Probing dynamic interference in high-order harmonic generation from long-range molecular ion: Bohmian trajectory investigation*

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Using Bohmian trajectory (BT) method, we investigate the dynamic interference in high-order harmonic generation from diatomic molecular ions. It is demonstrated that the main characteristics of the molecular harmonic spectrum can be well reproduced by only two BTs which are located at the two ions. This happens because these two localized trajectories can receive and store the whole collision information coming from all of the other re-collision trajectories. Therefore, the amplitudes and frequencies of these two trajectories represent the intensity and frequency distribution of the harmonic generation. Moreover, the interference between these two trajectories shows a dip in the harmonic spectrum, which reveals the molecular structure information.

Keywords: Bohmian trajectory, molecular high-order harmonic generation, dynamic interference

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1. Introduction

When atoms and molecules are irradiated by an intense laser pulse, high-order harmonics of the laser's frequency may be generated.[1–4] The mechanism of high-order harmonic generation (HHG) is a recombination of an ionized electron with its parent ion in the laser field.[5,6] HHG offers us a new way to detect this microscopic process with sub-angstrom spatial and attosecond temporal resolution.[7–9] HHG can also be applied to image molecular orbits,[10–12] and probe the electronic or nuclear dynamical behavior with attosecond resolution.[13–15]

The response of single atom or molecule of HHG has been studied theoretically by solving time-dependent Schrödinger equation (TDSE).[16,17] The TDSE calculation can provide an accurate result to simulate the experiment measurement. However, it is difficult to extract the dynamic information of the HHG process from the time-dependent wavefunction and to uncover the physical mechanism behind the process. To overcome this difficulty, one can adopt the BT scheme. Recently, this scheme has been used in research on the interaction between matter and a strong field. For example, by using this method, Takemoto *et al.*[18] investigated the attosecond electron dynamics of molecular ions, and Lai *et al.*[19] studied the correspondence between quantum and classical processes in strong field ionization. Furthermore, this scheme has been successfully applied to investigate the atomic HHG processes.[19–21] It is found that the result of the atomic HHG by solving the TDSE agrees well with that by the BT scheme.[21]

In this work, the BT method is applied to study the dynamic interference process in the molecular HHG processes. For the HHG of small linear molecules, there is a minimum in the harmonic spectra due to the interference of many atomic centers.[22–27] This minimum in the molecular HHG spectrum was first predicted theoretically by Lein *et al.*[16,17] and confirmed experimentally by Kanai *et al.*[23] The interference structure changes with the internuclear distance, the symmetry of molecular orbit, and the multi-orbit effect.[27,28] Thus, it is necessary to figure out the physical mechanism behind the interference structure in the molecular HHG spectrum.

To understand the interference mechanism, we investigate the HHG of H_2^+ molecular ion by using the BT scheme. It is found that the HHG can be reproduced qualitatively by using only two BTs, whose initial positions are located at the two atomic centers in the molecular ion. More importantly, the accurate interference structure in the molecular HHG spectrum can be illustrated through the dynamic analysis of the Bohmian particles' acceleration. These Bohmian particles play the role of an 'antenna' that can well reflect dynamic in-

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formation about the electron in the molecule.

2. Theoretical methods

We take the electron probability density as a fluid, and its flow can be analyzed by the Bohmian mechanics of many Bohmian particles whose trajectories are guided by the quantum wave-function.[17,29–31] The quantum wave-function $\psi(x,t)$ was obtained from the numerical solution of the one-dimensional TDSE (atomic units are used throughout this paper, unless otherwise stated): $\mathrm{i}\partial\psi(x,t)/\partial t = H\psi(x,t)$, where $H = -\partial^2/2\partial x^2 + V(x,R,t)$ is the Hamiltonian of the H_2^+ molecular ion. Here R is the distance between two protons, and x is an electronic coordinate in the molecular frame. Under dipole approximation and in length gauge, the time-dependent potential is

$$V(r,R,t) = V(x,R) + E(t)x,$$

where the soft-core potential

$$V(x,R) = -1/\sqrt{(x-R/2)^2+\alpha} - \sqrt{(x+R/2)^2+\alpha}$$

is chosen to represent the interaction between the electron and the nuclei, where α is the soft core parameter. The laser–electron interaction potential is $E(t)\cdot x$, where the laser's electric field is $E(t) = E_0 f(t)\sin(\omega t)$ with ω and E_0 being the frequency and peak amplitude of the laser pulse, respectively. The envelope of the laser's electric field is $f(t) = \exp[-4\ln 2((t-T/2)/\sigma)^2]$ with the total width being $T = 400$.

