phenomena are also well predicted by relevant dipoles in the second row of Fig. 4. Results in Fig. 5 for linear H_4^{3+} are similar to the cases of linear H_3^{2+}, where the spectra are not very sensitive to the change of the internuclear distance.

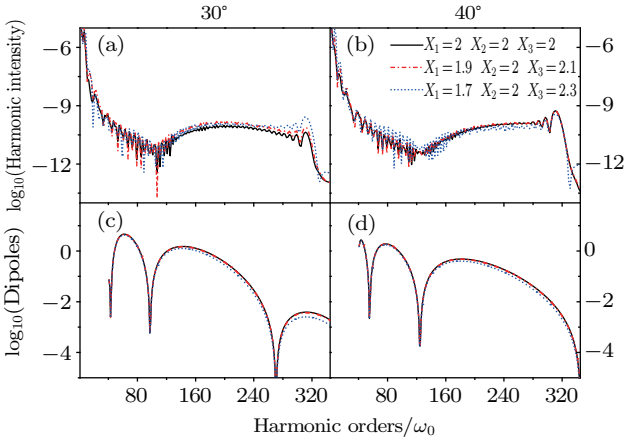


Fig. 5. (color online) Comparisons of odd HHG spectra of Eqs. 1(a) and 1(b) and dipoles $|D^4_{\rm odd}(\omega,\theta)|^2/\omega^4$ of Eqs. 2(c) and 2(d) for 2D linear model molecules H_4^{3+} with the same ionization potential of $I_{\rm p}=1.52$ a.u., but diverse internuclear distances of $X_1=2$ a.u., $X_2=2$ a.u., and $X_3=2$ a.u. (solid black curves), $X_1=1.9$ a.u., $X_2=2$ a.u., and $X_3=2.1$ a.u. (dash-dotted red curves), and $X_1=1.7$ a.u., $X_2=2$ a.u., and $X_2=2.3$ a.u. (dotted blue curves) at $\theta=30°$ (left) and $\theta=40°$ (right). The laser parameters used are $I=6\times10^{14}$ W/cm^2 and $\lambda=1200$ nm.

The results in Figs. 4 and 5 imply that as the minimum in odd spectra for multi-center linear molecules is not sensitive to the internuclear distance, it cannot be used as a tool to probe the positions of the nuclei in an ultrafast time scale.

One of the important characteristics for the HHG from asymmetric molecules is the emission of even harmonics. Next, we turn to even spectra for multi-center asymmetric linear molecules. In Fig. 6, we show the even spectra and relevant even dipoles for asymmetric linear molecules H_3^{2+} at different internuclear distances and orientation angles. Different from the odd cases in Fig. 4, one can observe from Fig. 6 that the even spectra differ remarkably as the internuclear distances change, in agreement with the predictions of relevant dipoles. On the whole, the even spectra are stronger with the increase of the molecular asymmetry (i.e., the increase of the difference between the distances X_1 and X_2). Results of even spectra for linear H_4^{3+} are similar to those in Fig. 6. So we do not present them here.

From the results in Figs. 4–6, we conclude that for multi-center asymmetric linear molecules, the odd spectra are not sensitive to the molecular structure, nor even spectra. However, in the situation, it remains possible to probe the structure of multi-center asymmetric linear molecules in the ultrafast time scale, with the combination of odd and even HHG spectra. Let us discuss this point in more detail. The exact difference between odd and even cases shown above arises

from different generation mechanisms of odd versus even harmonics. As discussed in Ref. [43], the emission of odd harmonics is mainly associated with the gerade component of the asymmetric orbital in the recombination process and the emission of even harmonics is mainly associated with the ungerade one. For multi-center asymmetric linear molecules, when one center of the multi-center system has a small change at position, the gerade component of the asymmetric orbital does not change basically and the ungerade component differs remarkably from the previous one. This difference is mapped in the odd–even HHG spectra, providing the possibility of probing the structural change of the multi-center system with odd–even HHG. Next, we explore the potential ultrafast probing procedure with odd–even harmonics.

