A scaling law of high-order harmonic generation*

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Using a nonperturbative quantum electrodynamics theory of high-order harmonic generation (HHG), a scaling law of HHG is established. The scaling law states that when the atomic binding energy E_b, the wavelength λ and the intensity I of the laser field change simultaneously to kE_b, λ/k, and k^3I, respectively. The characteristics of the HHG spectrum remain unchanged, while the harmonic yield is enhanced k^3 times. That HHG obeys the same scaling law with above-threshold ionization is a solid proof of the fact that the two physical processes have similar physical mechanisms. The variation of integrated harmonic yields is also discussed.

Keywords: high-order harmonic generation, scaling law, harmonic yield, nonperturbative quantum electrodynamics

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1. Introduction

Scaling analysis is a powerful technique to analyse various phenomena in the interaction of atoms and molecules with laser fields. For above-threshold ionization (ATI) of atoms and molecules irradiated by intense laser fields, a scaling law of photoelectron angular distributions (PADs) was established,[1] which discloses a simple physical mechanism as follows: the main features of PAD are determined by three dimensionless parameters: 1) the binding number e_b, which is the binding energy E_b in units of laser photon energy ω; 2) the ponderomotive number u_p, i.e., the ponderomotive energy U_p in units of ω; and 3) the number of the laser photons absorbed during ionization j. Keeping these three parameters unchanged, the angular distributions of photoelectrons do not change.

The high-order harmonic generation (HHG) is an important phenomenon accompanied with ATI. When atoms or molecules are irradiated by a strong laser field, the bound electrons could be ionized then accelerated by the electric field of the laser. If the ionized electron escapes from the laser field, the whole process belongs to ATI. However, if the electron recombines with its parent ion, the energy absorbed from the laser field is released simultaneously as a high-order harmonic photon, the whole process belongs to HHG. HHG has been studied extensively over the past two decades for its great applications in various research fields, such as spectroscopy analysis,[2,3] plasma diagnostics,[4,5] attosecond pulse generation,[6] and molecular dynamics.[7,8] Enhancing the cutoff energy and the harmonic yield is still the main difficulty.[9,10]

Recently, the interaction of atoms and molecules with mid-infrared laser fields draws a lot of attention, both for ATI[11−13] and HHG.[14−16] The advantage of using mid-infrared lasers in HHG is that the quasi-phase matching technique can be easily realized, which greatly enhances the conversion efficiency.[17,18] Many phenomena in the mid-infrared domain can be linked to those in the (near-) visible light domain by the scaling law, thus the scaling relation becomes important. For harmonic yield, various scaling relations on the wavelength λ of the driving laser field were established mainly by numerical methods. For instance, using the time-dependent Schrödinger equation (TDSE) method, Tate *et al.*[19] predicted that the integrated harmonic yield within a fixed energy interval follows a scaling relation as $\lambda^{-(5\sim6)}$; within the strong field approximation, Pérez-Hernández *et al.*[20] found that the harmonic yield follows a scaling relation as $\lambda^{-(3\sim4)}$; the *ab initio* quantum study of Frolov *et al.*[21] dis-

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closed a much faster decrease of the harmonic yield as wavelength increases, while Chen *et al.*[22] experimentally observed that the harmonic yield follows a scaling relation as $\lambda^{-(4\sim6)}$. Those scaling relations are far from uniform, which reflects that the essential physical mechanism is still missing. Thus, it becomes necessary to further study the scaling law of HHG and discuss the change of HHG yield under the scaling transformation. Moreover, the above-mentioned scaling relations on λ lay emphasis on the case that only the wavelength changes. If all the contributing factors change simultaneously, how does the HHG yield change?

