REGULAR ARTICLE

Role of Excited States in Asymmetric Harmonic Emission

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Abstract: Role of excited states is theoretically studied in the asymmetric harmonic generation from quasi-asymmetric molecule LiH^{3+}. The calculated energies and time-dependent populations on the relevant electron states demonstrate that there is the laser-induced electron transfer between the ground state and the first few excited states, which is responsible for the observed multiple frequencies enhancements on the harmonic emission process. In addition, such electron transfer process is very likely to be general for the quasi-asymmetric molecules, as suggested by calculations for LiH^{3+} at different internuclear distances and with different central frequencies, pulse intensities, carrier-envelope phases, pulse durations as well as for the other quasi-asymmetric molecule BeH^{4+}.

1 Introduction

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With the development of the laser technology, the interaction between the intense laser pulses and the atoms [1,2] or the molecules [3,4] becomes an issue of wide interest and has received a lot of attention. High-order harmonic generation (HHG) as one of the most important nonlinear phenomena has been investigated for many years due to its potential applications in the generation of coherent UV attosecond pulses [5, 6] and in the time-resolved dynamics measurement of the atomic/molecular structure [7].

Currently, the HHG process can be described by the semiclassical recollision model [8] that consists of three steps: tunnel ionization-acceleration-recombination where both ionization and recombination processes occurred on the ground electronic state. Very recently, it has been found that, for some quasi-asymmetric molecules with permanent dipoles and multiple centers such as HeH^{2+} [9,10], not only the ground state but also the excited states can be recorded by harmonic emission, especially by the lower order harmonics. For instance, Bian *et al* found that there is one electron transfer process between the $1s\sigma$ ground state and the $2p\sigma$ excited state of HeH^{2+} on the harmonic emission process [11]. Chen *et al* discovered that there are two characteristic electron trajectories on each cycle harmonic emission caused by the general ground state and the excited state [12]. However, it remains unclear whether this excited state effect is the general characteristics of the quasi-asymmetric molecules.

Thus, in this paper, to work out the above issue as well as for better understand the excited state effect on the asymmetric molecules, we carried out a time-dependent Schrödinger equation (TDSE) investigation on the asymmetric harmonic emission from the two quasi-asymmetric molecules of LiH^{3+} and BeH^{4+}. Atomic units (a.u.) are used throughout this paper unless stated otherwise.

2 Computational aspects

In our numerical simulations, the HHG spectra are obtained by solving the time-dependent Schrödinger equation [13-18]. Here, we assume that the molecule is aligned along the laser polarization direction. Thus, the Hamiltonian is given by $H = \left[-\partial^2 / \partial x^2 + V(x) + xE(t) \right]$. Here, $V(x) = -Z_1 / \sqrt{(x+R_1)^2 + a} - Z_2 / \sqrt{(x-R_2)^2 + a}$ is the Coulomb potential, which can be described by the soft-core parameters (a=0.223 for LiH^{3+} and a=0.1249 for BeH^{4+}). Z_1 and Z_2 are the effective charges of the two nuclei (Z_1=3, Z_2=1 for LiH^{3+} and Z_1=4, Z_2=1 for BeH^{4+}) and R is the internuclear distance with R_1=[Z_2/(Z_1+Z_2)]R and R_2=[Z_1/(Z_1+Z_2)]R. The laser field is expressed as: E(t)=Eexp[-4ln(2)t²/τ²]cos(ω₀t+φ), where E, ω₀, τ and φ denote the amplitude, the frequency, the pulse duration and the carrier-envelope phase (CEP) of the laser pulse. Further, the harmonic spectrum (S(ω)) is obtained by Fourier transforming the dipole