Journal of Atomic and Molecular Sciences

Global Science Press

ISSN 2075-1303

Progress of selection rules in high-order harmonic generation

Received: April 20, 2017,
Accepted: June 15, 2017,

DOI: 10.4208/jams.042017.061517a

http://www.global-sci.org/jams/

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Abstract. The high-order harmonic generation (HHG) has attracted much attention due to its wide application in attosecond science in last decades. The selection rules have also been broadly studied in experiments and theories since they play an important role in HHG. In this review, we give an overview of recent developments on selection rules of HHG from atoms to molecules. For targets with rotational symmetries, if the rotational symmetries of targets and laser pulses are the *M*-fold (the projective symmetry on the laser polarization plane) and *L*-fold, the selection rules are *Nk*±1, where *N* is the the greatest common divisor of *M* and *L*. However, for asymmetric molecules in non-Born-Oppenheimer approximation, the situation is more complicated, where the nuclear dipole acceleration can produce even harmonics, but it is three orders lower than that of the electron. Hence, the HHG is mainly relied on the electronic dipole acceleration. In this case, the broken degree of system-symmetry dominates the generation of even-order harmonics.

1. Introduction

When atoms or molecules interact with strong laser fields, many interesting intense field phenomena will occur, such as high-order harmonic generation (HHG) [1-10], non-sequential double ionization [11], below-threshold harmonic generation (BTH) [12-16], multiple ionization [17,18], above-threshold ionization [19], and Coulomb explosion (CE) [20], and so on. These phenomena deepen on our understanding of dynamic mechanisms in the interaction between atoms or molecules and intense fields.

The HHG attracts a lot of attention since it provides a new type of coherence light source of ultra-short wavelength. Many fancy phenomena and applications have also been found in last decades [21-26]. One of the most important achievements is that the HHG supplies us an important avenue to generate ultra-short attosecond laser pulses [27, 28], which pushes the investigations on the ultra-fast measurement from the femtosecond magnitude to the attosecond magnitude [26].

The step of HHG developments dramatically increased in 1980s. Many groups successfully observed HHG from gaseous targets in experiments [29-34]. However, the early experiments only observed the harmonics with few orders. In 1993, Macklin *et al*. [33] firstly found that harmonics beyond 100 orders could be produced by a laser field with the wavelength of 806nm and the peak intensity $\geq 10^{15}$ W/cm^2. At the same year, L'Huillier *et al*. also reported the generation of 135th harmonics from Ne driven by a 1ps, 1.06μm laser pulse [34]. These findings indicate the potential possibility for the high efficient generation of coherent radiation at the extreme ultraviolet region (XUV).

After the HHG potential applications were discovered, the HHG has been widely investigated [35-37]. So far the physical process of HHG can be well understood by the semi-classical three-step model [38]: ionization, acceleration and recombination. Moreover, the harmonic spectra produced from the atoms in a multi-cycle laser pulse present some common features: (i) the harmonic spectrum consists of three parts: a fast drop in the low-order yield, following a plateau and a sharp cutoff [39], the cutoff energy is around $I_p+3.17U_p$, where I_p is the ionization energy of the atom and U_p is the ponderomotive energy [40], (ii) only odd harmonics are produced [41-43]. In order to explain the disappearance of even harmonics, the early explanation is given upon the foundation of a perturbative analysis about the non-linear optical susceptibilities [44]. However, since perturbation theory has been broken down in so strong fields, one may expect that a more reasonable explanation can be proposed. Then a non-perturbative proof which invokes the concept of inversion symmetry is given by Ben-Tal et al. [45]. In this theory, they concluded that when the system possesses inversion symmetry, the selection rules for HHG are 2k±1 (k=1,2,3,...), that is to say, the harmonic spectrum is composed of only odd harmonics.