In this article, based on the HHG rate formula derived by Gao *et al.*,[23,24] we study the scaling relation of HHG as both the target atoms and the laser fields change simultaneously. Gao's formula was derived in the frame of the nonperturbative quantum scattering theory developed by Guo–Aberg–Crasemann (GAC),[25] in which the ionization and the recombination processes are described in full quantum form. Using the formula, Gao *et al.* successfully reproduced the plateau and the cutoff of the HHG spectrum. Following Gao's treatment, we calculate the HHG spectra of different atoms in various laser fields, then establish a scaling law of HHG. The scaling law answers the question that when the wavelength of laser field changes, how the target atoms and the intensity of the driving laser field are chosen to ensure the HHG spectrum unchanged. The scaling law of HHG is the same as that of ATI, which is a solid proof of the fact that the two physical processes have similar physical mechanisms.

The remainder of this article is arranged as follows. In Section 2, the nonperturbative quantum electrodynamics theory of HHG is briefly presented. In Section 3, the harmonic spectra in various conditions are calculated and discussed, then the scaling law of HHG is presented and analysed. In Section 4, the physical mechanism of the scaling law is discussed. The conclusions are drawn in Section 5.

2. High-order harmonic generation rate formula

Using the nonperturbative quantum scattering theory, within the strong-field and the single-active-electron approximations, one can express the differential generation rate of the q-th order harmonic as ($\hbar = c = 1$ throughout this paper)[23,24]

$$\begin{aligned}
&\frac{\mathrm{d}\omega}{\mathrm{d}\Omega'} \\
&= q u_\mathrm{p} \omega^6 \frac{e^2 m_\mathrm{e}^2}{4\pi^4} \Bigg| \sum_{j(j-j'=q)} \Bigg\{ (u_\mathrm{p} - j)\,(j - u_\mathrm{p} - e_\mathrm{b})^{1/2} \\
&\quad \times \boldsymbol{\varepsilon}'^{*} \cdot \Bigg\{ \int \mathrm{d}\Omega\, |\Phi(|\boldsymbol{P}|)|^2 \,[\boldsymbol{\varepsilon} \chi^*_{-j}(z,\eta) \chi_{-j'-1}(z,\eta) \\
&\quad + \boldsymbol{\varepsilon}^* \chi^*_{-j}(z,\eta) \chi_{-j'+1}(z,\eta) + \mathrm{c.c.}] \Bigg\} \Bigg\} \Bigg|^2, \qquad (1)
\end{aligned}$$

where j and j' are the numbers of laser photons transferred in the ionization and the recombination processes, respectively; $\boldsymbol{\varepsilon}'$ and $\boldsymbol{\varepsilon}$ denote the polarization vectors of the harmonic component and the laser field, respectively; and $\mathrm{d}\Omega = \sin\theta \mathrm{d}\theta \mathrm{d}\varphi$ is the solid angle in the momentum space, θ and φ are the polar angles of the electronic momentum $\boldsymbol{P}$. The generalized phased Bessel (GPB) function $\chi_j(z,\eta)$ is defined in terms of the phased Bessel function $\mathrm{X}_n(z)$ as[26]

$$\chi_j(z,\eta) = \sum_{m=-\infty}^{\infty} \mathrm{X}_{j-2m}(z) \mathrm{X}_m(\eta), \qquad (2)$$

and the two arguments read

$$\begin{aligned}
z &= \frac{2\,|e|\,\Lambda}{m_\mathrm{e}\omega} \boldsymbol{P} \cdot \boldsymbol{\varepsilon}, \\
\eta &= u_\mathrm{p} \boldsymbol{\varepsilon} \cdot \boldsymbol{\varepsilon}/2, \qquad (3)
\end{aligned}$$

where Λ is the half-amplitude of the vector potential of the laser field. The value of electron momentum $P \equiv |\boldsymbol{P}|$ is determined by the energy conservation as $P = \sqrt{2 m_\mathrm{e} \omega (j - u_\mathrm{p} - e_\mathrm{b})}$. The order of the GPB function, which also is the mute index of the summation in Eq. (1), denotes the number of transferred laser photons. The two arguments of the GPB function in Eq. (3) are determined by the electronic momentum $\boldsymbol{P}$ and the ponderomotive parameter u_p for a fixed-polarization laser field. For analytical simplicity, the wave function is chosen as the one that is for 1s state of a hydrogen-like atom, which in momentum space is given by