The TDSE is solved numerically by using the symmetrically splitting fast Fourier transformation scheme.[32,33] By using the time-dependent wave-function, the BTs $\{x_k(t)\,|k = 1,\ldots,N_{\mathrm{Tra}}\}$ are propagated by solving the following equation:

$$\dot{x}_k(t) = \mathrm{Im}\Big[\frac{1}{\psi(x,t)}\frac{\partial}{\partial x}\psi(x,t)|_{x=x_k(t)}\Big]. \tag{1}$$

The initial positions of the Bohmian particles are selected by the electron probability density of the ground state of H_2^+, and for each trajectory we assign the same weight. The following positions of these Bohmian particles are calculated from the integration of Eq. (1).[28] Furthermore, we can obtain the acceleration $a_k(t) = \ddot{x}_k(t)$ of the Bohmian particles by taking the derivative of Eq. (1). It should be noticed that, from a classical viewpoint, the square of the particle's acceleration is proportional to the instantaneous power of the radiation. Hence, the harmonic spectrum of each particle can be obtained by using the Fourier transformation scheme

$$P_k(\omega) = \Big|\frac{1}{t_\mathrm{f}-t_\mathrm{i}}\frac{1}{\omega^2}\int_{t_\mathrm{i}}^{t_\mathrm{f}} a_k(t)\,\mathrm{e}^{-\mathrm{i}\omega t}\,\mathrm{d}t\Big|^2, \tag{2}$$

where t_i and t_f are the initial and final instants of the laser pulse, respectively. The whole HHG of the molecular ion can be calculated from the Fourier transform of the acceleration of the total BTs

$$P(\omega) = \Big|\frac{1}{t_\mathrm{f}-t_\mathrm{i}}\frac{1}{\omega^2}\int_{t_\mathrm{i}}^{t_\mathrm{f}} a_{\mathrm{BT}}(t)\,\mathrm{e}^{-\mathrm{i}\omega t}\,\mathrm{d}t\Big|^2, \tag{3}$$

where $a_{\mathrm{BT}}(t) = \sum_{k=1}^{N_{\mathrm{Tra}}} a_k(t)/N_{\mathrm{Tra}}$.

For the purpose of comparison, we also calculate the HHG from TDSE using the time-dependent acceleration dipole

$$P_k(\omega) = \Big|\frac{1}{t_\mathrm{f}-t_\mathrm{i}}\frac{1}{\omega^2}\int_{t_\mathrm{i}}^{t_\mathrm{f}} a(t)\,\mathrm{e}^{-\mathrm{i}\omega t}\,\mathrm{d}t\Big|^2, \tag{4}$$

where

$$a(t) = \Big\langle\psi(x,t)\Big|\frac{\mathrm{d}V(x,R)}{\mathrm{d}x} - E(t)\Big|\psi(x,t)\Big\rangle. \tag{5}$$

