LASER-INTENSITY DEPENDENCE OF HIGH-ORDER HARMONIC FROM EXCITED HYDROGEN MOLECULAR ION

PHAN THI NGOC LOAN[*], TRAN PHI HUNG[**], HOANG VAN HUNG[***]

ABSTRACT

High-order harmonic generation (HHG) spectra from molecule H_2^+ initially prepared in the three lowest electronic bound states, placing in an intense laser, have been obtained by numerically solving time dependent Schrödinger equation. It is found that the ratios of HHG intensities from these states strongly depend on the laser intensities. In addition, we showed that HHG emission is closely correlated with the quality $\gamma(t)$ characterizing the ionization process of. H_2^+

Keywords: high-order harmonic generation, efficiency, excited state, ionization.

TÓM TẮT

Sự phụ thuộc của tín hiệu sóng điều hòa bậc cao của ion phân tử hydro bị kích thích vào cường độ laser

Chúng tôi thu được phổ phát xạ sóng điều hòa bậc cao (HHG) của phân tử H_2^+ được chuẩn bị ở ba trạng thái liên kết thấp nhất, đặt trong trường laser mạnh, bằng việc giải số phương trình Schrödinger phụ thuộc thời gian. Kết quả chỉ ra rằng, tỉ lệ cường độ HHG từ các trạng thái này phụ thuộc mạnh mẽ vào cường độ của laser. Thêm vào đó, chúng tôi chỉ ra rằng phát xạ HHG tương quan chặt chẽ với đại lượng $\gamma(t)$ đặc trưng cho quá trình ion hóa của H_2^+.

Từ khóa: sóng điều hòa bậc cao, hiệu suất, trạng thái kích thích, ion hóa.

1. Introduction

High-order harmonic generation (HHG) emitted from atoms/molecules is one of fundamental processes in strong-field laser physics. The atoms or molecules exposed to an intense ultrashort laser emit HHG radiation at frequencies that are integer multiples (called HHG order) of the incident laser's frequency. The envelop of HHG spectra reveals a rapid decrease at the lowest frequencies, then follow by a large plateau with roughly constant intensity, and finally, this plateau region ends at a sharp cutoff. Coherent radiation HHG can be used as a tool to extract atomic/molecular structure [9], or to probe electronic [1,13] and nuclear dynamics [12].

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The HHG spectra are usually explained in terms of semiclassical three-step model, proposed by Lewenstein et al. [10]. In the first step, the active electron tunnels into the continuum. Next, the liberated electron is accelerated away, and then driven back towards the parent ion when electric field reverses its direction of polarity. At last, the electron recombines into ground state, thus generating HHG.

Within above-mentioned model, only the ground state was considered and taken into accounts. However, a full quantum results obtained by solving time-dependent Schrödinger equation (TDSE) indicated some effects that caused by the excited states. The predominant recombination into the excited states leads to the disappearance of minima in HHG spectra of H_2^+ at large internuclear distances [7]. Role of excited states is also theoretically studied for HHG from quasi-asymmetric molecule [5]. It is showed that the laser-induced electron transfers between the ground state and the excited states is responsible for the appearance of some enhancements peaks in emission spectra.

One aim of HHG investigation is to improve the efficiency of generated spectra. Since ground state is the lowest-energy state, so atoms in this state is difficult to be ionized. Thus, it is obvious that one way to improve HHG efficiency is using excited states [14], or coherent superposition of states [11,17]. In 2005, Paul et al. successfully generated HHG spectra from rubidium atoms in highly excited states by a weak laser [14]. The magnitude of observed spectra increases several orders in comparision with that of the ground state. Besides, the development of the optical frequency comb technology [3,6] has provided a powerful method to generate a coherent superposition of bound states. Inspired by the technical achievements, Wang et al theoretically solved TDSE for He^+-like atom, and showed that the efficiency of HHG generated from atom with initial superposition of ground and first excited states is significantly enhanced in comparison with excited states alone [17]. Latter, Milosevic [11] based on Strong Field Approximation (SFA) obtained numerical and analytical results of HHG for two-state hydrogenlike atom model. It is showed that spectra contain resonant part with peaks around excitation energy. Moreover, the plateau region of HHG from coherent two-state atom can be extended with high intensity in comparision with that of excited states.

In the above-mentioned investigations, efforts to improve the HHG efficiency were restricted to atoms [11,14,17]. Hence, in this work, we expand our object to more complicated one - molecular ion hydrogen H_2^+. On the other hand, in previous works [11,17], HHG from atoms with only ground and the first excited states are considered, the second and higher excites states are neglected. Thus, in this study, we theoretically study of HHG of H_2^+ molecule in ground, the first and the second excited states. For convenience, we denoted these states by numbers 0, 1, 2, respectively. It is found that the emission efficiency of HHG for those states depend on the probe laser-intensity. In

addition, the influence of ionization probabilities is also analyzed to decipher the dependence of HHG on laser intensities. These results will establish a basic for our further investigation of HHG with coherent superposition state of hydrogen molecular ion.