$$\Phi(|\boldsymbol{P}|) = \frac{8(\pi a^3)^{1/2}}{(1 + P^2 a^2)^2}, \qquad (4)$$

where $a = 1/\sqrt{2 m_\mathrm{e} E_\mathrm{b}}$ is the Bohr radius. It is clear that for a hydrogen-like atom HHG in a long-pulse driving laser field is determined by the binding energy of the target atom, the wavelength and the intensity of the driving laser field.

3. Numerical results

According to the HHG rate formula, i.e. Eq. (1), we calculate the harmonic spectra of various target atoms in various laser fields of linear polarization focusing on the relative variation of the harmonic yield. When the wavelength of the laser field changes, how should the related physical quantities be changed in order to maintain the harmonic yield? For this purpose, we change the three quantities $E_{\rm b}$, λ, and I first in turn, then simultaneously to study the variation of HHG. Finally, a scaling law of HHG is established.

First, we show the influence of binding energy $E_{\rm b}$ on HHG spectrum and harmonic yield. The harmonic spectra from Xe with binding energy 12.1 eV and a model atom with the binding energy twice that of Xe are depicted in Fig. 1. The driving laser field has intensity $I = 2.0 \times 10^{14}$ W/cm^2 and wavelength $\lambda = 1064$ nm. Because a deeper potential well leads to a larger maximum kinetic energy at recombination, the harmonic spectrum from the model atom exhibits a wider plateau, which accords well with the cutoff law. However, the emission rate of the model atom is quite a lot lower, because less electrons are excited or ionized in a tighter bound. In order to show such a variation, in the inset we depict the integrated harmonic yields of the target atoms chosen as Xe, H, Ar and the model atom with the corresponding binding energies 12.1 eV, 13.6 eV, 15.76 eV, and 24.2 eV, respectively. We see that for a given laser field, the harmonic yield integrated over certain harmonic orders in the plateau decreases notably as the binding energy increases.

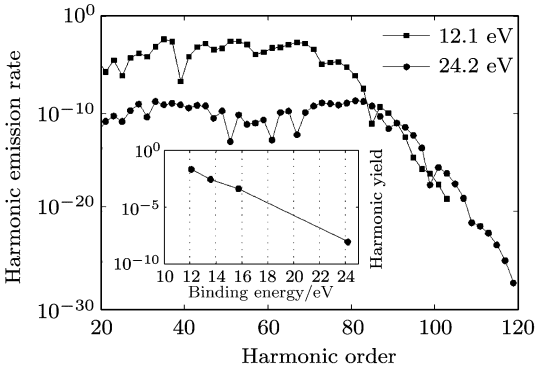


Fig. 1. Harmonic spectra from atoms with different binding energies in the laser field of wavelength 1064 nm and intensity 2.0×10^{14} W/cm^2. The inset displays the integrated harmonic yield, in which the emissions rates from the 23rd to the 65th harmonics are accumulated, versus binding energy. The emission rate and the harmonic yield are in arbitrary units.

Next, we display the influence of the laser wavelength λ. In the laser fields of intensity $I = 4.0 \times 10^{14}$ W/cm^2 and different wavelengths, the HHG spectra of Xe atoms are calculated. Figure 2(a) shows the spectra in the laser fields of wavelength 600 nm and 900 nm, respectively. The ponderomotive energy relates to the laser wavelength as $U_{\rm p} \propto I\lambda^2$, so the harmonic plateau of the atom in the laser field of longer wavelength is clearly wider for a larger maximum energy obtained from the field. We see from Fig. 2(a) that the harmonic emission rates in the plateau region in the laser field of wavelength 600 nm are slightly higher than those in the laser field of wavelength 900 nm. Comparing Fig. 2(a) with Fig. 1, we find that the increasing tendency of the harmonic emission rate caused by the decrease of wavelength is less than that caused by the decrease of the atomic binding energy.