3. Results and discussion

In order to explore the generation mechanism of the minima structure in the H_2^+ molecular HHG spectrum, we first calculate the BTs using Eq. (1). In the inset of Figure 1(a), we present the initial position of the Bohmian particles that are sampled from the electronic density distribution of the ground state, which is calculated from the H_2^+ molecular potential with $\alpha = 0.4$ and $R = 8.5$. In our calculation, the laser's frequency is $\omega = 0.057$, and the peak amplitude of the laser's electric field is $E_0 = 0.1$, where the shape of the laser's electric field is shown in the inset of Fig. 1(b). The BTs are shown in Fig. 1(a), where we may find that most of these trajectories are still located around the two atomic nuclei and a few trajectories are ionized after the laser pulse. In order to clearly distinguish the trajectories, we only present 400 trajectories in Fig. 1(a). One may ask: can these un-ionized trajectories play a role in HHG? If the answer is yes, then how do these bound BTs play roles in HHG? To answer these questions, we select two typical BTs which are initially located at $-R/2$ (BT(N)) and $R/2$ (BT(P)). The trajectory BT(N) is expressed in the cyan curve, and BT(P) is expressed in the green line in Fig. 1(a). We also present a re-collision trajectory (the red curve) in Fig. 1(a). For the purpose of comparison, in Fig. 1(b) we present the time-evolution picture of electronic probability density distribution by solving TDSE. We can find that the time-dependent electronic probability density agrees well with the evolution of the BTs. Under the dominance of the laser's electric field, the ionization mainly occurs at two moments ($t = 200$ and $t = 250$), which correspond to the peak positions of two half cycles of the laser electric field. Moreover, only for the ionization at about $t = 200$, the ionized electron has chance to come back to the nucleus (red line in Fig. 1(a)).

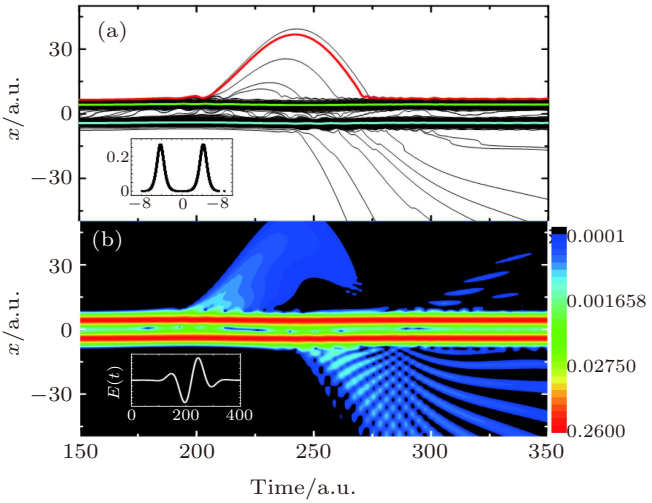


Fig. 1. (color online) (a) Time evolution of BTs; (b) time evolution of the electronic probability density and selected BTs: BT(N) and BT(P).

Using the two selected trajectories BT(N) and BT(P), we calculate the corresponding harmonic spectra by Eq. (2). The results are presented in Fig. 2(a), where the solid black line is the harmonic spectrum calculated from the trajectory BT(N), and the dotted red line is the harmonic spectrum calculated from the trajectory BT(P). It can be seen from Fig. 2(a) that the two harmonic spectra have almost the same intensities but they have different cutoffs. For the low-order region of the harmonic spectra, their harmonic structures are similar to each other. As the harmonic order is larger than 30, the difference between these two harmonic spectra increases with the harmonic order. As we calculate the total harmonic spectrum by summing up the contributions of these two trajectories, a clear minimum at about 33rd-order harmonic appears in the HHG spectrum caused by the interference between these two trajectories, as shown by the blue arrow in Fig. 2(b). This result qualitatively agrees with the numerical results from TDSE and that by summing up the contributions of all the BTs ($N_{\mathrm{Tra}} = 50000$), as shown by the dash-dotted green line and dotted red line in Fig. 2(b), respectively.

In order to explain why only two BTs can simulate the structure of the whole HHG spectrum, we investigate the dynamic behaviors of the two corresponding Bohmian particles and their accelerations. The time evolution of BT(N) and BT(P) is presented in Fig. 3(a). From Fig. 3(a) we can see that two particles oscillate with time, whose oscillation amplitudes are proportional to the change of electric field amplitude. The behaviors of the two Bohmian particles are similar at the beginning of the laser pulse and exhibit the difference at the moment of about $t = 200$. At this moment, in the trajectory BT(P) appears a fast oscillation with a frequency of about 10ω, while BT(N) moves smoothly. Furthermore, at moment $t = 250$, BT(P) moves smoothly while BT(N) oscillates. These oscillation mainly contributes to the low-order harmonics in the HHG spectrum, and its generation mechanism has been explained by Wang *et al.*[34]

