Theoretical study of the odd–even-order harmonic generation for asymmetric ions in non-Born–Oppenheimer approximation*

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We calculated the harmonic spectra generated from the asymmetric molecules of HD^+ and HeH^{2+}. It is found that HD^+ produces only odd harmonics, while HeH^{2+} produces both odd and even harmonics. Further analysis reveals that for both HD^+ and HeH^{2+}, the nuclear dipole acceleration can generate even harmonics, but it is three orders of magnitude lower than that of the electron. Hence, the electronic dipole acceleration dominates the harmonic generation. For HD^+, the electronic dipole acceleration only contributes to the generation of odd harmonics, but for HeH^{2+} it contributes to the generation of both odd and even harmonics. Besides, one concept of the broken degree of system-symmetry is proposed to explain the different odd-even property between the harmonic spectra of HD^+ and HeH^{2+}.

Keywords: strong field physics, odd–even-order harmonic, dipole acceleration, asymmetric ions

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1. Introduction

Over the last twenty years, the high-order harmonic generation (HHG) from atoms and molecules in the intense laser fields has been a hot subject.[1–5] Great progress has been made in understanding and application of HHG.[6,7] Not only does it offer the potential for creating new coherent light sources, single attosecond pulses,[8–10] or attosecond pulse trains (APT),[11,12] but also it is an important tool for ultrafast imaging of a dynamic molecular system.[13–17] So far, the physical process of HHG can be well comprehended by the semi-classical three-step model.[18] Firstly, the electron tunnels out of the potential barrier formed by the Coulomb potential and the laser field; then the ionized electron moves in the laser field and gains additional kinetic energy; finally, it recombines with the core and a photon is emitted (high-order harmonic). Besides the atomic system, HHG has been investigated in many other systems.[19–35]

As is well known, the harmonic spectrum from a single atom in a strong laser field presents some common and significant features.[19–21] Typically, only odd harmonics are observed in theoretical simulations and experiments.[21,22] One non-perturbative proof which invokes the concept of inversion symmetry has already been given by Ben-Tal *et al.*[23] This theory can work well for any system of the dynamic symmetry.[24]

In recent years, the molecular high-order harmonic generation (MHOHG) has been broadly studied, since molecules have more degrees of freedom which can be used to further control HHG.[26–30] The odd–even-order harmonic generation from molecular systems has been also investigated.[31–34] It has been shown that the molecules are more complicated than the atom,[35,36] but the harmonic generation spectra (HGS) are still composed only of odd harmonics for symmetric molecules in Born–Oppenheimer approximation.[31,32] Moreover, both odd and even harmonics were observed from asymmetric molecules with the fixed nuclei.[31] However, for HD molecule, even harmonics were first observed in non-Born–Oppenheimer approximation.[32] Recently, we found that the HD molecule still generated only odd harmonics in non-Born–Oppenheimer approximation though the generation of even harmonics is possible in principle.[33] Nevertheless, so far, it is still an open question whether the HD^+ can generate even harmonics in non-Born–Oppenheimer approximation, in which the nuclei are easier to dissociate than those in the HD molecule.

In this work, we give an affirmative answer. The HD^+ also generated only odd harmonics. Besides, we investigate the HHG of HeH^{2+} and find that both odd and even harmonics appear in the HGS. To our knowledge, the selection rules[23] cannot explain the different odd–even property occurring in the HGS of HD^+ and HeH^{2+}. Our theoretical analysis indicates that for HD^+ and HeH^{2+}, the nuclear dipole acceleration makes a contribution to producing even harmonics, but it is about three orders of magnitude less than that of the elec-

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tron. This leads to the fact that the impact of the nuclear dipole acceleration to the HGS can be ignored. Therefore, the HHG primarily depends on the contribution of the electronic dipole acceleration. For HD^+, the electronic dipole acceleration only results in the generation of odd harmonics. However, for HeH^{2+}, it can produce both odd and even harmonics. In addition, we also invoke the concept of the broken degree of system-symmetry to interpret the different odd–even property between the harmonic spectra of HD^+ and HeH^{2+}. Generally speaking, both odd and even harmonics can be produced when the system-symmetry is destroyed. Whereas, the visible even harmonics could be generated only if the breaking of system-symmetry gets to a certain degree. For HD^+, the system-symmetry is broken in a very small degree. Hence, only odd harmonics appear in the HGS. For HeH^{2+}, because the system-symmetry is drastically broken, there are both odd and even harmonics in the HGS.

