

## Efficient Grid Treatment of the Time Dependent Schrödinger Equation for Laser-Driven Molecular Dynamics

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**Abstract.** We present an efficient method to solve the time dependent Schrödinger equation for modeling the dynamics of diatomic molecules irradiated by intense ultrashort laser pulse without Born-Oppenheimer approximation. By introducing a variable prolate spheroidal coordinates and discrete variable representations of the Hamiltonian, we can accurately and efficiently simulate the motion of both electronic and molecular dynamics. The accuracy and convergence of this method are tested by simulating the molecular structure, photon ionization and high harmonic generation of  $H_2^+$ .

**AMS subject classifications:** 35Q41, 70H05, 70F07

**Key words:** Time-dependent Schrödinger equation (TDSE), Hydrogen molecule, non-Born-Oppenheimer approximation.

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## 1 Introduction

The interaction of atoms and molecules with intense ultrashort laser pulses has attracted increasing attention. Particularly, the high-order harmonic generation (HHG) [1, 2], above threshold ionization (ATI) [3] and dissociation [4], are extensively investigated in recent years. By using the high-order harmonics, attosecond coherent x-ray source has been produced [5, 6]. Such attosecond pulses make a breakthrough to image the motion of an electron inside atoms, molecules [7, 8] and lots of new applications are also developed [9–13]. For all of these investigations, an accurate and efficient theoretical model is of great importance to faithfully describe the electronic and molecular dynamics and to understand the underlying physics.

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The most accurate way to describe laser-atom and molecule interaction is *ab initio* solution of time-dependent Schrödinger equation (TDSE). This model has led to remarkable advancements in understanding the strong field phenomena. Nevertheless, this approach is quite computationally demanding because of the long range Coulomb potential and the high nonlinearity in the strong laser field. To accurately represent the Coulomb singularity, dense grid points near the nuclear origin must be employed. On the other hand, Coulomb effect is a long interaction force and also both HHG and ATI rely on a rescattering process [14]. According to this model, the electron is first ionized, then oscillates in the laser field and finally rescatters with the parent nucleus. The amplitude of electron motion is  $E_0/\omega_0^2$  [14], where  $E_0$  and  $\omega_0$  are the laser amplitude and wavelength, respectively. Therefore, to simulate the electron motion, the space region should be several times larger than  $E_0/\omega_0^2$ . Typically, a wide region of several tens angstrom or even larger is needed. Consequently, even with the state-of-the-art computer, it is still a big challenge to directly solve the full-dimensional TDSE for laser-atom interaction involving more than two electrons [15]. Fortunately, single-active-electron (SAE) is proven to be a good approximation and has been successfully utilized for describing the HHG, ATI and other phenomena. Within the SAE approximation, direct numerical solution of TDSE for laser-atom interaction has been well established. Nevertheless, compared with atoms, the molecules have additional degree of freedom and more complicated structure, the physical phenomena of molecular HHG [16–20] and ATI [23–25] are richer. These processes have attracted more and more interests because of the applications of imaging of molecular orbital [26]. Due to the extra internuclear motion, the response of molecules to strong fields usually depends on the structure and alignment of the molecules [16, 27], thus making it more complicated than that of atoms [17, 28]. Currently, the simulation is generally concentrated on the simple diatomic molecule. Although several numerical methods have been proposed for solving the TDSE, previous theoretical studies are mostly restricted either to the Born-Oppenheimer approximation [21, 22, 29–32] or to reduced-dimensionality TDSE [16, 17, 33], which however seems questionable to give an accurate results in quantity [34]. Whilst the nuclear motion was shown to play an important role in molecular HHG [17], therefore, the non-Born-Oppenheimer approach is indeed necessary for most simple molecules. In this work, to achieve a faithful theoretical model, we present an efficient way to *ab initio* solve the three dimensional TDSE without Born-Oppenheimer approximation, including both the electronic and molecular dynamics of diatomic molecules irradiated by intense ultrashort laser pulse.

## 2 Theoretical model

In this work, we focus on the interaction of diatomic molecule with a linearly polarized laser field. The simplest diatomic molecular ion  $H_2^+$  and its isotopes has been the basis of much study. Hence it is of special interest both in theory and experiment. Here we also set our model for treating  $H_2^+$ . The TDSE can be expressed as (atomic units (a.u.) are