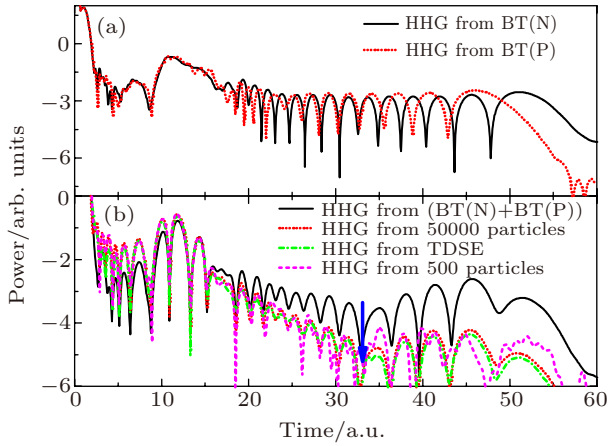


Fig. 2. (color online) Harmonic spectra from the diatomic molecule ion irradiated by the driving laser pulse, whose parameters are $E_0 = 0.1$ and $\omega = 0.057$: (a) calculated from the BT(N) and BT(P); (b) calculated from the TDSE and coherent sum of two or all Bohmian particles.

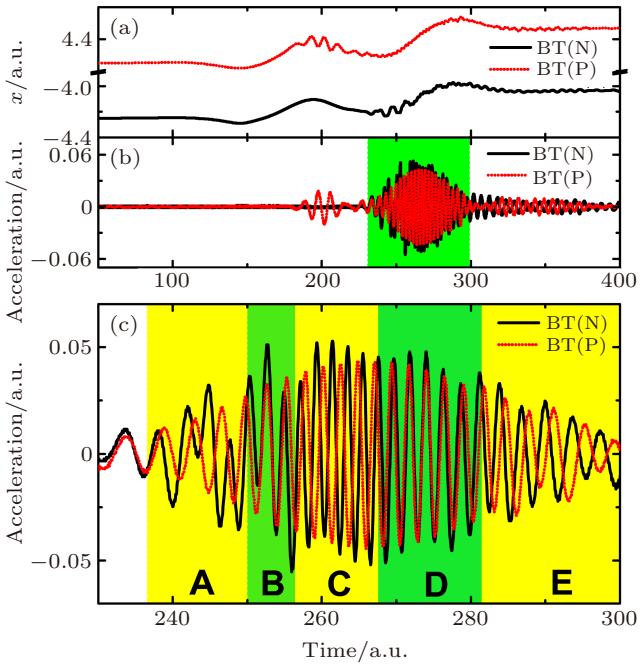


Fig. 3. (color online) Evolution of BTs BT(N) and BT(P): (a) time-dependent trajectories; (b) acceleration; (c) detailed acceleration.

Because the radiation of a particle is proportional to the module square of its acceleration,[35] we present the corresponding acceleration of two particles in Fig. 3(b). It can be clearly seen from Fig. 3(b) that around the instant 200, only BT(P) oscillates with a non-negligible amplitude, whose frequency is about 10ω. However, in the period of $t = 230$–300, a fast oscillation with high amplitude appears for both Bohmian particles with almost the same amplitude. In order to clearly investigate the dynamic interference profile of the HHG, we focus on the accelerations of the two Bohmian particles in the

period of $t = 230$–300, as shown in Fig. 3(c). From this figure we can see that the dipole accelerations of two particles have similar magnitude and oscillation frequency at every instant. However, their relative phase changes with time. In the regions 'A', 'C', and 'E', their accelerations exhibit opposite phases, while in the regions 'B' and 'D', their accelerations exhibit the same phases. From the viewpoint of the quantum interference, we may predict that when two oscillations have the same phases, the corresponding harmonic emission should be coherently enhanced. On the contrary, when the two oscillations have the opposite phases, the corresponding harmonic emission should be coherently reduced. To confirm our prediction, we perform the time-frequency analysis using wavelet transform for the Bohmian particles

$$A_\omega(t_0,\omega) = \int_{t_i}^{t_f} a_k(t) w_{t_0,\omega}(t)\,\mathrm{d}t, \tag{6}$$

where the wavelet kernel is $w_{t_0,\omega}(t) = \sqrt{\omega}W(\omega(t-t_0))$. Morlet wavelet[36,37] is used in this work.

