A nonperturbative quantum electrodynamic approach to the theory of laser induced high harmonic generation

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Using nonperturbative quantum electrodynamics, we develop a scattering theory for high harmonic generation (HHG). A transition rate formula for HHG is obtained. Applying this formula, we calculate the spectra of high harmonics generated from different noble gases shined by strong laser light. We study the cutoff property of the spectra. The data show that the cutoff orders of high harmonics are greater than that predicted by the “3.17” cutoff law. As a numerical experiment, the data obtained from our repeated calculations support the newly derived theoretical expression of the cutoff law. The cutoff energy of high harmonics described by the new cutoff law, in terms of the ponderomotive energy $U_p$ and the ionization potential energy $I_p$, is $3.34U_p + 1.83I_p$. The higher cutoff orders predicted by this theory are due to the absorption of the extra photons, which participate only the photon-mode up-conversion and do nothing in the photoionization process.

Keywords high harmonic generation (HHG), cutoff law, strong laser physics, nonperturbative quantum electrodynamics, transition rate formula

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1 Introduction

Laser induced high harmonic generation (HHG) from noble gases has attracted people’s attentions since the first observation in 1987 [1]. Many experimental and theoretical studies have been done over the last three decades. The HHG spectra carry information not only from the laser fields, but also from the atoms of gas media. The information extracted from atoms may be used in the tomographic reconstruction of atoms [2]. The observed high harmonics were of odd orders. Different electron recombination models were developed to account for HHG spectra. Most of the models were in the category of semiclassical theories. A notable one among those theories was a so-called three-step model [3, 4]. It successfully derived a cutoff law $q_c\hbar \omega = 3.17U_p + 1.32I_p$ for the HHG energy. In the expression of the cutoff law, $U_p$ is the pondermotive energy, $I_p$ the ionization potential energy, $\omega$ the laser photon frequency, and $q_c$ the cutoff order of HHG. There were also some nonperturbative quantum electrodynamics (NPQED) theories to account for phenomena in HHG with some artificial assumptions [5–7]. HHG is a very complicated physical process. New findings from new experiments in this area are still booming up. But all semiclassical and NPQED theories with artificial assumptions cannot truly account for all physical subprocesses. To correctly reveal all the physical subprocesses and to give experimental measurements better interpretations, it is necessary to develop a rigorous NPQED theoretical approach to HHG.

In a very recent experiment, ultrahigh-order harmonics were observed, up to orders greater than 5000 [8]. These high orders look going beyond the cutoff order predicted by the “3.17” law. To interpret this new experimental result, re-deriving HHG transition formula becomes an immediate need. With the transition rate formula, one can study the features of HHG and the cutoff law.

In this paper, we derive the HHG transition rate formula strictly according to the NPQED theory [9–10]. In this approach, quantum-field Volkov states are used as
where the laser photon mode and the spontaneously emitted photon mode, respectively. Here, \( n'_s \) and \( n''_s \) denote the spontaneously emitted photon mode, respectively. We consider a quantized laser field of frequency \( \omega \) corresponding to a wavevector \( \vec{k} \). The harmonic mode, treated as a spontaneous emission, is of frequency \( \omega' \) and has a wavevector \( \vec{k}' \). In this paper, we use \( \hbar = 1 \), \( c = 1 \), and \( e = -|e| \). In the Schrödinger picture, the Hamiltonian of the atom–radiation interaction system is [13]

\[
H = H_0 + U(r) + V + V',
\]

where

\[
H_0 = \frac{(-i\nabla)^2}{2m_e} + \omega N_a + \omega' N'_a
\]

is the noninteraction part of the Hamiltonian. Here, \( N_a \) and \( N'_a \) are photon number operators of the laser and the spontaneously emitted photon mode, respectively. And \( U(r) \) is the atomic binding potential, \( m_e \) the electron rest mass. The total electron–photon interaction is \( V + V' \) with

\[
V = \frac{e}{2m_e}[\vec{A}(r) \cdot (-i\nabla) + (-i\nabla) \cdot \vec{A}(r)] + \frac{e^2 \vec{A}(r)^2}{2m_e},
\]