The paper is organized as follows. In Section 2 we describe numerical calculation method TDSE to compute the HHG of H_2^+ in ground and excited states. The obtained HHG for various laser-intensities is presented in Section 3. In this Section, we also discuss the correlation between the ionization probability and emission efficiency. Section 4 contains concluding remarks.

2. Theory and calculation method

After being ionized, electron moves along the electric vector of linearly polarized laser field. In addition, molecule H_2^+, as an electric dipole in an electric field, quickly aligns along the polarization vector of laser. Thus, we employ one-dimensional model (1D) to reduce the amount of calculations to solving the time-dependent Schrödinger equation.

In this Section the TDSE method for calculating HHG of H_2^+ in ground and excited states is presented. Because the movement of two nuclei is very slow compared to electronic motion, so we assume that the two nuclei are frozen with the internuclear separation 2.0 a.u. The time-dependent Schrödinger equation of molecule H_2^+ interacting with the laser pulse has the following form (atomic units with $\hbar = e = m_e = 1$ are used throughout)

$$i\frac{\partial}{\partial t}\Psi(x,t) = \left(-\frac{\partial^2}{2\partial x^2} + V_C(x) + V_L(x,t)\right)\Psi(x,t), \quad (1)$$

where x is the electronic coordinate with respect to the center mass. The soft Coulomb potential of molecule $V_C(x)$ is

$$V_C(x) = \frac{1}{2} - \frac{1}{\sqrt{(x-1)^2 + a}} - \frac{1}{\sqrt{(x+1)^2 + a}}. \quad (2)$$

Here the soft-Coulomb parameter $a = 1.55$ is added to avoid the singularities and to mimic the real 3D potential energy of H_2^+ in the ground state. The ionization potentials are 29 eV, 19 eV and 12 eV for ground state, the first and the second excited states, respectively at R = 2.0 a.u. These values are in good agreement with the real ones, which are well known in literature [16].

The electron-field interaction $V_L(x,t)$ in length gauge is taken of the form $V_L(x,t) = x.E(t)$, where

$$E(t) = E_0 \sin^2\left(\frac{\pi t}{\tau}\right)\sin(\omega_0 t + \varphi), \tag{3}$$

E_0, τ, ω_0, and φ are the peak field amplitude, pulse duration, laser carrier frequency and carrier-envelope phase, respectively. Laser 800 nm and $\varphi = 0^o$ are used in all calculations.

Initially, the molecular ion H_2^+ is prepared in state *n*. The initial wave function of the n^{th} state of H_2^+ is found using the imaginary time propagation method [8]. During the interaction with laser, the time-propagating wave function is given by applying the second-order split operator technique [4]

$$\begin{aligned}\Psi(x,t+\Delta t) \approx \exp\left(-i\frac{V(x,t)}{2}\Delta t\right)\exp\left(-i\hat{T}\Delta t\right) \\ \times\exp\left(-i\frac{V(x,t)}{2}\Delta t\right)\Psi(x,t) + O\left(\Delta t^3\right),\end{aligned} \tag{4}$$

where $V(x,t) = V_C(x) + V_L(x,t)$ is the total potential, and $\hat{T}$ is the kinetic energy operator of the system. In this work, the time-step is $\Delta t = 0.027$ a.u. We use a numerical grid of 1600 a.u. with step-size of 0.1 a.u. All parameters have been checked for the convergence of the results.

In this work, HHG spectra are calculated in acceleration form $\mathbf{a}(t) = -\langle\Psi(x,t)|\nabla V_C + \mathbf{E}|\Psi(x,t)\rangle$. The HHG spectrum $S_{HHG}(\omega)$ can be obtained through Fourier transforming of this acceleration

$$S_{HHG}(\omega) = \left|\int_{-\infty}^{+\infty} a(t)\, e^{-i\omega t} dt\right|^2. \tag{5}$$

The time-dependent ionization probability can be computed as

$$P(t) = \int_{S_i} |\Psi(x,t)|^2 dx, \tag{6}$$

where S_i is the ionization region and defined as $S_i : -20\text{ a.u.} \le x \le 20\text{ a.u.}$

3. Result and discussion

In this work, we use a 5-optical cycles (13 fs duration) laser pulse with sine-square envelope and wavelength of 800 nm. The intensity of laser is chosen as

5×10^{13} W/cm^2 (weak laser), 2×10^{14} W/cm^2 (intermediate laser) and 1×10^{15} W/cm^2 (strong laser). The electric field $\mathbf{E}(t) = -\partial\mathbf{A}(t)/\partial t$ and the vector potential $\mathbf{A}(t)$ of such a laser pulse are depicted in Fig.1.