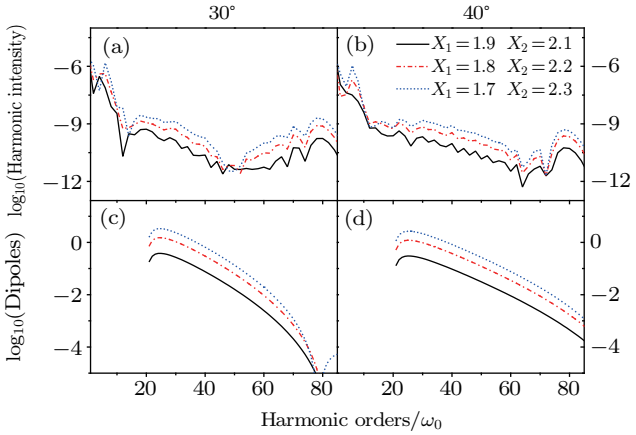


Fig. 6. (color online) Comparisons of even HHG spectra of Eqs. 1(a) and 1(b) and dipoles $|D^3_{\text{even}}(\omega,\theta)|^2/\omega^4$ of Eqs. 3(c) and 3(d) for 2D linear model molecules H_3^{2+} with the same ionization potential of $I_p = 1.52$ a.u., but diverse internuclear distances of $X_1 = 1.9$ a.u and $X_2 = 2.1$ a.u (solid black curves), $X_1 = 1.8$ a.u and $X_2 = 2.2$ a.u. (dash-dotted red curves), and $X_1 = 1.7$ a.u and $X_2 = 2.3$ a.u. (dotted blue curves) at $\theta = 30°$ (left) and $\theta = 40°$ (right). The laser parameters used are $I = 1\times10^{15}$ W/cm^2 and $\lambda = 600$ nm.

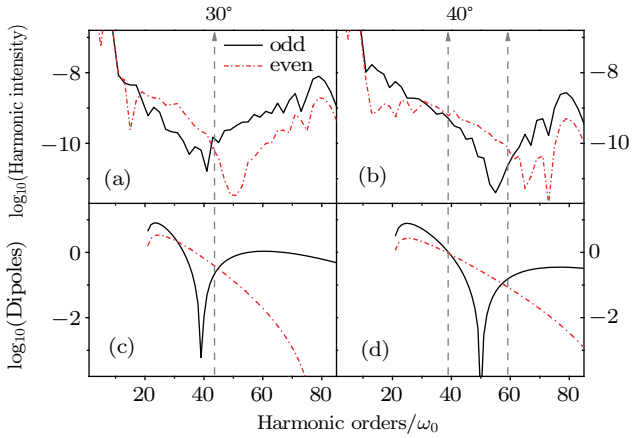


Fig. 7. (color online) Comparisons of odd–even HHG spectra of Eqs. 1(a) and 1(b) and dipoles $|D^3_{\text{odd(even)}}(\omega,\theta)|^2/\omega^4$ of Eqs. (2), (3c), and (3d) for 2D linear model molecules H_3^{2+} with $X_1 = 1.7$ a.u. and $X_2 = 2.3$ a.u. at $\theta = 30°$ (left) and $\theta = 40°$ (right). The dashed arrows indicate the intersection points of odd–even spectra and dipoles. The laser parameters used are $I = 1\times10^{15}$ W/cm^2 and $\lambda = 600$ nm.

In Fig. 7, we plot the odd–even HHG spectra from asymmetric linear molecules H_3^{2+} with $X_1 = 1.7$ a.u. and $X_2 = 2.3$ a.u. at different orientation angles in the first row. For comparison, odd and even harmonics are presented using solid and short-dashed lines, respectively. From Fig. 7, we can see that the relative yields of odd versus even harmonics presented here show a clear dependence on the orientation angle. For example, in Fig. 7(a), the yields of even harmonics are lower than odd ones for low and high harmonic orders. The situation reverses for intermediate harmonic orders, with the odd–even HHG spectra showing two clear intersections. As the orientation angle increases, the positions of the intersections shift towards higher harmonic orders, as shown in Fig. 7(b). The characteristics are also reproduced by the relevant dipoles shown in the second row of Fig. 7. We use dashed arrows to indicate the intersections located at harmonic orders with energy near or higher than $2I_p$ (about H40). For these harmonic orders, the influence of the Coulomb effect on the continuum electron is smaller and the plane-wave approximation for the continuum electron in Eq. (4) is expected to be more applicable.