\[
V' = -\frac{e}{2m_e}[\vec{A}'(r) \cdot (-i\nabla) + (-i\nabla) \cdot \vec{A}'(r)] + \frac{e^2 \vec{A}'(r) \cdot \vec{A}'(r)}{m_e},
\]

where \( \vec{A}(r) \) and \( \vec{A}'(r) \) denote the vector potential of the laser photon mode and the spontaneously emitted photon mode, respectively. We use \( \vec{A}(r) = g(e^{ik \cdot r} a + e^{-ik \cdot r} a^\dagger) \), \( \vec{A}'(r) = g(e^{ik' \cdot r} a' + e^{-ik' \cdot r} a'^\dagger) \), where \( g = (2V_0 \omega)^{-1/2} \), with \( V_0 \) being the normalization volume of the photon field, and \( e = e_\perp \cos(\xi/2) + ie_\parallel \sin(\xi/2) \) being the polarization vector. The angle \( \xi \) determines the degree of polarization, such that \( \xi = \pi/2 \) corresponds to circular polarization and \( \xi = 0 \) to linear polarization.

The transition matrix element, according to Guo, Åberg, and Crasemann (GAC) [9], is

\[
T_{fi} = \sum_{\mu(\xi_i = \xi_i)} \langle \Phi_f, n_f, 1(\Psi_\mu, n'_s) + | \Psi_\mu, n''_s \rangle' \times(\langle \Psi_\mu, n'_s | + \langle \Psi_\mu, n''_s |)(V + V')| \Phi_i, n_i, 0),
\]

where

\[
| \Psi_\mu, n'_s \rangle' = \sum_\mu' \langle \Psi_\mu', n''_s | V' | \Psi_\mu, n'_s \rangle \begin{pmatrix} \mu \times \mu' \end{pmatrix} + \langle \mu | \mu' \rangle | \Psi_\mu, n''_s \rangle,
\]

is the first-order correction to the wavefunction perturbed by the harmonic mode. Here, \( n'_s \) and \( n''_s \) denote the spontaneously emitted photon number, \( n_i \) the initial photon number of laser mode, and \( n_f \) the final one. The index \( \mu \equiv \{ Pn \} \) characterizes a quantum-field Volkov state [14]:

\[
| \Psi_\mu \rangle = V_e^{-1/2} \sum_l \exp\{i[P + (u_p - l)k]|r\} \times \chi_{-l}(z_c, z_s)(n + l),
\]

which describes a nonrelativistic electron with momentum \( P \) moving in a field of photon number \( n \) and the corresponding energy \( E_\mu = P^2/(2m_e) + (n + u_p + 1/2)\omega \), where \( V_e \) is the normalization volume of electron, \( n + l \) the Fock state of the laser photon mode, \( l \) the number of transferred photons, \( u_p = U_p/\omega \) the ponderomotive energy per laser photon energy. In latter text, we use \( e_b \) instead of \( I_p/\omega \), and \( e'_b = e_b + u_p \) for simplification.

The phased Bessel function \( X_m(z) \) and the generalized phased Bessel (GBP) functions [15] \( \chi(z, z_s) \) are defined as

\[
X_m(z) = J_m(|z|) e^{iz\omega},
\]

\[
\chi(z, z_s) = \sum_m X_{j-2m}(z_s) X_m(z),
\]

with \( z = [2|e|A/(m_e\omega)]P \cdot \epsilon \) and \( z_s = (1/2)u_pe \cdot \epsilon \).