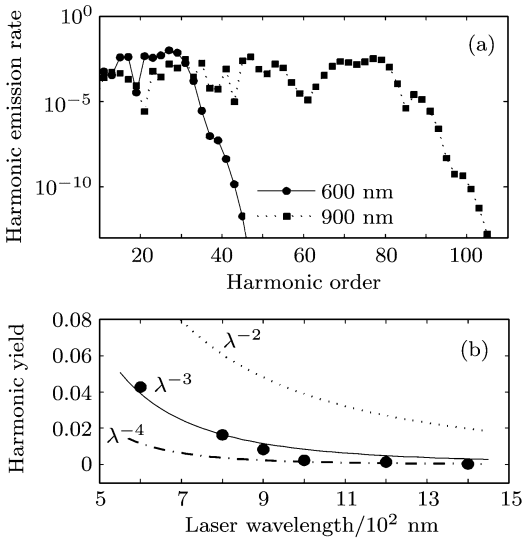


Fig. 2. (a) Harmonic spectra of Xe in the laser fields of different wavelengths and the fixed intensity 4.0×10^{14} W/cm^2. (b) The integrated harmonic yield versus laser wavelength. The solid circles denote the harmonic yield integrated from the 11st order to the 33rd order; the dotted, solid and dash-dot lines present the corresponding $\lambda^{-2.0}$, $\lambda^{-3.0}$, and $\lambda^{-4.0}$ curves. The emission rate and the harmonic yield are in arbitrary units.

The width of the harmonic plateau is more sensitive to the wavelength of the laser field than to the atomic binding energy. That is to say, the atomic binding energy plays a key role in affecting the harmonic emission rate, but the laser field majorly determines the width of the plateau. The yields of the given harmonics in the plateaus are depicted in Fig. 2(b), in which the emissions rates of harmonics from the 11st order to the 33rd order are integrated. Clearly, the yield decreases nonlinearly as the wavelength of the laser

field increases, approximately following a variation of wavelength as $\lambda^{-3.0}$ within the wavelength range involved in our calculations. The λ^{-2} dependence comes from the electronic transition from the ground state to the continuum states and λ^{-1} comes from the energy conservation in the harmonic generation process. Such variation tendencies coincide with the previous experimental observations by Shan and Chang[27] and the theoretical results given by Pérez-Hernández *et al.*[20]

Then we study the influence of laser intensity I on HHG. The calculated spectra are shown in Fig. 3. The ponderomotive energy of the laser field depends linearly on laser intensity for a given laser wavelength, the width of the HHG plateau increases linearly as the laser intensity increases. Owing to the nonlinear dependence of the harmonic emission rate on laser intensity, the integrated harmonic yield increases nonlinearly as the laser intensity increases. The calculations are depicted in the inset of Fig. 3, in which the harmonic emissions rates from the 11st order to the 69th order are integrated.

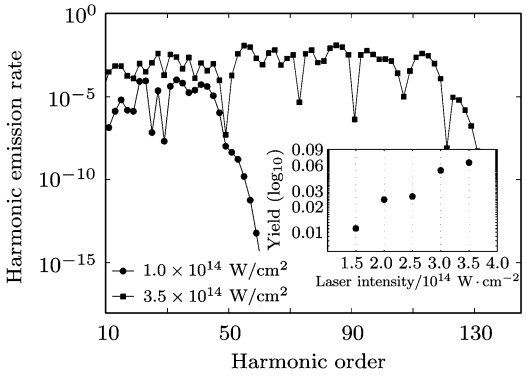


Fig. 3. Harmonic spectra of Xe in the laser fields of wavelength 1064 nm and different intensities. The inset shows the variation of the harmonic yield integrated from the 11st order to the 69th order, with laser intensity. The emission rate and the harmonic yield are in arbitrary units.