2. Simulation methods

For the numerical simulations, we have used the three-body model with two nuclei and one electron, where the motion of nuclei and electron is restricted to the polarization direction of the laser field in the one-dimensional (1D) model. Within this model, the coupling to the laser field is treated within the dipole approximation. As a result, the center-of-mass motion does not couple to the laser field. Then, the time-dependent Schrödinger equation (TDSE) can be given as (atomic units are used throughout)[32,37,38]

$$\mathrm{i}\frac{\partial}{\partial t}\psi(R,z;t) = [H_0 + V_{\mathrm{I}}(t)]\,\psi(R,z;t), \tag{1}$$

with the field-free Hamiltonian

$$H_0 = -\frac{1}{2\mu_{\mathrm{e}}}\frac{\partial^2}{\partial z^2} - \frac{1}{2\mu_n}\frac{\partial^2}{\partial R^2} + V_{\mathrm{c}}, \tag{2}$$

$$V_{\mathrm{c}} = \frac{q_a q_b}{\sqrt{R^2+\beta}} - \frac{q_a}{\sqrt{(z-z_a)^2+\alpha}} - \frac{q_b}{\sqrt{(z+z_b)^2+\alpha}}, \tag{3}$$

where z is the electron coordinate when the origin is the center of mass; R is the inter-nuclear distance; q_a and q_b are the electric charges of two nuclei; $z_a = m_b/(m_a+m_b)R$ and $z_b = m_a/(m_a+m_b)R$ are the positions of the first nucleus and the second nucleus, respectively; $\mu_{\mathrm{n}} = m_a m_b/(m_a+m_b)$ and $\mu_{\mathrm{e}} = (m_a+m_b)/(m_a+m_b+1)$ are the reduced mass of nucleus and electron, with m_a and m_b the masses of the first nucleus and the second nucleus. The interaction of the electron with the laser pulse is taken into account via $V_{\mathrm{I}}(t)$ given in length gauge as

$$V_{\mathrm{I}}(t) = \hat{D}\cdot E(t) = \left[-\left(1+\frac{q_a+q_b-1}{m_a+m_b+1}\right)z - \lambda R\right]E(t), \tag{4}$$

where $\hat{D}$ is the dipole operator and $\lambda = (q_a m_b - q_b m_a)/(m_a+m_b)$ is the asymmetry parameter. The laser field has the form $E(t) = E_0 f(t)\sin\omega t$, where E_0 and ω are the peak amplitude and circular frequency. The envelope function $f(t)$ is a linear ramp from zero to one during the first five optical cycles. Afterwards, the field amplitude is held constant. The total time of propagation is 10 optical cycles. In our simulations, the wavelength of the laser pulse is 770 nm and the peak intensity is 4×10^{14} W/cm^2. By adopting this laser field, the peak of spectra appears much cleaner (Fig. 1). Hence, our conclusion is more explicit. In fact, the shape of envelope and the carrier-envelope phase (CEP) have no influence on our conclusion, when the red-shifted is less prominent.

Before the time evolution of the wave function, an initial state is requisite. For H_2^+ and HD^+, the initial states can be obtained by propagation in imaginary time. We choose the soft-core parameters $\alpha = 1.0$ and $\beta = 0$ such that the energy of the ground state and equilibrium distance are -0.7763 a.u. and 2.745 a.u., respectively. However, a bound state cannot be obtained by this method for HeH^{2+}, because the lowest state $1s\sigma$ is dissociated.[40] In order to solve this problem, we adopt the spectral method.[39] Then, the first excited state $2p\sigma$ which is one bound state of HeH^{2+} is chosen to be the initial state. The soft-core parameters $\alpha = 0.3$ and $\beta = 0.01$ are adopted to reproduce the ionization energy of 1.03 a.u. and equilibrium distance of 3.89 a.u.[40]