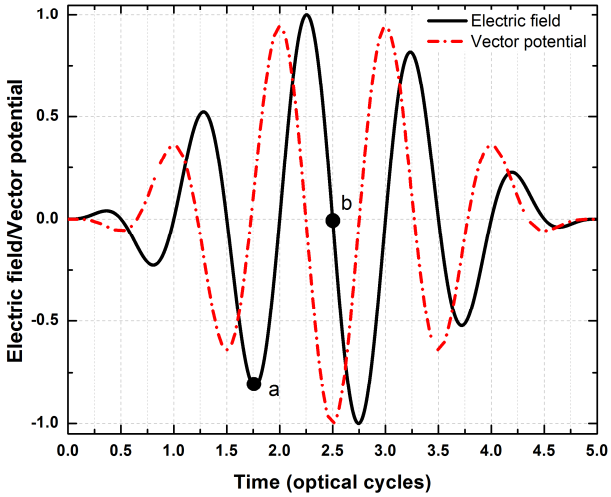

Fig.1. *Electric field and vector potential for a 5-cycle duration laser pulse with wavelength of 800 nm.*

The HHG spectra from H_2^+ placed in short pulse lasers are numerically obtained and depicted in Fig.2. Generally, the HHG spectra of H_2^+ in all cases exhibit a typical feature of plateau and cutoff. When H_2^+ interacts with laser with intensity of 5×10^{13} W/cm^2, the cutoff of spectra are $29\omega_0$, $21\omega_0$, and $17\omega_0$ for the ground state, first and second excited states. For laser of 2×10^{14} W/cm^2, these values are $45\omega_0$, $39\omega_0$, and $37\omega_0$, respectively. It is easily understood that the ground state has larger ionization potential than the excited state's one, which leads to the flattening of the plateau region of spectrum. This result is quite identical with that predicted by semiclassical three-step model. Moreover, it is found that the spectra of excited states exhibit double-plateau structure when molecule exposed to the strong laser field 1×10^{15} W/cm^2. The cutoffs of the first and second plateau regions are $139\omega_0$, and $103\omega_0$ (arrows in Fig.2c$_1$) for these states. The double-plateau was observed for atomic case He^+ in [17], now we also found for molecule H_2^+ in strong laser field. This phenomenon was can be understood that there are many trajectories that electron can recombine with parent ion at different time, leading to different cutoffs. These trajectories contribute to the harmonics with distinct strength, thus creating the double-plateau in spectrum [17].

Now we pay attention to the efficiency of HHG spectra (Fig.2, left panels). It is shown that the HHG intensities from ground state, first and second excited states are significantly different and strongly depend on the laser-intensity parameter. In the weak laser field 5×10^{13} W/cm^2, the magnitude of HHG of H_2^+ with the first excited state is about three orders higher than that of the ground state, and one order less than that generated from the second excited state. With higher intensity at 2×10^{14} W/cm^2, the emission efficiency of the first excited state reveals stronger than that of two other. Specifically, its magnitude is 1.5 and 5 orders higher than that of the second excited and ground states, respectively. For strong field with laser-intensity 1×10^{15} W/cm^2, in contrast to the previous cases, the HHG intensity of spectra of both two excited states are lower than that of ground state. Obviously, sorting the state's HHG magnitude is differently expressed in dependence on strength of laser field.