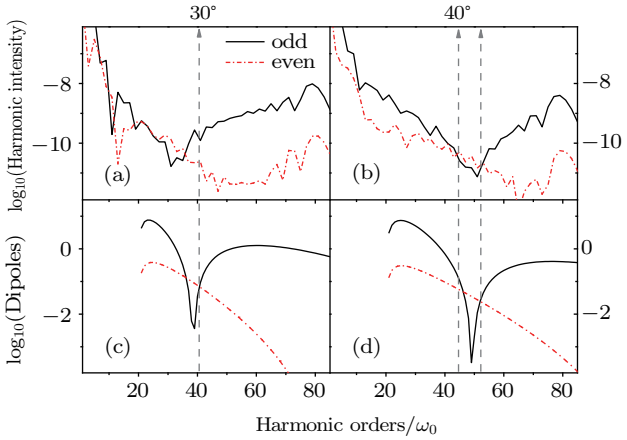


Fig. 8. (color online) Same as Fig. 7, but $X_1 = 1.9$ a.u. and $X_2 = 2.1$ a.u.

In Fig. 8, we show results for other internuclear distances with a small asymmetry in comparison with the case of Fig. 7. On the whole, the odd–even HHG spectra also show some striking intersections, the positions of which agree with the predictions of relevant odd–even dipoles. The calculated odd–even harmonic spectra and relevant dipoles for asymmetric linear H_4^{3+} also show similar phenomena as those presented in Figs. 7 and 8. Based on the above results, we propose the probing of the structure of multi-center asymmetric molecules using the intersections of odd versus even HHG spectra.

It should be stressed that at smaller orientation angles, the intersections of odd–even HHG spectra are located at lower harmonic orders near the threshold, where the influence of the parent ions on the continuum electron is stronger. In this case,

equation (4) based on the plane-wave approximation is not applicable. So we focus our discussion on these two larger angles of $\theta = 30^\circ$ and 40° here. The proposed procedure discussed below is also more applicable for larger angles.

3.2. Probing procedure

Here, all of the terms $G^3_{\rm odd}$ and $G^3_{\rm even}$ are functions of the internuclear distances X_1 and X_2 (see the expressions below Eqs. (2) and (3)). The solution of the above equation gives the predictions of these distances X_1 and X_2. Similarly, one can reconstruct the bond lengths of linear asymmetric molecules H_4^{3+} using the following expressions:

$$\begin{aligned} G^4_{\rm odd}(\omega_1,\theta_1) &= G^4_{\rm even}(\omega_1,\theta_1),\\ G^4_{\rm odd}(\omega_2,\theta_2) &= G^4_{\rm even}(\omega_2,\theta_2),\\ G^4_{\rm odd}(\omega_3,\theta_3) &= G^4_{\rm even}(\omega_3,\theta_3). \end{aligned} \tag{5}$$

Here, ω_1, ω_2, and ω_3 are also the harmonic energy corresponding to the intersections of odd–even HHG spectra at three angles of θ_1, θ_2, and θ_3, respectively. Note, for a certain angle, the odd and even HHG spectra can have two intersections located at higher harmonic orders (see Figs. 7 and 8). Both of the intersections can be used in the reconstruction. In our simulations, we change the orientation angles from $\theta = 20^\circ$ to $\theta = 60^\circ$ and average the bond lengthes reconstructed with odd–even HHG at different intersections and different angles. Relevant results are presented in Fig. 9.

