

## EFFECT OF MOLECULAR CHARGE ASYMMETRY ON EVEN-TO-ODD RATIO OF HIGH-ORDER HARMONIC GENERATION

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**Abstract.** *Recently, asymmetric molecules, such as  $\text{HeH}_2^+$ , CO, OCS, HCl, have been evolved much attention since its rich information in the high-order harmonic generation (HHG), whose ratio of adjacent even and odd harmonics characterizes the asymmetry of molecules. In this paper, we study the dependence of even-to-odd ratio on the asymmetric parameters, in particular, the nuclear-charge ratio, and the permanent dipole, by exploiting a simple but general model of asymmetric molecules  $Z_1Z_2$  subjected to an intense laser pulse. The HHG is simulated by the numerical method of solving the time-dependent Schrödinger equation. We find out that this even-to-odd ratio strongly depends on the nuclear-charge ratio. In particular, the even-to-odd ratio reaches its maximum when the nuclear-charge ratio is about from 0.5 to 0.7. Besides, the dependence on the permanent dipole of the even-to-odd ratio has a non-trivial law. To explain, we calculate the analytical ratio of the transition dipole according to the emission of even and odd harmonics, and this ratio is well consistent with the even-to-odd ratio of the HHG.*

**Keywords:** HHG, even harmonics, even-to-odd ratio, asymmetric molecule, permanent dipole.

**Classification numbers:** 42.65.Ky.

## I. INTRODUCTION

In the recent decades, high-order harmonic generation (HHG) emitted from atoms, molecules, or solids interacting with an ultrashort intense laser pulse has been a hot topic since its wide applications in strong-field physics and attosecond science [1–5]. The HHG can be well understood by the three-step model, where electron: (i) tunnels through the atomic/molecular potential barrier, (ii) then propagates in the continuum state, and (iii) recombines to the parent ion and converts its kinetic energy into the photon energy [1, 2]. The HHG spectra have a typical shape with a flat plateau ended by a cutoff; after that, the HHG intensity dramatically drops [1, 2].

In the past, many studies have been focused on atoms [2, 6, 7], and then expanded to symmetric molecules [3, 4, 8–12], whose HHG spectra contain only odd harmonics due to the time-spatial symmetry of the laser-atom/molecule system [13, 14]. Recently, much attention has been paid to asymmetric molecules such as  $\text{HeH}^{2+}$ , CO, OCS, HCl [15–22]. The HHG spectra of those molecules possess both odd and even harmonics due to the symmetry breaking of the laser-molecule systems [14, 17, 19, 22]. The odd-even HHG spectra can be applied for reconstructing the asymmetric molecular orbital [18], probing electron dynamics [17, 23–26], nuclear dynamics [27], and orientation degree of asymmetric molecules [28, 29]. Notably, in 2017, Hu *et al.* have first found the pure-even HHG spectra, i.e., the spectra contain only even harmonics when the laser electric field is perpendicular to the molecular axis of CO [19]. This finding is discovered theoretically by the time-dependent density functional theory. For other orientations, both odd and even harmonics appear in HHG spectra [19]. We have examined these results by the method of numerically solving the time-dependent Schrödinger equation (TDSE) [22]. Moreover, we have also indicated a non-trivial dependence of even-to-odd ratio, i.e., the ratio of intensity between the adjacent even and odd harmonics, on the molecular orientation [22]. Indeed, with the increasing of the orientation angle to  $90^\circ$ , the even-to-odd ratio for the parallel HHG decreases to zero, while this ratio for the perpendicular HHG grows up to infinity. There is a phase transition from the odd-even state to the pure-odd or pure-even state of HHG spectra when the orientation angle reaches  $90^\circ$ . It reflects a transition between the symmetry-breaking state into the symmetry one of the molecule-laser system.

Clearly, the even-to-odd ratio is strictly dependent on the molecular orientation controlling the symmetry of the molecule-laser system. However, the dependence of this ratio on the other asymmetric parameters such as the nuclear-charge ratio, or the permanent dipole is undiscovered. Therefore, in this paper, we investigate the influence of these asymmetric parameters, specifically, the nuclear-charge ratio, and the permanent dipole on the even-to-odd ratio of HHG spectra from the asymmetric molecule. For this purpose, we choose a simple model of an asymmetric molecule  $Z_1Z_2$  with one electron to investigate for easier adjustment of the nuclear charges. Despite the simplicity, this model still ensures the generality of real asymmetric molecules which are usually modeled as one active electron molecule in the theoretical investigation [18, 20, 22]. The HHG spectra are simulated by the TDSE method.

The rest of the paper is organized as follows. In Sec. II, we present the main points of the TDSE method for calculating the harmonic spectra of  $Z_1Z_2$  molecules and the analytical analysis of the transition dipoles responsible for the generation of odd and even harmonics. In Sec. III, we show our main results and discussion of the sensitivity of the even-to-odd ratio on the asymmetric parameters. A summary is given in Sec. IV.