After the initial states have been obtained, we solve Eq. (1) using the Crank–Nicholson method[41]

$$\begin{aligned}\psi(R,z;t+\delta t) = {} & \mathrm{e}^{-\mathrm{i}H_z\delta t/2}\,\mathrm{e}^{-\mathrm{i}H_R\delta t}\,\mathrm{e}^{-\mathrm{i}H_z\delta t/2}\psi(R,z;t) \\ & + O(\delta t^3).\end{aligned} \tag{5}$$

Then the dipole acceleration expectation value can be calculated by the Ehrenfest theorem

$$\begin{aligned} a(t) &= \frac{\mathrm{d}^2}{\mathrm{d}t^2}\langle\psi(t)|\hat{D}|\psi(t)\rangle \\ &= \frac{1}{\mu_{\mathrm{e}}}\langle\psi(t)|\frac{\partial H}{\partial z}|\psi(t)\rangle \\ &\quad + \frac{\lambda}{\mu_{\mathrm{n}}}\langle\psi(t)|\frac{\partial H}{\partial R}|\psi(t)\rangle \\ &= a_{\mathrm{e}}(t) + \lambda a_{\mathrm{n}}(t), \end{aligned} \tag{6}$$

where $\langle\psi(t)|\hat{D}|\psi(t)\rangle$ is the dipole moment

$$\begin{aligned} &\langle\psi(t)|\hat{D}|\psi(t)\rangle = -\langle\psi(t)|z|\psi(t)\rangle \\ &-\lambda\langle\psi(t)|R|\psi(t)\rangle = d_{\mathrm{e}}(t) + \lambda d_{\mathrm{n}}(t). \end{aligned} \tag{7}$$

$d_{\mathrm{e}}(t)$ and $d_{\mathrm{n}}(t)$ are the electronic dipole moment and the relative nuclear dipole moment, respectively. $a_{\mathrm{e}}(t)$ and $a_{\mathrm{n}}(t)$ are the electronic dipole acceleration and the relative nuclear dipole acceleration. Through the Fourier transforming of the dipole acceleration $a(t)$, the MHOHG spectrum can be written as

$$P(\omega) = |\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{+\infty} a(t)\,\mathrm{e}^{-\mathrm{i}\omega t}|^2, \tag{8}$$

where ω is the frequency of the high-order harmonics.

3. Results and discussion

The harmonic spectra generated by H_2^+, HD^+, and HeH^{2+} are shown in Fig. 1. The left column is the results of 1D simulations. In our calculation, we use a grid from -400 a.u. to 400 a.u. containing 8000 grid points in the z-axis, and a grid from 0 to 100 a.u. containing 1000 grid points in the R-axis. The time step of wave function propagation is set to be 0.05 a.u. In order to avoid spurious reflections of the wave function from the boundary $z = \pm 400$ and $R = 100$, the wave function is multiplied by a cos1/8 mask function over a range of $|z| = 356$–400 and $R = 88$–100 per evolving step. From Figs. 1(a)–1(c), one can see that the HGS of H_2^+ and HD^+ are composed only of odd harmonics, while the HGS of HeH^{2+} are composed of both odd and even harmonics. In fact, for HeH^{2+}, a two-plateau structure and a continuum spectrum have been observed for long wavelength.[42,43] But in this paper, the wavelength of laser is just 770 nm, so the spectra of HeH^{2+} just exhibit one cut-off, which is in agreement with the results of Refs. [42] and [43] in short wavelength. In addition, we also display the results of 3D simulations in the right column. It is clear that in two cases (1D and 3D), HD^+ produces only odd harmonics, while HeH^{2+} produces both odd and even harmonics.