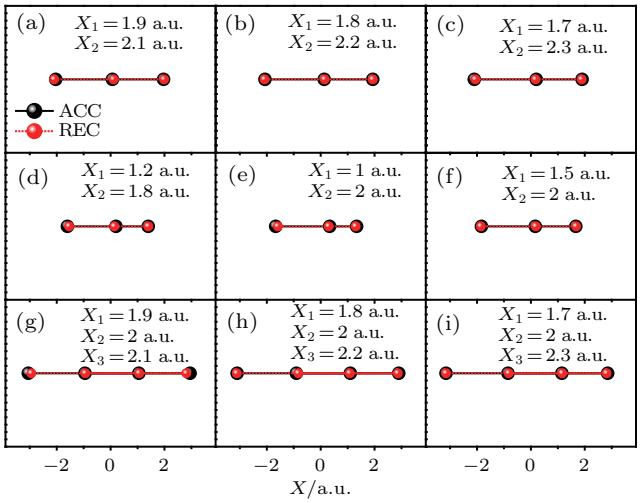


Fig. 9. (color online) Comparisons of the exact results (big-black spheres, ACC) and the reconstructed results of Eqs. (5) and (6) (small-red spheres, REC) for geometry of linear molecules H_3^{2+} ((a)–(f)) and H_4^{3+} ((g)–(i)) with the same ionization potential of $I_{\rm p} = 1.52$ a.u. and diverse internuclear distances. The laser parameters used are $I = 1\times10^{15}$ W/cm^2 and $\lambda = 600$ nm for H_3^{2+}, and $I = 6\times10^{14}$ W/cm^2 and $\lambda = 1200$ nm for H_4^{3+}.

In the first row of Fig. 9, we show the results for H_3^{2+} with $X_1 + X_2 = 4$ a.u. In the second row, those are for H_3^{2+} with $X_1 + X_2 = 3$ a.u. and in the third row for H_4^{3+} with $X_1 + X_2 + X_3 = 6$ a.u. One can observe from Fig. 9, for all of the cases, the reconstruction procedure based on Eqs. (5) and (6) basically reproduces the internuclear distances of the multi-center molecules, implying the applicability of this procedure. We have validated our above discussions with performing simulations at other laser and molecular parameters.

4. Conclusions

In summary, we have studied the HHG from multi-center asymmetric linear molecules H_3^{2+} and H_4^{3+}. We have shown that different from symmetric diatomic molecules such as H_2^+, the odd HHG spectra, especially the interference minima in the odd spectra of multi-center asymmetric molecules are not very sensitive to the change of the internuclear distance, so do the even spectra. The phenomena reflect the fact that when the position of one of the atomic centers of the multi-center system has a small change, the gerade component of the asymmetric orbital (which is mainly responsible for the emission of odd harmonics in the recombination process) does not change basically, but the ungerade component (which is mainly responsible for the emission of even harmonics) differs remarkably from the previous one. In this situation, the intersection of odd versus even HHG spectra can be used as a sensitive tool to probe the structure of multi-center molecules in an ultrafast time scale. We expect that our studies on the HHG of these simple systems will shed light on other multi-center molecules with more complex symmetries.

References

[1] McPherson A, Gibson G, Jara H, Johann U, Luk T S, McIntyre I A, Boyer K and Rhodes C K 1987 *J. Opt. Soc. Am. B* **4** 595
[2] L'Huillier A, Schafer K J and Kulander K C 1991 *J. Phys. B* **24** 3315
[3] Corkum P B and Krausz F 2009 *Nat. Phys.* **3** 381
[4] Krausz F and Ivanov M 2009 *Rev. Mod. Phys.* **81** 163
[5] Lépine F, Ivanov M Y and Vrakking M J J 2014 *Nat. Photon.* **8** 195
[6] Chen J G, Yang Y J, Chen J and Wang B B 2015 *Phys. Rev. A* **91** 043403
[7] Corkum P B 1993 *Phys. Rev. Lett.* **71** 1994
[8] Lein M, Hay N, Velotta R, Marangos J P and Knight P L 2002 *Phys. Rev. Lett.* **88** 183903
[9] Liu C D, Zeng Z N, Wei P F, Liu P, Li R X and Xu Z Z 2010 *Phys. Rev. A* **81** 033426
[10] Zhao J and Zhao Z X 2010 *Chin. Phys. Lett.* **27** 063301
[11] Li W Y, Dong F L, Yu S J, Wang S, Yang S P and Chen Y J 2015 *Opt. Express* **23** 31010
[12] Itatani J, Levesque J, Zeidler D, Niikura Hiromichi, Pepin H, Kieffer J C, Corkum P B and Villeneuve D M 2004 *Nature* **432** 867
[13] Zhou X X, Tong X M, Zhao Z X and Lin C D 2005 *Phys. Rev. A* **72** 033412
[14] Li Y, Zhu X S, Lan P F, Zhang Q B, Qin M Y and Lu P X 2014 *Phys. Rev. A* **89** 045401
[15] Du H, Pan X F, Liu H F, Zhang H D, Zhang J, Guo J and Liu X S 2016 *Chin. Phys. B* **25** 093202
[16] de Nalda R, Heesel E, Lein M, Hay N, Velotta R, Springate E, Castillejo M and Marangos J P 2004 *Phys. Rev. A* **69** 031804
[17] Kanai T, Minemoto S and Sakai H 2005 *Nature* **435** 470
[18] Vozzi C, Calegari F, Benedetti E, Caumes J P, Sansone G, Stagira S, Nisoli M, Torres R, Heesel E, Kajumba N, Marangos J P, Altucci C and Velotta R 2005 *Phys. Rev. Lett.* **95** 153902
[19] Le A T, Tong X M and Lin C D 2006 *Phys. Rev. A* **73** 041402
[20] Lein M 2007 *J. Phys. B* **140** R135