The characteristics of the harmonic spectrum including the harmonic yield and the cutoff order of the plateau change with one of the three quantities as partially mentioned in previous work. Considering the scaling law of PADs and noting that HHG and ATI are coexistent physical processes, we wonder whether the principal characteristics of HHG are comparable, with the ponderomotive parameter $u_{\rm p}$ and the binding number $e_{\rm b}$ kept unchanged. We change these three quantities, say, $E_{\rm b}$, λ, and I to $kE_{\rm b}$, λ/k, and k^3I correspondingly, then compare the HHG spectra. We find that the variation tendency of the harmonic emission rate with the harmonic order exhibits almost no change and the plateau cuts off in the same order. Moreover, the numerical calculations reveal that the harmonic emission rate in the plateau region increases k^3 times against the original, which is called the scaling law of HHG. Figure 4(a) shows the harmonic spectra of Xe and Ar. Since the binding energy of Ar is about 1.3 times that of Xe, we set $k = 1.3$. The HHG spectrum from Ar irradiated by the laser field, which is chosen according to the scaling law, is surprisingly the same as that from Xe, except for a tiny common difference in the overall emission rate. The scaling relation on the intensity is the key feature of the scaling law, since it is not difficult to understand that the atomic binding energy and the laser photon energy increases proportionally. Figure 4(b) shows the spectra from Ar in the laser fields of intensity k times and k^2 times that of the original field respectively. Not only the cutoff harmonic order but also the shape of the harmonic spectrum is notably different from that of the curve A in Fig. 4(a). The difference becomes more apparent as the laser intensity deviates further from the scaling value.

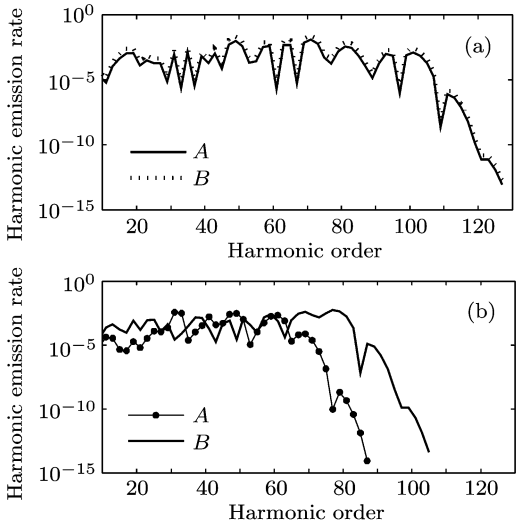


Fig. 4. (a) Curve A (solid line) denoting the harmonic spectrum of Xe in the laser field with wavelength 1064 nm and intensity 3.0×10^{14} W/cm^2, curve B (dotted line) representing the harmonic spectrum of Ar in the laser field of wavelength 818.5 nm and intensity 6.6×10^{14} W/cm^2. (b) Harmonic spectra of Ar at laser fields with wavelength 818.5 nm and intensities 3.9×10^{14} W/cm^2 (A) and 5.07×10^{14} W/cm^2 (B). The emission rate and the harmonic yield are in arbitrary units.

4. Discussion

In the following, we look for the mechanism behind the scaling law of HHG. According to Eq. (1) the harmonic emission rate is determined by two parts.