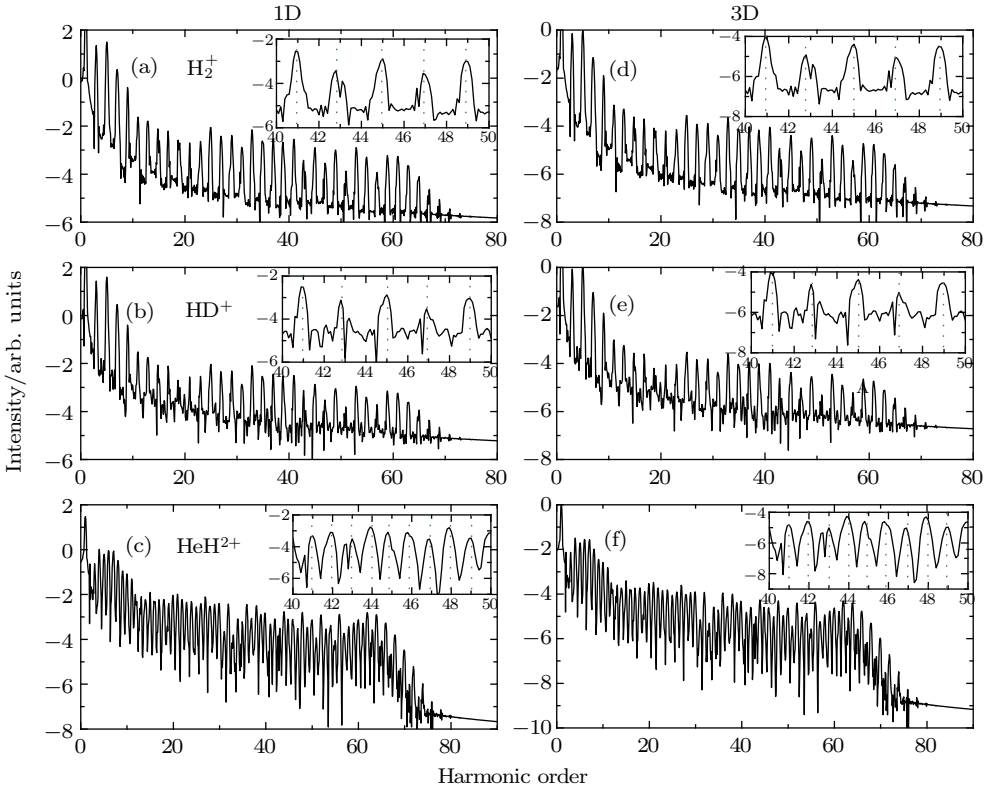


Fig. 1. Harmonic spectra generated from (a) H_2^+, (b) HD^+, and (c) HeH^{2+} by a laser with the peak intensity of 4×10^{14} W/cm^2 and the wavelength of 770 nm. The corresponding 3D simulation results are shown in the right column. The insets are the enlarged area of the harmonic spectra from 40th to 50th order.

The result of H_2^+ can be well understood with the selection rules in Ref. [23], which can be described in the following way. The probability to produce the nth harmonic from a system is[23]

$$\sigma^{(n)} \propto n^4 |\langle\langle \phi(\boldsymbol{r},t)|\hat{D}\,\mathrm{e}^{-\mathrm{i}n\omega t}|\phi(\boldsymbol{r},t)\rangle\rangle|^2, \tag{9}$$

where the double bracket stands for the integration over space and time. $\hat{D}$ represents the dipole operator. In the case of a symmetric system, the Hamiltonian $H(t)$ is invariant under the second order dynamic symmetry (DS) operator $\hat{P}_2 = (\boldsymbol{r} \to -\boldsymbol{r}, t \to t + \pi/\omega)$. The nth harmonic is therefore emitted only if $\langle\langle\phi|\hat{D}\,\mathrm{e}^{-\mathrm{i}n\omega t}|\phi\rangle\rangle = \langle\langle\hat{P}_2\phi|\hat{P}_2\hat{D}\,\mathrm{e}^{-\mathrm{i}n\omega t}\hat{P}_2^{-1}|\hat{P}_2\phi\rangle\rangle \neq 0$, leading to $\hat{D}(\boldsymbol{r})\,\mathrm{e}^{-\mathrm{i}n\omega t} = \hat{D}(-\boldsymbol{r}) \times \exp[-\mathrm{i}n\omega(t+\pi/\omega)]$ .It is fulfilled only with odd n for a symmetric system. Therefore, only the odd harmonics are generated for H_2^+. However, for HD^+ and HeH^{2+}, the selection rules cannot explain the difference of the HGS in non-Born–Oppenheimer approximation. Namely, only odd harmonics are produced for HD^+, but both odd and even harmonics for HeH^{2+}.