In Eq. (5), the summing index \( \mu' \) means \( \{ P'n' \} \). In Eq. (4), \( n'_s \) and \( n''_s = 0 \) or 1, denoting none or one harmonic photon, respectively. Treating the harmonic interaction as a perturbation, we obtain the reduced transition matrix element as a sum of the following three terms [13]:

\[
T_1 = \sum_{\mu(\xi_i = \xi)} \langle \Phi_f, n_f, 1|Pn_1, 1|\Phi_i, n_i, 0\rangle,
\]

\[
T_2 = \sum_{\mu(\xi_i = \xi)} \langle \Phi_f, n_f, 1|Pn_1, 1|V|\Phi_i, n_i, 0\rangle,
\]

\[
T_3 = \sum_{\mu(\xi_i = \xi)} \langle \Phi_f, n_f, 1|Pn_0, 0|V|\Phi_i, n_i, 0\rangle,
\]

where the sum over \( \mu \) can be performed as \( \sum_\mu \equiv \sum_{Pn} = V_e^{-1}(2\pi)^3 \sum_{n} d^3P \).

According to the energy conservation between the initial state and the final state required by the scattering theory, we obtain the frequency condition for the harmonic

\[
\omega' = E_{bf} - E_{bi} + (n_i - n_f)\omega.
\]
In the case when the excited photoelectron jumps back to the initial state, we have $\Phi_f = \Phi_i$ and $E_{bf} = E_{bi}$. Then Eq. (8) becomes

$$\omega' = (n_i - n_f)\omega \equiv q\omega. \quad (9)$$

After a lengthy manipulation, we obtain explicit expressions for these three terms:

$$T_1 = \frac{|e|g' e^{*z_c}}{m_e (2\pi)^3} \sum_{j \geq |e'_f| + 1} \int d^3 p \phi_f (p')^* \phi_i (p + k' - q k)$$

$$\times \mathcal{X}_{-j}(z_c, z_s) \{ (p' - q k) \mathcal{X}_{-j-q}(z_c, z_s) \}^*$$

$$+ |e| [\epsilon \mathcal{A} \mathcal{X}_{-j-q+1}(z_c, z_s)]^*$$

$$+ e^* \mathcal{A} \mathcal{X}_{-j-q-1}(z_c, z_s)^* \}, \quad (10)$$

with

$$P' = P + (u_p - j + q) k,$$

$$P'' = P + (u_p - j') k,$$

$$T_2 = \frac{|e|g' e^{*z_c}}{m_e (2\pi)^3} \sum_{j \geq |e'_f| + 1} \int d^3 P \phi_i (p'')^* \phi_i (p + k' - q k)$$

$$\times \mathcal{X}_{-j}(z_c, z_s) \{ (p'' + j k) \mathcal{X}_{-j'+q}(z_k') \}^*$$

$$+ \frac{1}{2} k [z_c X_{j'-j+q-1}(z_k') + z_s X_{j'-j+q+1}(z_k')] + 2(z_s X_{j'-j+q-2}(z_k') + z_s X_{j'-j+q+2}(z_k'))$$

$$+ |e| A \mathcal{X}_{j'-j+q-1}(z_k')$$

$$+ e^* \mathcal{X}_{j'-j+q+1}(z_k') \}, \quad (12)$$

with

$$P = \sqrt{2m_e \omega (j - e'_f)},$$

$$P' = P - (j - j' - q) k - k', \quad P'' = P + (u_p - j) k,$$

$$\Delta KE = \frac{P - P'}{m_e} \cdot [(j - j' - q) k + k'],$$

$$P' = P + (u_p - j + q) k + k', \quad P'' = P + (u_p - j') k,$$

$$\Delta KE = \frac{P - P'}{m_e} \cdot [(j' - j + q) k - k'],$$

$$T_3 = \frac{|e|g' e^{*z_c}}{m_e (2\pi)^3} \sum_{j \geq |e'_f| + 1} \int d^3 P \phi_i (p'')^*$$

$$\times \mathcal{X}_{-j}(z_c, z_s) \{ (p'' + j k) X_{j'-j+q}(z_k') \}^*$$

$$\times \frac{(u_p - j) \omega}{\Delta KE + (j' - j) \omega} \{ (p'' + j k) X_{j'-j+q}(z_k') \}^*$$

$$\times \mathcal{X}_{-j}(z_c, z_s) \{ (p'' + j k) X_{j'-j+q}(z_k') \}^* \} \}, \quad (13)$$

This formula allows us to calculate HHG propagating in all directions. In our all-direction calculations, we find that the rate of HHG in any other direction is much less than that in the propagation direction of the laser beam. This fact also agrees with experimental observations [16]. In this paper, we only calculate spectra of high harmonics propagating in the laser beam direction.