One is the dynamical process described by the three GPB functions, the other is the wave function that describes the probability distribution of the electrons in the momentum space. Under the scaling transformation, they change independently. The GPB functions, say $\chi_{-j}(z,\eta)$ and $\chi_{-j'\mp1}(z,\eta)$, describe the physical responses of electrons to intense laser fields and thus they determine the main features of the harmonic spectrum. If the three physical quantities $E_{\rm b}$, λ and I change to corresponding $kE_{\rm b}$, λ/k and k^3I simultaneously, the values of $e_{\rm b}$ and $u_{\rm p}$ keep unchanged. The number of the absorbed photons j in the ionization process is confined solely by $e_{\rm b}+u_{\rm p}$ to ensure ionization. For a given harmonic order q, because of the energy conservation in the whole process, the number of the emitted photons in the recombination process satisfies $j'=j-q$. According to the above discussion and Eqs. (1) and (3), for a given harmonic order, the variation ranges of the orders and the arguments of GPB functions are constant, which guarantees the physical process to be unchanged. So the features of harmonic spectra from Xe and Ar depicted in Fig. 4(a) are almost identical, while the tiny difference in emission rate is caused by the wave function. When $E_{\rm b}$, λ and I change according to the scaling law, the electronic momentum changes from P to $k^{1/2}P$ and the Bohr radius changes from a to $k^{-1/2}a$. For the hydrogen-like atom, the 1s state wave function changes $k^{-3/4}$ times the original one. Considering the ω^6 factor in front of the sum, the overall harmonic emission rate is enhanced k^3 times.

The scaling law of HHG relates the HHG spectra from different target atoms in different laser fields to each other, thus it provides a tool to determine the equivalent HHG processes in various cases. Since the wave function used in our calculations is that for the hydrogen-like atom, there may appear more detailed differences stemming from the trivial differences in the wave function of the target atoms. These differences can be used to extract detailed information about the valence electron in the reconstruction of atomic and molecular shells, as done by Itatani *et al.*[28] At present, the usage of hydrogen-like wave function serves as a reference.

Furthermore, the scaling law is of practical importance in the generation of higher energy harmonics: when the physical quantities $E_{\rm b}$, λ, and I change to $kE_{\rm b}$, λ/k, and k^3I, correspondingly, the width of plateau in the HHG spectrum does not change, however, the energy of a single photon increases k times that of the original. Meanwhile, the emission rates are enhanced k^3 times the original. As a result, more higher energy harmonics can be generated. Compared with using the laser field of longer wavelength to obtain higher energy harmonics, transforming the quantities according to the scaling law avoids getting a wider plateau at the cost of decreasing the harmonic yield.

Finally, we emphasize that HHG shares the same scaling law with ATI, in the sense that when the wavelength of the driving laser field changes $1/k$ times, if the atomic binding energy changes k times and the laser intensities changes k^3 times, the main features of the HHG spectrum and the PADs of ATI remain unchanged. This phenomenon reflects that HHG and ATI are coexistent homologous physical processes.[29] The scaling law of ATI holds in few-cycle laser pluses,[30] accordingly we expect that HHG in few-cycle pulses also obeys the scaling law, which will be instructive for the generation of attosecond pulses.

5. Conclusions

In this paper, using a nonperturbative quantum electrodynamics theory, we established a scaling law of HHG. Our conclusions are drawn as follows.

(i) The scaling law of HHG states that the harmonic spectrum from an atom with binding energy $E_{\rm b}$ in the laser field of wavelength λ and intensity I is the same as that from an atom with binding energy $kE_{\rm b}$ in the laser field of wavelength λ/k and intensity k^3I, where k is a real number;

(ii) When $E_{\rm b}$, λ, and I change according to the scaling law, the increase of the harmonic yield depends on the wave function of the target atom. For the hydrogen-like atom, the harmonic yield within the harmonic plateau increases k^3 times the original one;

(iii) The scaling law indicates a way to obtain more higher energy harmonics. Under the scaling transform, both the energy of a single laser photon and the harmonic emission rate increase, while the highest harmonic order remains unchanged, thus more higher energy harmonics must be obtained;

(iv) HHG and ATI obey the same scaling law, which is strong evidence of the fact that the two physical processes coexist and have similar physical mechanisms.

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