As shown in Ref. [33], the nuclear dipole acceleration may play a role in the HHG beyond Born–Oppenheimer approximation. It can be seen from Eq. (6) that for H_2^+, $a(t) = a_e(t)$ due to the asymmetry parameter $\lambda = 0$. However, for HD^+ and HeH^{2+}, the asymmetry parameter $\lambda \neq 0$, and thus the nuclear dipole acceleration can contribute to the HHG. In order to examine the role of nuclear dipole acceleration, figure 2 presents the harmonic spectra calculated with $a_n(t)$ for

HD^+ and HeH^{2+}. As shown in Fig. 2, the cutoff of the harmonic spectrum is smaller than that in Fig. 1 for both cases (harmonic order lower than 15). This is because the nuclei are easy to dissociate when the only electron of HD^+ and HeH^{2+} is ionized by the intense laser pulse. Besides, it is found that there are even harmonics in both of the harmonic spectra. Consequently, in principle, the generation of even harmonics is possible for HD^+ and HeH^{2+}. But the intensity of harmonics is lower than that in Fig. 1.

For further capturing the signatures of nuclear dipole acceleration, figure 3 shows the electronic dipole acceleration $a_e(t)$ and the nuclear dipole acceleration $a_n(t)$ for HD^+ and HeH^{2+}. For comparison, $a_n(t)$ is amplified by μ_n times. Clearly, for both HD^+ and HeH^{2+}, the electronic dipole acceleration is about three orders of magnitude larger than the nuclear dipole acceleration. Then we can conclude that the contribution of the nuclear dipole acceleration to HHG can be ignored. In order to verify it, we also present the harmonic spectra of HD^+ and HeH^{2+} calculated with $a_e(t)$ in Figs. 3(c) and 3(d). It is found the harmonic spectra calculated with $a_e(t)$ are the same as that calculated with $a_e(t)+\lambda a_n(t)$. This implies that the influence of the nuclear dipole acceleration to HHG can be indeed ignored for both HD^+ and HeH^{2+}, though it can contribute to the even-order harmonic generation. Apparently, it is not the reason why the odd–even property of the HGS of HD^+ and HeH^{2+} is different.

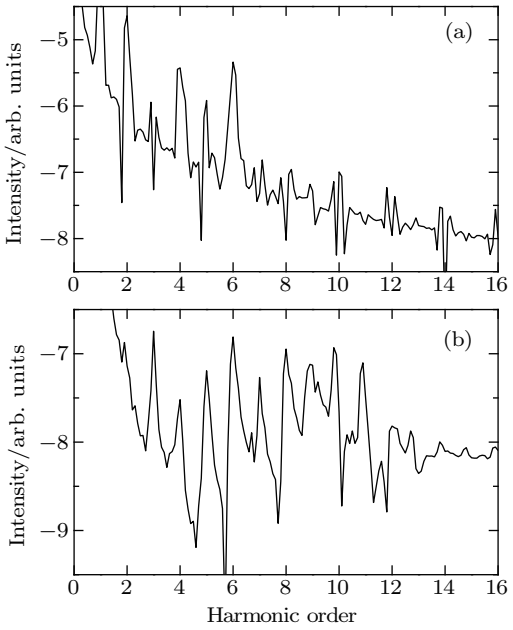


Fig. 2. The harmonic spectra calculated with $a_n(t)$ for (a) HD^+ and (b) HeH^{2+}.