## II. THEORETICAL BACKGROUND

In this section, first, we present the TDSE method for calculating the time-dependent wave function [20, 22] and, as a consequence, HHG emitted from the molecule in the strong laser pulse. Then, we present an analytical method to theoretically describe the conversion efficiency of odd and even harmonics in the HHG spectra from an asymmetric molecule.

### II.1. TDSE method for simulating HHG

In this paper, we study the model of asymmetric molecules  $Z_1Z_2$ , which consists of two nuclei and one electron. The molecule  $Z_1Z_2$  has diverse nuclear charges and internuclear distances. This model has been popularly used in many studies, such as Refs. [17, 18, 21, 23, 25, 28]. Despite its simplicity, this model is acceptable to mimic the HHG spectra from multielectron molecules. It is well known that for the interaction with intense laser pulses, a molecule can be described by the single-active-electron model [30, 31]. According to this model, only the HOMO electron interacts with the laser and with the effective potential created by the remaining electrons and the nuclei. Therefore, to control the molecular parameters and investigate their influence on the HHG spectra, using the two-center molecule  $Z_1Z_2$  with one active electron as a simplified model is quite appropriate.

The molecular model is presented in Fig. 1(a). The two nuclear centers are proposed to lie on the Oz axes. The center-of-charge coordinate system is used. The molecule is subjected to the laser field  $\mathbf{E}(t)$  with an orientation angle  $\theta$ , an angle between the electric vector and the molecular axis. In this paper, we study the case of  $\theta = 0^\circ$ . The magnitude of the electric field has the form of  $E(t) = E_0 f(t) \sin(\omega_0 t)$ , where  $E_0$ ,  $\omega_0$  respectively are the amplitude and carrier frequency;  $f(t)$  is the envelope function of the laser pulse. In this paper, we use the laser with the intensity of  $1.5 \times 10^{14}$  W/cm<sup>2</sup>, and the wavelength of 800 nm. To obtain sharper harmonic peaks, we use a long trapezoidal pulse of ten optical cycles with two cycles turns on and off, and eight cycles in the flat part, as shown in Fig. 1(b).

To obtain the HHG spectra, we utilize the TDSE method, i.e., the temporal wave function is numerically calculated from the time-dependent Schrödinger equation. It can be written in the atomic units of  $\hbar = e = m_e = 1$  as following

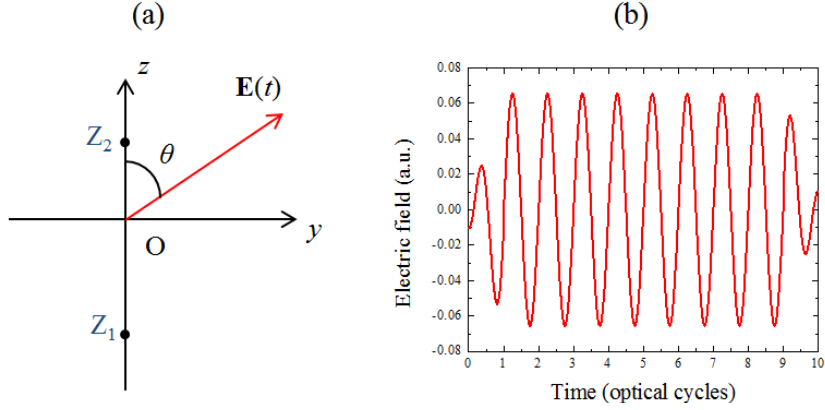
$$i \frac{\partial}{\partial t} \psi(\mathbf{r}, t) = \left( -\frac{1}{2} \nabla^2 + V(\mathbf{r}) + \mathbf{r} \cdot \mathbf{E}(t) \right) \psi(\mathbf{r}, t). \quad (1)$$

Here,  $\psi(\mathbf{r}, t)$  is the wave function of the active electron. The Coulomb potential  $V(\mathbf{r})$  has the following form

$$V(\mathbf{r}) = -\frac{Z_1}{\sqrt{r^2 - 2rz_1 \cos \alpha + z_1^2}} - \frac{Z_2}{\sqrt{r^2 - 2rz_2 \cos \alpha + z_2^2}}, \quad (2)$$

where  $Z_1$  and  $Z_2$  are effective charge of the two nuclei;  $z_1 = RZ_2/(Z_1 + Z_2)$  and  $z_2 = RZ_1/(Z_1 + Z_2)$  respectively are the coordinates of the two nuclei;  $R$  is the internuclear distance; and  $\alpha$  is the angle between the electron position vector  $\mathbf{r}$  and the axis Oz.