Molecules have more degrees of freedom than that of atoms, thus the selection rules for the molecular high-order harmonic generation (MHOHG) are more complex. Specifically, in a circularly polarized (CP) laser pulse, the allowed harmonics for molecular targets are determined by the rotational symmetries of molecules. If a molecule possesses M-fold rotational symmetry, the allowed harmonic orders in one CP laser field are kM±1 (M is a positive integer and k=0,1,2,...), in which the efficiency of HHG is low [46]. In a linearly polarized (LP) laser field, the molecules with inversion symmetry obey the same selection rules as atoms [47]. Very recently, the group of professor Peixiang Lu [47] investigated the selection rules in HHG from more complicated molecules driven by different laser fields and got an important conclusion: the allowed harmonic orders can be directly judged by the associated rotational symmetries (ARS) of the target-laser system. For the stereoscopic targets, the ARS is determined by the symmetry of the projection of the targets rather than by the symmetry of the targets itself. For the laser pulse, the symmetry contributing to ARS can be decided by the symmetries of the Lissajous figure and its dynamical directivity [47]. Recently, the circularly polarized molecular high-order harmonics have been generated due to the

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optional selection rules of HHG in a bicircular laser [48, 49]. However, for asymmetric molecules in non-Born-Oppenheimer approximation, the situation is confusing. In 2001, Kreibich et al. firstly found that a model HD molecule can produce intense even harmonics in non-Born-Oppenheimer approximation [50]. Then the even harmonics were also observed from asymmetric molecules in Born-Oppenheimer approximation [51]. These works seemed to indicate that the even-order harmonics could appear as long as the systemic symmetry was broken regardless of in Born-Oppenheimer approximation or non-Born-Oppenheimer approximation. Nevertheless, in 2016, Du et al. found that the HD molecule still generated only odd harmonics in non-Born-Oppenheimer approximation though the generation of even harmonics is possible in principle [52]. Then Yue et al. investigated the HHG for HD^+ and HeH^{2+} in non-Born-Oppenheimer approximation and found that HD^+ generated only odd harmonics while HeH^{2+} generated both odd and even harmonics [53]. And they invoke a concept of broken degree of system-symmetry to interpret the different odd-even property between the harmonic spectra of asymmetric molecules HD^+ and HeH^{2+} [53].

The purpose of this review is to give an account of the history and recent status of the studies on the selection rules for HHG from atoms or molecules in Born-Oppenheimer approximation and non-Born-Oppenheimer approximation. The organization of this paper is as follows. In Sec. 2, we briefly introduce the methods of solving the time-dependent Schrödinger equation (TDSE). In Sec. 3, we will present the selection rules for HHG in Born-Oppenheimer approximation and non-Born-Oppenheimer approximation. Finally, we will summarize in Sec. 4.

2. Theoretical methods

In this section, we will sketch some theoretical methods for the HHG, including the numerical solution to TDSE in Born-Oppenheimer approximation and non-Born-Oppenheimer approximation.

2.1. Numerical solution of TDSE in Born-Oppenheimer approximation

Here we just present a one-dimensional (1D) numerical solution of TDSE using the split-operator method for the targets with two nuclei. For multiple-dimensional numerical solutions with different methods (e. g. B-spline). The readers can gain more details in relevant references and a recent review [54].

The 1D TDSE describing the interaction between targets and strong laser pulses in the dipole approximation and the length gauge can be given as (atomic units are used throughout)

$$\mathrm{i}\frac{\partial}{\partial t}\Psi(x;t)=\left[-\frac{\partial^2}{2\partial x^2}+V(x)+E(t)x\right]\Psi(x,t), \qquad (1)$$

with $\mathrm{V}(x)=-Z_1/\sqrt{a+(x+R/2)^2}-Z_2/\sqrt{a+(x-R/2)^2}$, where Z_1 and Z_2 are the electric charges of two nuclei, and a is the soft-core parameter. R is the inter-nuclear distance. *E(t)* is the laser field. In Born-Oppenheimer approximation, it assumes that the molecular vibrational period is longer than the laser pulse duration. Thus the nuclei are considered to be frozen. After the initial state is obtained by propagation in imaginary time. The equation (1) can be numerically solved by the split-operator method [55].

$$\Psi(x;t+\delta t)=e^{-iT\delta t/2}e^{-iV\delta t}e^{-iT\delta t/2}\Psi(x;t)+O(\delta t^3), \qquad (2)$$

where T is the kinetic operator, and V is the interaction potential considering all the potential energy of systems. Then through the Ehrenfest's theorem [56], the dipole acceleration a(t) can be given as $a(t)=-\langle\Psi(t)|\vec{\nabla}V|\Psi(t)\rangle$. Finally, one can obtain the harmonic spectrum by Fourier-transforming the dipole acceleration

$$\mathrm{S}_q=\left|\int_{-\infty}^{+\infty}a(t)\exp(-iq\omega_0 t)dt\right|^2 \qquad (3)$$

q is the harmonic order and ω_0 is the circular frequency of laser field.