[21] Baker S, Robinson J S, Lein M, Chirila C C, Torres R, Bandulet H C, Comtois D, Kieffer J C, Villeneuve D M, Tisch J W G and Marangos J P 2008 *Phys. Rev. Lett.* **101** 053901
[22] Cui H F and Miao X Y 2017 *Chin. Phys. B* **26** 103302
[23] Zhang J, Pan X F, Xu T T, Zhang H D, Du H, Guo J and Liu X S 2017 *Chin. Phys. B* **26** 013202
[24] Li Y P, Yu S J, Duan X Y, Shi Y Z and Chen Y J 2016 *J. Phys. B* **49** 075603
[25] Chen Y J, Liu J and Hu Bambi 2009 *J. Chem. Phys.* **130** 044311
[26] Chen Y J and Hu Bambi 2009 *J. Chem. Phys.* **131** 244109
[27] Smirnova O, Mairesse Y, Patchkovskii S, Dudovich N, Villeneuve D, Corkum P and Ivanov M Yu 2009 *Nature* **460** 972
[28] Li W Y, Wang S, Shi Y Z, Yang S P and Chen Y J 2017 *J. Phys. B* **50** 085003
[29] Kamta G L, Bandrauk A D and Corkum P B 2005 *J. Phys. B* **38** L339
[30] Bian Xue-Bin and Bandrauk A D 2010 *Phys. Rev. Lett.* **105** 093903
[31] Chen Y J and Zhang B 2011 *Phys. Rev. A* **84** 053402
[32] Zhang B, Chen Y J, Jiang X Q and Sun X D 2013 *Phys. Rev. A* **88** 053428
[33] Miao X Y and Du H N 2013 *Phys. Rev. A* **87** 053403
[34] Miao X Y and Zhang C P 2014 *Phys. Rev. A* **89** 033410
[35] Lu R F, Yu C, Wang Y H, Shi Q and Zhang Y D 2014 *Phys. Lett. A* **378** 90
[36] Zhang J, Liu H F, Pan X F, Du H, Guo J and Liu X S 2016 *Chin. Phys. B* **25** 053202
[37] Li Y P, Yu S J, Li W Y and Chen Y J 2017 *Phys. Rev. A* **95** 063412
[38] Wang S, Cai J and Chen Y J 2017 *Phys. Rev. A* **96** 043413
[39] Wang R H, Jiang H B, Yang H, Wu C Y and Gong Q H 2005 *Chin. Phys. Lett.* **22** 1913
[40] Zhu X S, Zhang Q B, Hong W Y, Lan P F and Lu P X 2011 *Opt. Express* **19** 436
[41] Du H, Luo L, Wang X and Hu B 2012 *Phys. Rev. A* **86** 013846
[42] Pan Y, Zhao S F and Zhou X X 2013 *Phys. Rev. A* **87** 035805
[43] Chen Y J, Fu L B and Liu J 2013 *Phys. Rev. Lett.* **111** 073902
[44] Wu J, Zeng H and Guo C L 2006 *Phys. Rev. A* **74** 031404
[45] Bian X B and Bandrauk A D 2010 *Phys. Rev. Lett.* **105** 093903
[46] Etches A, Gaarde M B and Madsen L B 2011 *Phys. Rev. A* **84** 023418
[47] Heslar J, Telnov D and Chu S I 2011 *Phys. Rev. A* **83** 043414
[48] Yue S J, Du H C, Wu H M, Xue S, Zhao J C and Hu B T 2017 *Chin. Phys. B* **26** 074215
[49] Frumker E, Hebeisen C T, Kajumba N, Bertrand J B, Wörner H J, Spanner M, Villeneuve D M, Naumov A and Corkum P B 2012 *Phys. Rev. Lett.* **109** 113901
[50] Kraus P M, Rupenyan A and Wörner H J 2012 *Phys. Rev. Lett.* **109** 233903