The time-dependent Schrödinger equation is solved by the procedure presented in detail in Refs. [20, 22]. Accordingly, the time-dependent wave function is found by the expansion of the field-free (time-independent) wave functions. The time-dependent coefficients of the expansion are then calculated by the evolution from the ground state. Since the crucial role of the ground



**Fig. 1.** Asymmetric molecular model  $Z_1Z_2$  (a) and the laser pulse used in the simulation (b).

state in the harmonics generation [19], we eliminated other excited states in constructing the initial state. After getting the time-dependent wave function, the induced dipole is calculated by the formula

$$\mathbf{d}(t) = \langle \psi(\mathbf{r}, t) | \mathbf{r} | \psi(\mathbf{r}, t) \rangle. \quad (3)$$

The HHG spectra are proportional to the Fourier transform of the induced dipole acceleration

$$S(\omega) = \left| \int \hat{\mathbf{e}} \cdot \ddot{\mathbf{d}}(t) e^{-i\omega t} dt \right|^2, \quad (4)$$

where  $\hat{\mathbf{e}}$  is an unit vector. In our study, we are interested in only the parallel HHG, i.e., HHG with the polarization parallel to the electric field. The other with the perpendicular polarization is absent for the case of  $\theta = 0^\circ$  [19, 22]. The permanent dipole of the asymmetric molecule is calculated by

$$\mathbf{P} = \langle \psi(\mathbf{r}, 0) | \mathbf{r} | \psi(\mathbf{r}, 0) \rangle. \quad (5)$$

Here,  $\psi(\mathbf{r}, 0)$  is the ground-state wave function of the molecule in the absence of the laser field, which are calculated by the B-spline method in this paper. Since at  $t = 0$ , the molecule is symmetric about the  $z$ -axis, the permanent dipole is aligned on this axis.

For the numerical simulation, we use 50 partial waves, 180 B-spline functions, and a box with a radius of 150 a.u. with 360 grid points. To avoid the reflection due to a finite box, we utilize the  $\cos^{1/8}$  mask function beyond the radius of 90 a.u. The time step is 0.055 a.u. We limit the number of basis functions of the time-dependent wave function by truncating the maximum energy of the system to be about 6 a.u.

## II.2. Analytical expression of transition dipole

The HHG of the asymmetric molecule consists of both odd and even harmonics, as shown in Refs. [15–22]. To interpret the HHG intensity of the odd- and even-harmonic generation, Chen and Zhang have derived analytical expressions of the corresponding transition dipoles [17]. Here, we will briefly recall some relevant equations of this work.

According to the three-step model, the harmonic photon is emitted at the last step when the ionized electron recombines to the parent molecular ion [2]. Therefore, the HHG efficiency

is proportional to the transition dipole between the continuum and ground states, i.e.,  $S(\omega) \propto |D(\omega)|^2$  [4]. The transition dipole  $|D(\omega)|$  is defined by the equation

$$\mathbf{D}(\omega) = \langle 0 | \hat{\mathbf{e}} \cdot \mathbf{r} | \mathbf{p}(\omega) \rangle, \quad (6)$$

where  $|0\rangle$  is the wave function for the ground state, and the wave function for the continuum state  $|\mathbf{p}(\omega)\rangle$  is assumed to be a plane wave  $e^{i\mathbf{p}(\omega) \cdot \mathbf{r}}$ . The electron momentum  $\mathbf{p}(\omega)$  and the HHG frequency  $\omega$  are related by the dispersion formula  $|\mathbf{p}(\omega)| = \sqrt{2\omega}$ .

In Refs. [17, 18, 22], it has been shown that, for asymmetric molecules, the electron recombination in the gerade part of the ground state leads to the generation of the odd harmonics. Meanwhile, the recombination into the ungerade part is responsible for the emission of the even ones. These facts are equivalent to the conclusion that the odd and even harmonics result from the recombination of the ungerade and gerade parts of the continuum states into the ground state, respectively [17]. Therefore, the transition dipole separates into two components as

$$\mathbf{D}(\omega) = i \langle 0 | \hat{\mathbf{e}} \cdot \mathbf{r} | \sin(\mathbf{p}(\omega) \cdot \mathbf{r}) \rangle + \langle 0 | \hat{\mathbf{e}} \cdot \mathbf{r} | \cos(\mathbf{p}(\omega) \cdot \mathbf{r}) \rangle. \quad (7)$$

Here, the first component corresponds to the generation of the odd harmonics, while the second one is responsible for the emission of the even harmonics.

As shown in Ref. [17], the ground-state wave function of a two-atomic molecule can be assumed as a linear combination of the atomic wave functions

$$|0\rangle \equiv \psi(\mathbf{r}, 0) \propto ae^{-\kappa|\mathbf{r} + \hat{\mathbf{e}}_{z1}|} + be^{-\kappa|\mathbf{r} + \hat{\mathbf{e}}_{z2}|}, \quad (8)$$

where  $a = Z_1 / \sqrt{Z_1^2 + Z_2^2}$  and  $b = Z_2 / \sqrt{Z_1^2 + Z_2^2}$  are the contribution coefficients;  $\kappa = \sqrt{2I_p}$  with the ionization potential  $I_p$ . Substitute the wave function (8) into the Eq. (7), we obtain

$$\mathbf{D}(\omega) \propto (iG^o(\omega) + G^e(\omega)) \langle e^{-\kappa r} | \hat{\mathbf{e}} \cdot \mathbf{r} | \sin(\mathbf{p}(\omega) \cdot \mathbf{r}) \rangle, \quad (9)$$