2.2. Numerical solution of TDSE in non-Born-Oppenheimer approximation

To show the effect of the nuclear motion in the process of HHG, the numerical calculation of TDSE in non-Born-Oppenheimer approximation is proposed [50]. In this part, we only introduce a numerical calculation for a typical HD molecule with two electrons and two nuclei. In this calculation, the interaction between the HD molecule and the laser field is treated within dipole approximation. Then the Hamiltonian reads

$$H=-\frac{1}{2\mu_n}\frac{\partial^2}{\partial R^2}-\frac{1}{2\mu_e}\left(\frac{\partial^2}{\partial z_1^2}+\frac{\partial^2}{\partial z_2^2}\right)+\frac{1}{\mathrm{R}}+w(z_1,z_2)$$

$$-\sum_{j=1}^{2}w\left(z_j,\frac{M_2}{Mn}R\right)-\sum_{j=1}^{2}w\left(z_j,\frac{M_1}{Mn}R\right)+E(t)\hat{D} \qquad (4)$$

with the dipole operator $\hat{D}=-(z_1+z_2)-\lambda R$. For the interactions, it chooses the soft Coulomb potential [57]

$$w(x,y)=\frac{1}{\sqrt{(x-y)^2+1}} \qquad (5)$$

z_1 and z_2 are the coordinates of two electrons. R is the inter-nuclear distance, and Mn=M1+M2 is the total nuclear mass. $\mu_e=\mathrm{M_n}/(\mathrm{M_n}+1)$ and $\mu_n=\mathrm{M_1M_2}/\mathrm{M_n}$ are respectively the reduced mass of nucleus and electron. E(t) is the laser field, and $\lambda=(\mathrm{M_2}-\mathrm{M_1})/\mathrm{M_n}$ is the mass-asymmetry parameter. The time-dependent wave function can be obtained by numerical solving TDSE using the Crank-Nicolson method [58]. Then the dipole moment can be got by

$$\mathrm{d}(t)=<\psi(t)|\hat{D}|\psi(t)>=-<\psi(t)|z_1+z_2|\psi(t)>$$
$$-\lambda<\psi(t)|R|\psi(t)>=d_e(t)+\lambda d_n(t) \qquad (6)$$

$d_e(t)$ and $d_n(t)$ are the electronic dipole moment and the relative nuclear dipole moment, respectively. The corresponding dipole acceleration is given via Ehrenfest's theorem [56]

$$\mathrm{a}(t)=-\frac{1}{\mu_e}<\psi(t)|\frac{\partial H}{\partial z_1}+\frac{\partial H}{\partial z_2}|\psi(t)>$$
$$-\frac{\lambda}{\mu_n}<\psi(t)|\frac{\partial H}{\partial R}|\psi(t)>=a_e(t)+\lambda a_n(t) \qquad (7)$$

where ae(t) and an(t) are the electronic dipole acceleration and the relative nuclear dipole acceleration. The harmonic spectrum can be easily got by Fourier-transforming the dipole acceleration.

For the numerical calculation of TDSE for molecular ion, it is similar to that of HD molecule. The relative details can be found in Refs. [50, 53, 59].

3. The selection rules for HHG from atoms and molecules in Born-Oppenheimer approximation and non-Born-Oppenheimer approximation

After the HHG being a hot topic, the selection rules as an important feature of HHG attract much attention. We will start from investigations of the selection rule for atomic HHG, including the early experimental results and the theoretical studies. Then we will review the selection rules of HHG from molecules possessing symmetry to asymmetric molecules in Born-