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# Theoretical exploration of the interference minimum in molecular high-order harmonic generation

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**Abstract.** The interference minimum in the molecular high-order harmonic generation is studied by numerically solving the non-Born-Oppenheimer approximation time-dependent Schrödinger equation. The results show that two kinds of interference minima appear in the high-order harmonic spectrum when the hydrogen molecular ion is exposed to the laser pulse. Furthermore, by adjusting wavelength of the laser as well as initial condition of nucleus, it is found that two kinds of interference minima are ascribed to the molecular structure and the electronic dynamics behavior induced by the laser field, respectively. Besides, the time-frequency distributions and the electron wave packet distributions are calculated to better understand the mechanism of the minimum.

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**Key words**: High-order harmonic generation, Interference minimum, Non-Born-Oppenheimer approximation

## 1 Introduction

Owing to the rapid advancement of laser technology, the dynamics of atoms or molecules exposed to an intense laser field [1-4] has become the subject of many theoretical and experimental investigations. Many nonlinear nonperturbative phenomena, such as charge resonance enhanced ionization (CREI) [5], above threshold ionization (ATI) [6] and high-order harmonic generation (HHG) [7, 8] have been discovered. In the past few years, high-order harmonic radiation has become a very active discussion in this field for the potential to produce ultrashort pulse (XUV) and X-ray source. The HHG process can be well understood by the semiclassical three-step model [9]: the electrons tunnel through

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the barrier formed by the Coulomb potential and the laser field, then they oscillate in the laser field, and finally may recombine with the parent ion and emit a harmonic photon with energy equal to the ionization potential plus the kinetic energy of the recombining electron.

In recent years, molecular harmonic generation has been intensively investigated, since it contains more physical information than atom. On the one hand, as there are two or more nuclei in molecules, the returning electron experiences the two-center or multicenter potential, which induces the interference effect on the emission process of molecular HHG other than the atomic case. On the other hand, the molecular HHG is sensitive to the rotational [10] and vibrational motion [11] of nuclei, which provides a measuring means of molecular dynamics in intense laser field. For instance, Lein found that the harmonic spectra generated from the molecules would be approximately relevant to the nuclei vibrational autocorrelation function [11]. Furthermore, Feng *et al.* proved that the harmonic emissions from the $H_2^+$ and $D_2^+$ were sensitive to the initial vibrational state, with more intense harmonics generated in the lighter isotopes [12]. In addition, the main discovery in aligned molecular HHG spectra was the double-slit-type interference effect [13]. This interference effect was experimentally observed in aligned $CO_2$ molecules by Kanai *et al.* [14] and this phenomenon was theoretically achieved in the simplest diatomic molecules $H_2^+$ and $H_2$ by Lein *et al.* [15]. Most of these studies indicate the importance of the nuclear motion and molecular structure in molecular dynamics. In fact, when molecules are irradiated by intense laser fields, it can be found that the interference effects have already appeared in many processes. To further research the interference effect which is caused by the molecular multicenter, the time-dependent Schrödinger equation (TDSE) is solved in this paper when a model $H_2^+$ is exposed to an intense pulse. Numerical results show that the minimum in the low-order region is largely dependent on both the nuclear mass and the initial vibrational level, which demonstrates that the minimum in the low-order region mostly derives from molecular structure. In contrast, the minimum in the high-order region is strongly dependent on the fundamental wavelength, which indicates that this minimum is associated with the dynamic process caused by laser.

The rest of this paper is organized as follows. The numerical method in this work is presented in Sec. II. The results and discussions of interference effects are presented in Sec. III. The conclusion of our paper is given in Sec. IV. Atomic units are used throughout this paper unless stated otherwise.

## 2 Theoretical method

In this paper, the theoretical calculation adopts the one dimensional model that electron and two nuclei were fixed in a straight line. The HHG can be studied by numerically solving the non-Born-Oppenheimer approximation (NBOA) time-dependent Schrödinger equation via the parallel quantum wave-packet computer code LZH-DICP