where  $\langle e^{-\kappa r} | \hat{\mathbf{e}} \cdot \mathbf{r} | \sin(\mathbf{p}(\omega) \cdot \mathbf{r}) \rangle$  is similar to the transition dipole between the continuum and the ground states of an atom.  $G^{o/e}(\omega)$  are the molecular interference factors causing odd and even harmonics [17]

$$G^o(\omega) = a \cos(pz_1 \cos\theta) + b \cos(pz_2 \cos\theta), \quad (10)$$

$$G^e(\omega) = a \sin(pz_1 \cos\theta) - b \sin(pz_2 \cos\theta). \quad (11)$$

### III. RESULTS AND DISCUSSION

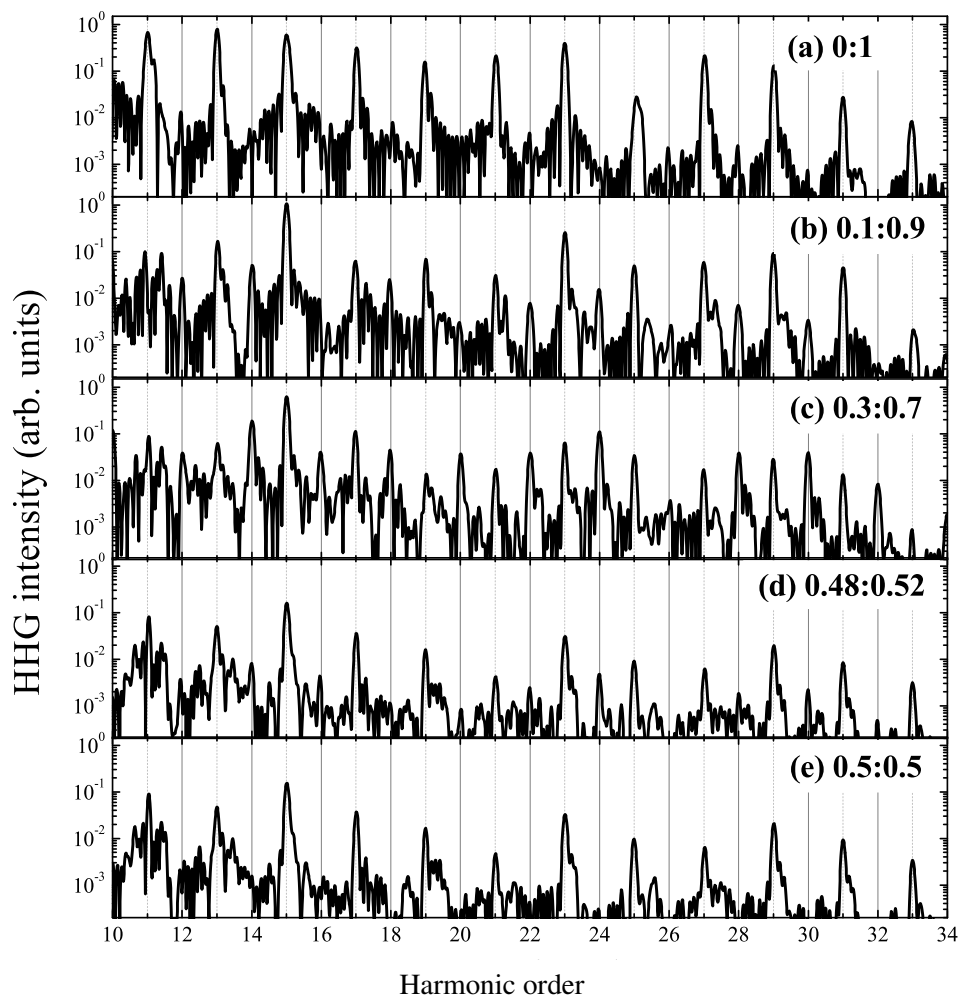
In this section, we present the HHG spectra emitted from the asymmetric molecule  $Z_1Z_2$  with different nuclear-charge ratios  $Z_1/Z_2$ . We need the ionization probability and the cutoff not being changed; thus, we also vary the total charge  $Z_1 + Z_2$  so that the ionization potential of the molecule, meaning the absolute value of the ground state energy, is fixed by the value of 0.515 a.u. This value is chosen similarly to that of the real molecule CO.

#### III.1. Dependence of even-to-odd ratio on nuclear-charge ratio

##### *The case of internuclear distance $R=2$ a.u.*

First, we consider the case of the asymmetric molecules  $Z_1Z_2$  with the internuclear distance of  $R = 2$  a.u. The HHG spectra are exhibited in Fig. 2 for molecules with different nuclear-charge ratios. It is shown that the correlation between the intensities of odd and even harmonic orders in HHG spectra is strongly dependent on the  $Z_1/Z_2$  ratio. Indeed, for the case  $Z_1 : Z_2 = 0 : 1$ ,

when the molecule becomes an atom (symmetric), the HHG spectra contain only odd harmonics, as indicated in Fig. 2a. With the increase of the nuclear-charge ratio, the intensity of the even harmonics first gradually enhances (Fig. 2b), then becomes comparable to, and even exceeds the intensity of the odd ones (Fig. 2c). After that, with the ratio  $Z_1/Z_2$  continuing to increase, the intensity of even harmonic orders reduces (Fig. 2d) and is completely depressed at  $Z_1 = Z_2$  when the molecule becomes symmetric (Fig. 2e).