[51] Chen J, Yu S, Li Y, Wang S and Chen Y 2017 *Chin. Phys. B* **26** 094209
[52] Frumker E, Kajumba N, Bertrand J B, Wörner H J, Hebeisen C T, Hockett P, Spanner M, Patchkovskii S, Paulus G G, Villeneuve D M, Naumov A and Cokum P B 2012 *Phys. Rev. Lett.* **109** 233904
[53] Kraus P M, Baykusheva D and Wörner H J 2014 *Phys. Rev. Lett.* **113** 023001
[54] Yu S, Zhang B, Li Y, Yang S and Chen Y 2014 *Phys. Rev. A* **90** 053844
[55] Kraus P M, Tolstikhin O I, Baykusheva D, Rupenyan A, Schneider J, Bisgaard C Z, Morishita T, Jensen F, Madsen L B and Wörner H J 2015 *Nat. Commun.* **6** 7039
[56] Zhang B, Yu S J, Chen Y J, Jiang X Q and Sun X D 2015 *Phys. Rev. A* **92** 053833
[57] Li W Y, Yu S J, Wang S and Chen Y J 2016 *Phys. Rev. A* **94** 053407
[58] Shi Y Z, Zhang B, Li W Y, Yu S J and Chen Y J 2017 *Phys. Rev. A* **95** 033406
[59] Yu S J, Li W Y, Li Y P and Chen Y J 2017 *Phys. Rev. A* **96** 013432
[60] Baker S, Robinson J S, Haworth C A, Teng H, Smith R A, Chirilă C C, Lein M, Tisch J W G and Marangos J P 2006 *Science* **312** 424
[61] Madsen C B and Madsen L B 2007 *Phys. Rev. A* **76** 043419
[62] Madsen C B, Abu-samha M and Madsen L B 2010 *Phys. Rev. A* **81** 043413
[63] Wong M C H, Brichta J P and Bhardwaj V R 2010 *Phys. Rev. A* **81** 061402
[64] Ciappina M F, Becker A and Jaroń-Becker A 2007 *Phys. Rev. A* **76** 063406
[65] Baykusheva D, Ahsan M S, Lin N and Wörner H J 2016 *Phys. Rev. Lett.* **116** 123001
[66] Vampa G, McDonald C R, Orlando G, Klug D D, Corkum P B and Brabec T 2014 *Phys. Rev. Lett.* **113** 073901
[67] Wu M, Ghimire S, Reis D A, Schafer K J and Gaarde M B 2015 *Phys. Rev. A* **91** 043839
[68] Guan Z, Zhou X X and Bian X B 2016 *Phys. Rev. A* **93** 033852
[69] Du T Y, Guan Z, Zhou X X and Bian X B 2016 *Phys. Rev. A* **94** 023419
[70] Yu C, Zhang X R, Jiang S C, Cao X, Yuan G L, Wu T, Bai L H and Lu R F 2016 *Phys. Rev. A* **94** 013846
[71] Jiang S, Wei H, Chen J, Yu C, Lu R and Lin C D 2017 *Phys. Rev. A* **96** 053850
[72] Jiang S C, Yu C, Yuan G L, Wu T, Wang Z W and Lu R F 2017 *J. Phys.: Condens. Matter* **29** 275702
[73] Kraus P M, Mignolet B, Baykusheva D, Rupenyan A, Horny L, Penka E F, Grassi G, Tolstikhin O I, Schneider J, Jensen F, Madsen L B, Bandrauk A D, Remacle F and Wörner H J 2015 *Science* **350** 790
[74] Rupenyan A, Kraus P M, Schneider J and Wörner H J 2013 *Phys. Rev. A* **87** 031401
[75] Feit M D, Jr Fleck J A and Steiger A 1982 *J. Comput. Phys.* **47** 412
[76] Shi Y Z, Wang S, Dong F L, Li Y P and Chen Y J 2017 *J. Phys. B* **50** 065004