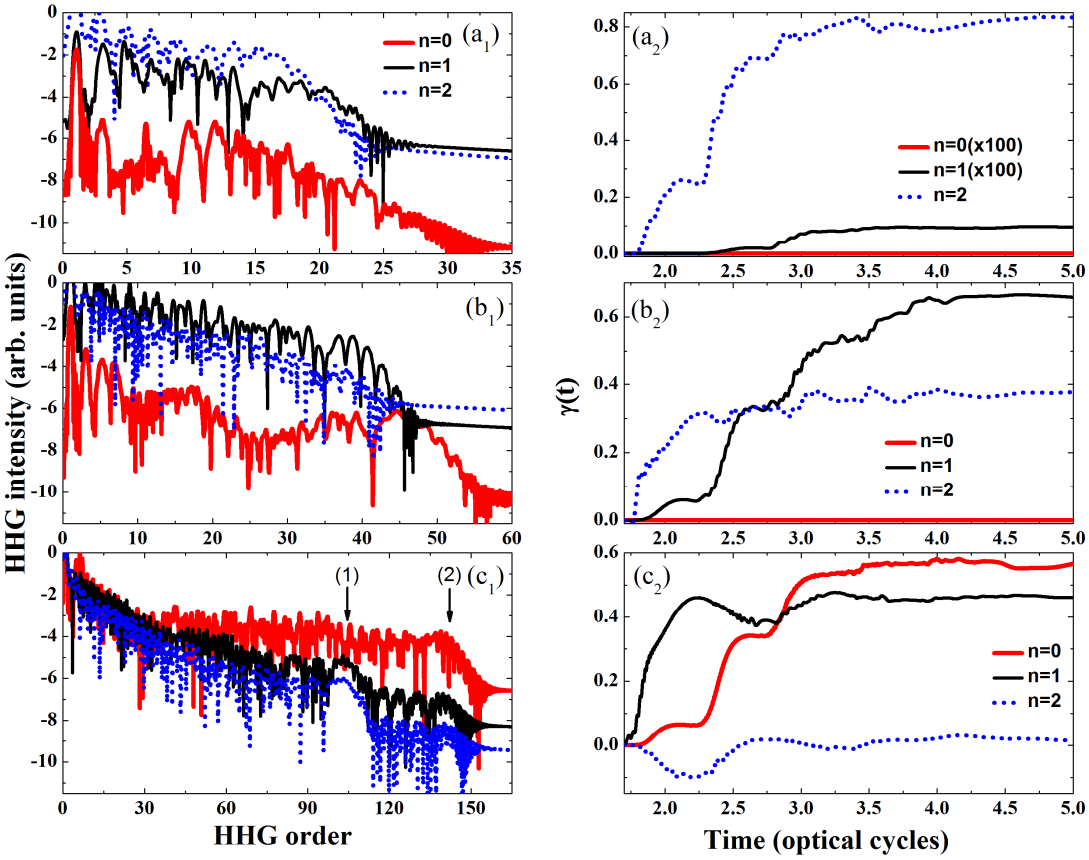

Fig.2. *The HHG spectra (left panels) and the corresponding ionization probabilities (right panels) of H_2^+ from the ground n=0 (thick line), first n=1 (thin line) and second n=2 (dot line) excited states. The laser with intensities of 5×10^{13} W/cm^2(a_1, a_2), 2×10^{14} W/cm^2(b_1, b_2) and 1×10^{15} W/cm^2(c_1, c_2) is used.*

To elucidate the above-mentioned effects, we calculated the time-dependent ionization probabilities of these states through equation (6). Physically, the HHG efficiency is directly proportional to the ionization probability. In addition, within classical treatment [2,15], the authors have proven that the electron will never return back and recombine to the parent ion if it is ionized before the peaks of laser. Only electron born at time near "a" (Fig.1) can return to the target at time near the maximum of vector potential "b". Hence, we evaluated only a part of ionization probability, which is directly contributed to the HHG generation via $\gamma(t) = P(t) - P(\mathrm{a})$. Numerical calculation of this quantity is plotted in Fig.2 (right panels). More clearly, we multiply these probabilities by appropriate factors. It is easy to see that the state's order of $\gamma(t)$ in most of the time is identical with order of corresponding emission efficiencies.

We consider for the case of weak laser with intensity of $5\times10^{13}\,\mathrm{W/cm^2}$ (Fig.2a_2). The transition from the ground state to the continuum is negligibly small, that the emission intensity is low. With increasing quantum numbers of bound states, the ionization probabilities are stronger, thus leading to more efficient emitted harmonics. For higher laser field with intensity $2\times10^{14}\,\mathrm{W/cm^2}$ (Fig.2b_2), it is shown that 66% electron ionized from first excited state can return and recombine to the target while for the second excited state it is smaller, 37%. Therefore, H_2^+ prepared in the second excited state generates less efficiency spectrum than that in the first excited state. Finally, for the sufficient strong field $10^{15}\,\mathrm{W/cm^2}$, the HHG spectra reveal a distinguished behavior with the most efficiency emission from H_2^+ in the ground state. In this state, since the ionization probability reaches 56% (Fig.2c_2), much larger than that for lower laser field, leading to higher HHG intensity. In strong field, the recombination probability of ground state is significantly larger 57% in compared with 46% for the first and 1.4% for the second excited states. The excited states are almost depleted before the laser pulse reaches its maximum. Consequently, H_2^+ in ground state emits higher HHG than those in excited states.

4. Conclusions

In this paper, we have systematically studied the HHG of H_2^+ initially prepared in the three lowest states exposed to intense laser. It is shown that the ratio of HHG intensity from the three lowest states is strongly sensitive to the intensity of the laser field. The HHG spectra exhibit different behavior in various range of laser-intensity. Importantly, influence of ionization probability on HHG is also analyzed. The two main factors contribute to the HHG spectrum are (i) ionization, and (ii) recombination processes. In addition, we have showed a closely relation between the HHG efficiency and the quantity $\gamma(t)$, which is proportional to the recombination process. For strong laser field, the excited states are almost depleted before the laser pulse reaches its

maximum, leading to the lower-intensity emission in compared with ground state's one. Base on these results, the study of highly efficient generation HHG from H_2^+ with coherent superposition of ground and excited states will be further carried out.

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