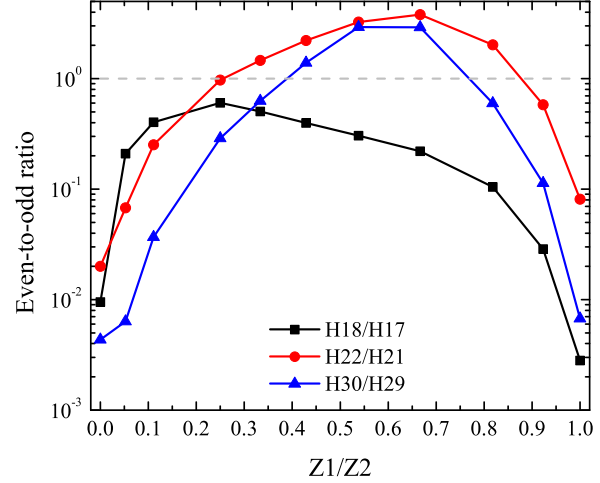


**Fig. 2.** Odd-even HHG spectra from the molecule  $Z_1Z_2$  with different ratios  $Z_1/Z_2$  calculated by the TDSE method. The molecule is chosen with internuclear distance  $R = 2$  a.u. and ionization potential  $I_p = 0.515$  a.u. The HHG spectra contain only odd harmonics for the cases: (a)  $Z_1 : Z_2 = 0 : 1$  (the molecule becomes an atom) and (e)  $Z_1 : Z_2 = 0.5 : 0.5$  (the molecule is symmetric). For other ratios  $Z_1/Z_2$ , the HHG spectra possess both odd and even harmonics.

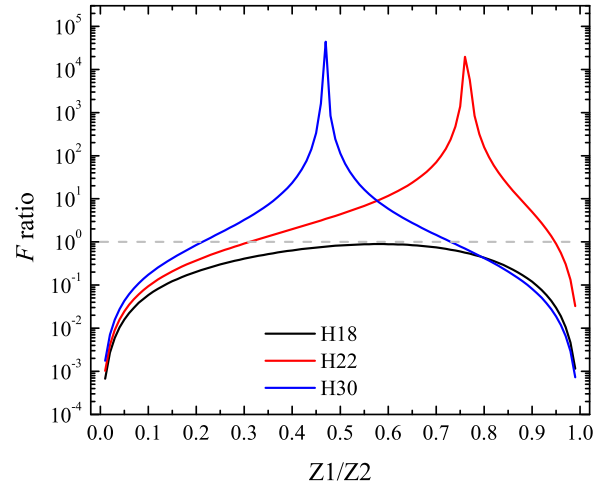
The same results are also presented in Fig. 3 in another way for a clearer illustration. We calculate the ratio between the intensities of each selected pairs of adjacent harmonics (with different parity), i.e., of the even and the nearest odd harmonic neighbors. We call it the even-to-odd ratio and plot it as a function of the ratio  $Z_1/Z_2$  in Fig. 3. The figure shows that there is no sudden phase jump from pure-odd spectra into the odd-even one. Indeed, with increasing the ratio  $Z_1/Z_2$ , the even-to-odd ratio first gradually grows up from zero; then, after reaching a maximum, the even-to-odd ratio drops to zero again when  $Z_1/Z_2 = 1$ . For harmonics in the middle of the plateau and near cutoff of HHG spectra, the maximum even-to-odd ratio is achieved when the  $Z_1/Z_2$  ratio is about  $0.5 \div 0.7$ . Clearly, there is a gradual transition between the odd-even state to the pure-odd state of HHG spectra.

From the above discussion, we infer that the even-to-odd ratio of the HHG intensity as a function of  $Z_1/Z_2$  reflects the parity of the molecule-field system. For the cases  $Z_1/Z_2 = 0$  or  $Z_1/Z_2 = 1$ , the atom/molecule-field system is symmetric with respect to the spatial inversion ( $\mathbf{r} \rightarrow -\mathbf{r}$ ) combined with the temporal translation by a half carrier-wave period ( $t \rightarrow t + T/2$ , where  $T = 2\pi/\omega_0$ ). As a consequence, the HHG spectra contain only odd harmonics [13, 14]. For other ratios  $Z_1/Z_2$ , when the molecule becomes asymmetric, the symmetry mentioned above is broken that results in the generation of both even and odd harmonics [14, 17, 22]. Thus, the even-to-odd ratio strongly relates to the degree of symmetry breaking of the molecule-laser system.