It should be noticed that the dynamic emission profile of the harmonic can be clearly observed from the corresponding acceleration of the Bohmian particle. In Fig. 4(c), we present the time–frequency profile of the harmonic generated from the summation of BT(P) and BT(N). For the higher-order harmonics, in the period of $t = 230$–300, the interference pattern between these trajectories can be observed. Compared with time-dependent acceleration in Fig. 3(c), we find that in the time region of 'A', 'C', and 'E', where the two particles are out of phase, the harmonic intensity is low. On the contrary, in the period of 'B' and 'D', the Bohmian particles' accelerations are in phase, and hence the corresponding harmonic emission has higher intensity. Thus, we can clearly observe the dynamic interference profile of molecular HHG through the analysis of BTs. The time–frequency profiles of the harmonics generated from two Bohmian particles are shown in Figs. 4(a) and 4(b). In Figure 4(d), we calculate the time–frequency profile from the dipole calculated from the TDSE. Comparing Figures 4(d) with 4(c), we can find that, in the high-energy part, this dynamic profile of HHG agrees well with the result by the coherent summation of the two BTs.

The reason why the main characteristics of the molecular harmonic spectrum can be generated by only two BTs that are located respectively at the two ions is that the two localized trajectories can receive and store the whole collision information coming from all of the other re-colliding trajectories. The motion of Bohmian particle is not affected by classical force but is affected by the quantum force.[21] Therefore, using the information about the two BTs, we can detect the re-colliding process.

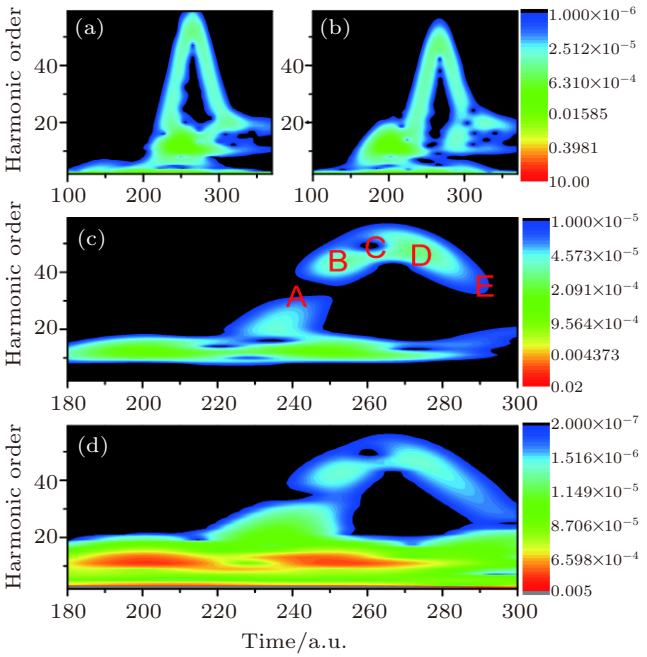


Fig. 4. (color online) Time-frequency behavior of harmonic emission calculated from: (a) BT(N); (b) BT(P); (c) coherent sum of BT(P) and BT(N); (d) TDSE.

4. Conclusion

In conclusion, utilizing a BT scheme, the dynamic coherent process of the molecular high-order harmonic generation is investigated in this paper. By analyzing the temporal characteristics of BTs, we find that the minimum in the molecular HHG spectrum is attributed to the interference of the BTs located at the two centers of molecule. The BT scheme can clearly reflect the dynamic process of harmonic interference, and can be taken as a detector to explore the dynamic process of complicated molecular system.