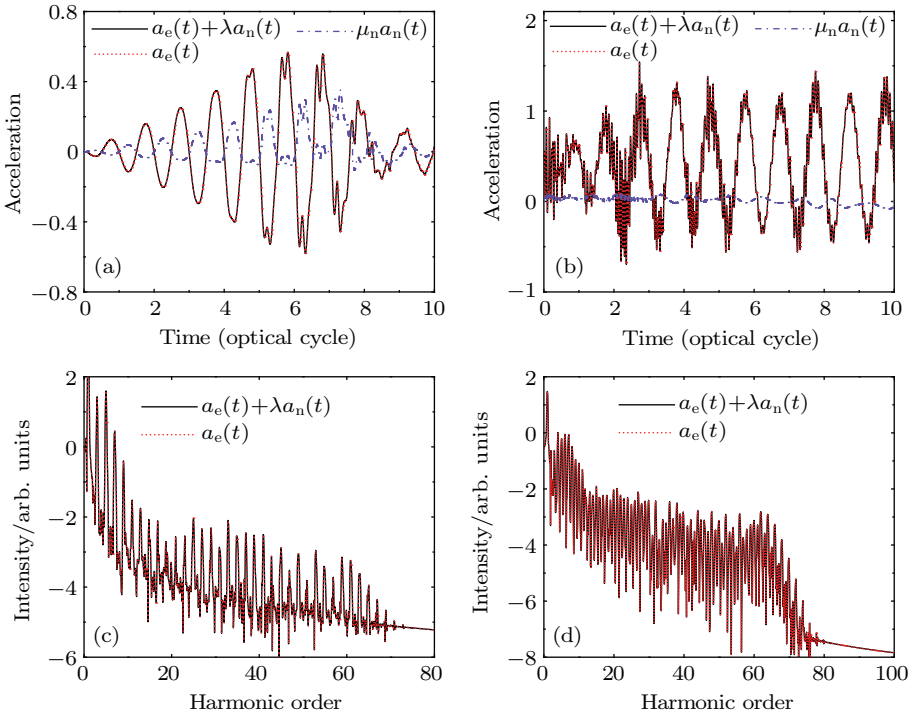


Fig. 3. (color online) The electronic dipole acceleration $a_e(t)$ (red dotted line), the nuclear dipole acceleration $a_n(t)$ (violet dashed-dotted line), and the total dipole acceleration $a_e(t)+\lambda a_n(t)$ (black solid line) for (a) HD^+ and (b) HeH^{2+}. For clarity, $a_n(t)$ is amplified by μ_n times. (c) Harmonic spectra generated from HD^+. Black solid line is calculated with $a_e(t)+\lambda a_n(t)$, and red dotted line is calculated with $a_e(t)$. (d) Same as panel (c) but for HeH^{2+}.

As is already known, the contribution of the nuclear dipole acceleration to the HHG can be ignored. Therefore, the electronic dipole acceleration mainly contributes to the harmonic generation. In order to confirm our inference, the total dipole acceleration $a_e(t)+\lambda a_n(t)$ is displayed in Fig. 3. As shown in Figs. 3(a) and 3(b), for both HD^+ and HeH^{2+},

the total dipole acceleration $a_e(t)+\lambda a_n(t)$ and the electronic dipole acceleration $a_e(t)$ are almost equal. Moreover, it can be clearly seen from Figs. 3(c) and 3(d) that the harmonic spectra calculated with $a_e(t)$ are the same as that calculated with $a_e(t)+\lambda a_n(t)$. All of these imply that the HHG mainly relies on the contribution of the electronic dipole acceleration. The detailed theoretical derivation is discussed in Ref. [33], which is in good agreement with our conclusion.

In order to further understand the difference between the harmonic spectra of HD^+ and HeH^{2+}, figure 4 shows the Coulomb potential for H_2^+, HD^+, and HeH^{2+}. In Fig. 4, for H_2^+, the potential well is perfectly symmetric. Certainly, H_2^+ obeys the selection rules in Ref. [23]. For HD^+, the potential well is still in symmetry, though it has a small translation in the $z$axis. One can see that the potential well of HD^+ and H_2^+ could coincide by a small translation in the z axis. By contrast, for HeH^{2+}, the potential-well symmetry is drastically destroyed. Thus, one concept of the broken degree of system-symmetry can be proposed to explicate the difference of the HGS of HD^+ and HeH^{2+}. In detail, for HD^+, the system-symmetry is only destroyed into a small extent, which shows that the nuclear acceleration can contribute to the HHG. However, its contribution can be ignored. Therefore, only odd harmonics are observed for HD^+. While for HeH^{2+}, the system-symmetry is drastically destroyed. Hence, there are both odd and even harmonics in the spectrum.