To interpret in-depth the dependence of even-to-odd ratio on the nuclear-charge ratio  $Z_1/Z_2$ , we consider the analytical expression of interference factors presented in Eqs. (10) and (11), causing odd and even harmonics [17]. Since the HHG signal is proportional to the transition dipole as described in Subsec. II.2, we predict that the even-to-odd ratio of the harmonic intensity must



**Fig. 3.** The dependence of the even-to-odd ratio on the ratio  $Z_1/Z_2$  for harmonics in the plateau of HHG spectra obtained by the TDSE method. The dashed line presents the even-to-odd ratio equal to one. The molecular model is the same as used in Fig. 2.



**Fig. 4.** The ratio of interference factors of even and odd harmonics as a function of the ratio  $Z_1/Z_2$ .

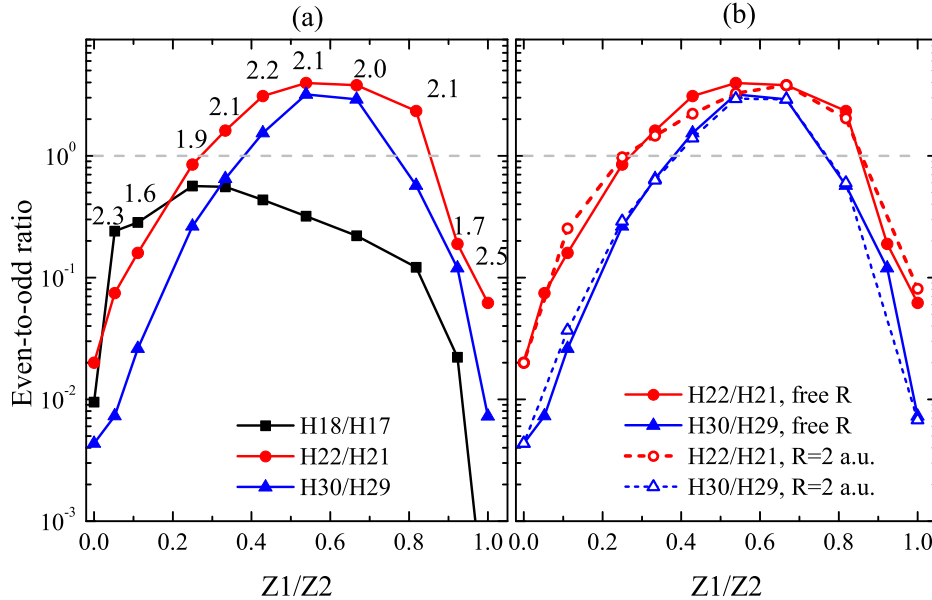
proportionate to the ratio defined as follows

$$F = \left( \frac{G^e(\omega)}{G^o(\omega)} \right)^2 = \left( \frac{Y \sin \frac{X}{1+Y} - \sin \frac{XY}{1+Y}}{Y \cos \frac{X}{1+Y} + \cos \frac{XY}{1+Y}} \right)^2 \quad (12)$$

with the notations:  $X = p(\omega)R$  and  $Y = Z_1/Z_2$ . If the even-to-odd ratio indeed relates to the ratio  $F$ , then we can see from Eq. (12) the straightforward dependence of the even-to-odd ratio on the ratio  $Z_1/Z_2$  and the factor  $X$ . In Fig. 4, we plot the ratio  $F$  as a function of the ratio  $Z_1/Z_2$  for some harmonics in the plateau. The figure indicates a similar tendency as for the even-to-odd ratio calculated by the TDSE method, as shown in Fig. 3. Specifically, with increasing the ratio  $Z_1/Z_2$ , the ratio  $F$  sharply increases, then approaches or exceeds one, before dropping dramatically.

It is noticed that for harmonics near the cutoff, the ratio  $F$  undergoes a sharp maximum due to the destructive interference effect and this occurs for odd harmonics and is absent in even ones [23, 26]. However, these sharp maximums are not seen in Fig. 3 due to the large step of the ratio  $Z_1/Z_2$  in our study. The interference effect in HHG spectra from asymmetric molecules is a complicated issue and will be investigated in our further studies.

### Changing the internuclear distance



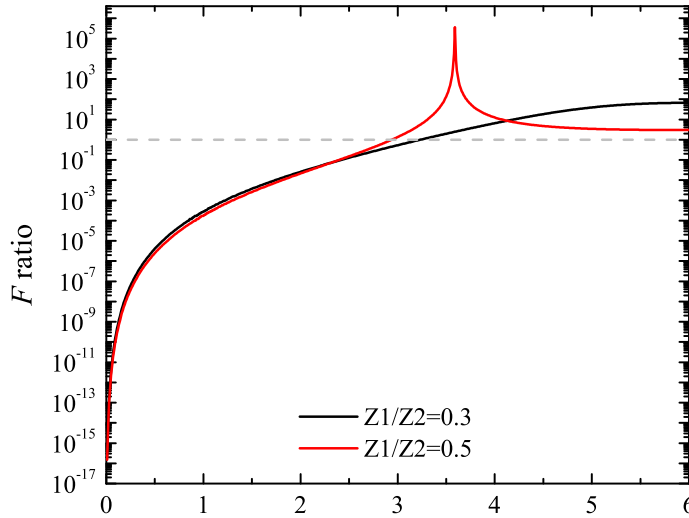
**Fig. 5.** The even-to-odd ratio of harmonics in the plateau of HHG spectra from molecule  $Z_1Z_2$  with arbitrary internuclear distance  $R$  (a) and the comparison with the case of  $R = 2$  a.u. (b). The values of  $R$  are enclosed for each point in the figure.