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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] Ferray M, L'Huillier A, Li X F, Lompré L A, Mainfray G and Manus C 1998 *J. Phys. B* **21** L31
[3] Popmintchev T, Chen M C, Popmintchev D, Arpin P, Brown S, Ališauskas S, Andriukaitis G, Balčiunas T, Mücke O D, Pugzlys A, Baltuška A, Shim B, Schrauth S E, Gaeta A, Hernández-García C, Plaja L, Becker A, Jaron-Backer A, Murnane M M and Kapteyn H C 2012 *Science* **336** 1287
[4] Lein M 2007 *J. Phys. B* **40** R135
[5] Corkum P B 1993 *Phys. Rev. Lett.* **71** 1994
[6] Lewenstein M, Balcou Ph, Ivanov M Y, L'Huillier A and Corkum P B 1994 *Phys. Rev. A* **49** 2117
[7] Salières P, Maquet A, Haessler S, Caillat J and Taïeb R 2012 *Rep. Prog. Phys.* **75** 062401
[8] Goulielmakis E, Schultze M, Hofstetter M, Yakovlev V S, Gagnon J, Uiberacker M, Aquila A L, Gulikson E M, Attwood D T, Kienberger R, Krausz F and Kleineberg U 2008 *Science* **320** 1614

[9] Krausz F, Ivanov M 2009 *Rev. Mod. Phys.* **81** 163
[10] Itatani J, Levesque J, Zeidler D, Niikura H, Pépin H, Kieffer J C, Corkum P B and Villeneuve D M 2004 *Nature* **432** 867
[11] McFarland B K, Farrell J P, Bucksbaum P H and Gühr M 2008 *Science* **322** 1232
[12] Haessler S, Caillat J and Salières P 2011 *J. Phys. B* **44** 203001
[13] Li W, Zhou X, Lock R, Patchkovskii S, Stolow A, Kapteyn H C and Murnane M M 2008 *Science* **322** 1207
[14] Smirnova O, Mairesse Y, Patchkovskii S, Dudovich N, Villeneuve D, Corkum P and Ivanov M Y 2009 *Nature* **460** 972
[15] Wörner H J, Bertrand J B, Kartashov D V, Corkum P B and Villeneuve D M 2010 *Nature* **466** 604
[16] Lein M, Hay N, Velotta R, Marangos J P and Knight P L 2002 *Phys. Rev. Lett.* **88** 183903
[17] Takemoto N and Becker A 2011 *J. Chem. Phys.* **134** 074309
[18] Lai X Y, Cai Q Y and Zhan M S 2009 *New J. Phys.* **11** 113035
[19] Wu J, Augstein B B and Faria C 2013 *Phys. Rev. A* **88** 023415
[20] Song Y, Li S Y, Liu X S, Guo F M and Yang Y J 2013 *Phys. Rev. A* **88** 053419
[21] Song Y, Guo F M, Li S Y, Chen J G, Chen G and Yang Y J 2012 *Phys. Rev. A* **86** 033424
[22] Lein M, Hay N, Velotta R, Marangos J P and Knight P L 2002 *Phys. Rev. A* **66** 023805
[23] Kamta G L and Bandrauk A D 2005 *P Phys. Rev. A* **71** 053407
[24] Kanai T, Minemoto S and Sakai H 2005 *Nature* **435** 470
[25] Han Y C and Madsen L B 2010 *J. Phys. B* **43** 225601
[26] Etches A, Gaarde M B and Madsen L B 2011 *Phys. Rev. A* **84** 023418
[27] Wu Y, Zhang J T, Ye H L and Xu Z Z 2011 *Phys. Rev. A* **83** 023417
[28] Han Y C and Madsen L B 2013 *Phys. Rev. A* **87** 043404
[29] Bohm D 1952 *Phys. Rev.* **85** 166
[30] Bohm D 1952 *Phys. Rev.* **85** 180
[31] Wyatt R E 2005 *Quantum Dynamics with Trajectories* (NewYork: Springer)
[32] Yang Y J, Chen G, Chen J G and Zhu Q R 2004 *Chin. Phys. Lett.* **21** 652
[33] Yang Y J, Chen J G, Chi F P, Zhu Q R, Zhang H X and Sun J Z 2007 *Chin. Phys. Lett.* **24** 1537
[34] Wang J, Chen G, Guo F M, Li S Y, Chen J G and Yang Y J 2013 *Chin. Phys. B* **22** 033203
[35] Cowan R D 1981 *The Theory of Atomic Structure and Spectra* (California: University of California Press)
[36] Chen J G, Zeng S L and Yang Y J 2010 *Phys. Rev. A* **82** 043401
[37] Chen J G, Yang Y J, Zeng S L and Liang H Q 2011 *Phys. Rev. A* **83** 023401