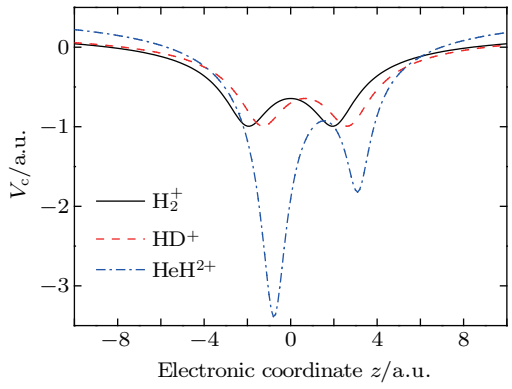


Fig. 4. (color online) Coulomb potential for H_2^+ (black solid line), HD^+ (red dashed line), and HeH^{2+} (blue dashed-dotted line). For H_2^+ and HD^+, $R=2.745$ a.u. For HeH^{2+}, $R=3.89$ a.u.

To examine the influence of the broken degree of system-symmetry, we used the model molecule with different $\Delta q = q_b - q_a$ ($q_a = 1, q_b \geq q_a$). Within this model, one can change the broken degree of system-symmetry by manipulating Δq, where the bigger Δq is, the more completely the broken extent is. The results are shown in Fig. 5. It can be seen that as Δq increases, the even harmonics gradually appear and the intensity becomes higher and higher. Hence, we invoke the concept of the broken degree of system-symmetry to explain the different odd–even property between the harmonic spectra of HD^+ and HeH^{2+}. In other words, to observe obvious even harmonics, the system-symmetry should be broken to some extent.

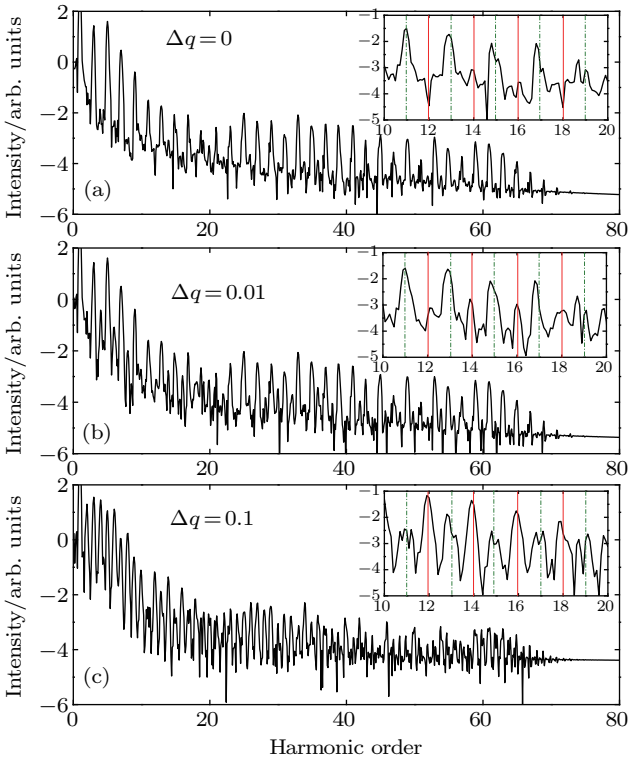


Fig. 5. The harmonic spectra with $\Delta q = 0$, 0.01, and 0.1. The insets are the enlarged area of the harmonic spectra from order 10 to 20.

4. Conclusion

In summary, we have studied the HHG of H_2^+, HD^+, and HeH^{2+} in non-Born–Oppenheimer approximation. It is found that HD^+ generates only odd harmonics, while HeH^{2+} generates both odd and even harmonics. Note that the selection rules[23] cannot explain this difference. Then theoretical analysis implies that the dipole acceleration of the electron and the nuclei jointly contribute to HHG. However, for both HD^+ and HeH^{2+}, the nuclear dipole acceleration has no effects on the HHG, since it is three orders of magnitude lower than that of the electron. Thus, the electronic dipole acceleration is the key of the HHG. For HD^+, the electronic dipole acceleration only leads to the odd-order harmonic generation, but for HeH^{2+}, it leads to the odd–even-order harmonic generation. Additionally, we proposed the concept of the broken degree of system-symmetry to explain the difference of the HGS of HD^+ and HeH^{2+}. It is found that the visible even harmonics could be generated only if the breaking of system-symmetry gets to a certain degree. For HD^+, due to the tiny destruction of system-symmetry, it generates only odd harmonics. However, for HeH^{2+}, both odd and even harmonics are produced, which is attributed to the drastic breaking of system-symmetry.

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