Now, we move to the case of the asymmetric molecule  $Z_1Z_2$  with the fixed ionization potential but with the internuclear distance changing, as a parameter, in the range from 1.6 a.u. to 2.5 a.u. Fig. 5 shows the dependence of the even-to-odd ratio on the ratio  $Z_1/Z_2$ . The value of



$R$  is added for each point in the figure. Surprisingly, this dependence is similar to the one presented in Fig. 3 for the case of internuclear distance  $R = 2$  a.u. This analogy demonstrates that the even-to-odd ratio is almost not sensitive to the changing of the internuclear distance.

To explain the above-mentioned result, we examine the ratio  $F$  of the interference factors of even and odd harmonics, see Eq. (12). Clearly, this ratio depends on the internuclear distance  $R$  through the factor  $X$ . In Fig. 6, we plot the ratio  $F$  as a function of  $X$ . It is shown that, after a sudden growth, the ratio  $F$  slowly changes with increasing of  $X$  value, except for a sharp maximum due to the destructive interference of odd HHG spectra [23, 26]. For harmonics in the plateaus of HHG spectra, and  $R$  values as indicated in Fig. 5, the  $X$  factor is ranged from about 2.5 a.u. to 5.5 a.u. In this range,  $F$  is almost stable, as shown in Fig. 6. Therefore, the even-to-odd ratio of HHG spectra from  $Z_1Z_2$  molecule is almost independent on the internuclear distance.



**Fig. 6.** The ratio of interference factors of even and odd harmonics as a function of  $X = p(\omega)R$

### III.2. Non-trivial dependence of even-to-odd ratio on molecular permanent dipole

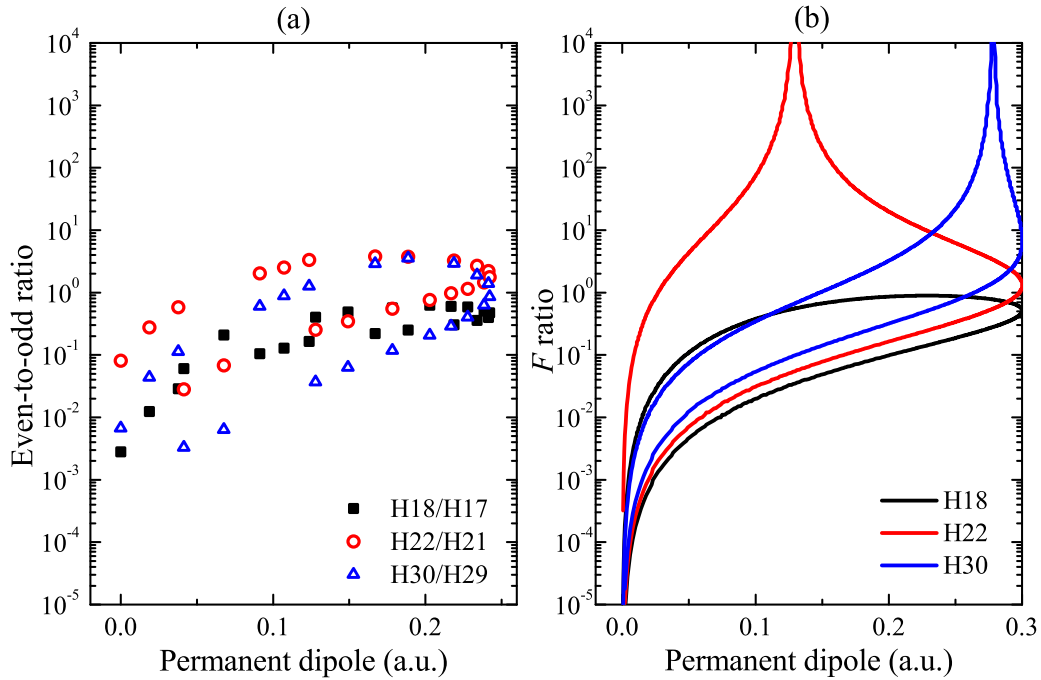
Finally, we consider the dependence of the even-to-odd ratio on the permanent dipole of the asymmetric molecules. To vary the permanent dipole, we change the initial wave packet by controlling the  $Z_1/Z_2$  ratio (see Eq. (5)). The internuclear distance and ionization potential are  $R = 2$  a.u. and 0.515 a.u., respectively. The even-to-odd ratio as a function of the permanent dipole calculated by the TDSE method is presented in Fig. 7(a). The results show that the even-to-odd ratio increases with increasing molecular permanent dipole. The dependence is not one-to-one, because there is one value of the permanent dipole corresponding to two values of the  $Z_1/Z_2$  ratio. Thus, the figure has a hysteresis-like shape where two even-to-odd ratios for the same permanent dipole can be distinguished.

To explain this anomalous dependence, we examine the analytical expression Eq. (12) for the ratio  $F$  of the interference factors. It clearly shows that the ratio  $F$  depends only on the ratio

$Z_1/Z_2$ , but not on the molecular dipole moment. We can also get an analytical formula for the permanent dipole  $P$  by substituting the molecular wave function Eq. (8) into Eq. (5) to obtain

$$P \approx -a^2 z_1 + b^2 z_2 = R \frac{Y(1-Y)}{(1+Y^2)(1+Y)}. \quad (13)$$

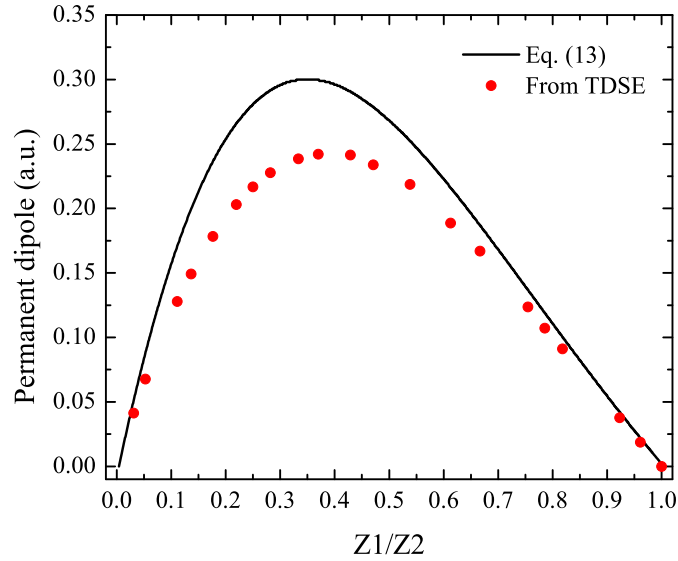
Here, for simplicity, we assume that the overlap between the atomic orbitals are negligible. It shows that the permanent dipole is controlled by the ratio  $Y = Z_1/Z_2$  only. To validate the analytical expression (13), we also calculate the permanent dipole numerically by solving the Eq. (1) without the laser field and put the solution into Eq. (5). The results are plotted in Fig. 8 as a function of the ratio  $Z_1/Z_2$ . For comparison, in Fig. 8, we also plot the analytical permanent dipole, calculated by Eq.(13). The figure shows that the dependencies of the permanent dipole on the ratio  $Z_1/Z_2$  calculated by both methods, numerical and analytical, agree with each other although the analytical values are a little bit higher than the numerical ones because of the omission of the wave function overlap. Therefore, the analytical expression Eq. (13) is reliable for us in our further explanation.



**Fig. 7.** The even-to-odd ratio (a) and the ratio of the interference factors (b) as a function of the molecular permanent dipole for harmonics in the plateau of HHG spectra. The molecular model is the same as used in Fig. 2.

Using Eqs. (12) and (13), we calculate the dependence of the ratio  $F$  on the permanent dipole and show it in Fig. 7(b). It is indicated that this dependence also forms the hysteresis-like curve like that of the even-to-odd ratio presented in Fig. 7(a), except for some special peak values

where the destructive interference effect occurs. Fig. 8 also helps us to understand the hysteresis-like shape of the even-to-odd ratio on the permanent dipole. Indeed, from Fig. 8 we can see two values of ratio  $Z_1/Z_2$  corresponding to one value of the permanent dipole. One value is small nearby the case of a (symmetric) atom-like molecule, and another one is near 1 for a symmetric molecule. In summary, we have properly explained the non-trivial dependence of the even-to-odd ratio on the molecular permanent dipole.



**Fig. 8.** The permanent dipole of the  $Z_1Z_2$  molecule calculated by the analytical formula Eq. (13) and by the numerical method by solving Eq. (1) in the absence of the laser field. The molecular model is the same as used in Fig. 2.

#### IV. CONCLUSIONS

In summary, we have studied the dependence of the even-to-odd ratio of harmonics in the plateau of HHG spectra emitted from the asymmetric molecule  $Z_1Z_2$  on the asymmetric parameters by the TDSE method. Besides, we have utilized the analytical model to interpret the obtained results.

Firstly, we have shown that the even-to-odd ratio strongly depends on the nuclear-charge ratio  $Z_1/Z_2$ . Indeed, with increasing of  $Z_1/Z_2$  ratio, the even-to-odd ratio first gradually increases, then drops to zero when  $Z_1/Z_2$  reaches 1. Moreover, we have claimed that the even-to-odd ratio is almost not sensitive to the changing of the internuclear distance. These results are well consistent with the analytical ratio of the interference factors of even and odd harmonics according to the strong-field approximation. In addition, we have indicated the non-trivial dependence of the even-to-odd ratio on the molecular permanent dipole of the asymmetric molecules and explained it related to the ratio  $F$  of the interference factor of the transition dipole.

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