In the direction $k'//k$, the three terms simplify as:

$$T_1 = \frac{|e|g' (2m_e \omega)^{3/2} }{(2\pi)^3} \sum_{j \geq |e'_f| + 1} \int d \Omega \phi_f (p)^*$$

$$\times \phi_i (p) \mathcal{X}_{-j}(z_c, z_s) \{ (\epsilon^{e'} \cdot P \mathcal{X}_{-j-q}(z_c, z_s))^*$$

$$+ |e| \epsilon \mathcal{A} \mathcal{X}_{-j-q+1}(z_c, z_s)^*$$

$$+ e^* \cdot \epsilon \mathcal{X}_{-j-q-1}(z_c, z_s)^* \}, \quad (14)$$

$$T_2 = \frac{|e|g' (2m_e \omega)^{3/2} }{(2\pi)^3} \sum_{j \geq |e'_f| + 1} \int d \Omega \phi_f (p)^*$$

$$\times \phi_i (p) \mathcal{X}_{-j}(z_c, z_s) \{ (\epsilon^{e'} \cdot P \mathcal{X}_{-j-q}(z_c, z_s))^*$$

$$+ |e| \epsilon \mathcal{A} \mathcal{X}_{-j-q+1}(z_c, z_s)^*$$

$$+ e^* \cdot \epsilon \mathcal{X}_{-j-q-1}(z_c, z_s)^* \}, \quad (15)$$

$$T_3 = \frac{|e|g' (2m_e \omega)^{3/2} }{(2\pi)^3} \sum_{j \geq |e'_f| + 1} \int d \Omega \phi_f (p)^*$$

$$\times \phi_i (p) \mathcal{X}_{-j}(z_c, z_s) \{ (\epsilon^{e'} \cdot P \mathcal{X}_{-j-q}(z_c, z_s))^*$$

$$+ |e| \epsilon \mathcal{A} \mathcal{X}_{-j-q+1}(z_c, z_s)^*$$

$$+ e^* \cdot \epsilon \mathcal{X}_{-j-q-1}(z_c, z_s)^* \}, \quad (16)$$

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This feature can be understood directly from the parity coefficients of $D$ where we use $\epsilon$ and $2$. Wave functions are calculated with Dirac–Hartree–Fock calculations. We choose Xe 5 as a plateau and a clear cutoff structure.

In these spectra, there are no even order harmonics. In the following we show the main results from our calculations. We compute HHG spectra for He, Ne, Ar, and Kr, shined by linearly polarized laser light of wavelength 800 nm and intensity $1.5\times10^{15}$ W/cm$^2$. These spectra show some common features: a well formed plateau and a sharp cutoff structure. The calculated spectra are presented in Fig. 2.

3 Numerical calculations and discussion

With the transition rate formula, we are able to calculate the HHG spectra from all atoms. The atomic wave functions are calculated with Dirac–Hartree–Fock method.

In the following we show the main results from our calculations. We choose Xe 5P$_{1/2}$ as the sample atomic bound state, shined by linearly polarized laser light of wavelength 800 nm. The laser beam intensity is chosen in four different values from $5\times10^{14}$ to $2\times10^{15}$ W/cm$^2$. Figure 1 shows the calculated HHG spectra which have a plateau and a clear cutoff structure.

\[ T_3 = \frac{|e|g'(2m_ee^2\epsilon)^{3/2}}{(2\pi)^3} \sum_{j\geq|\epsilon|+1} (j-e_j)^{1/2} \int d\Omega_p \Phi_f(P)^* \times \Phi_i(P) \chi_{-j}(z_c, z_s)^* \left\{ \epsilon^* \cdot P \frac{U_p-j}{-Bq} \chi_{-j+q}(z_c, z_s) + |e| |\epsilon^* \cdot \epsilon \frac{U_p-j}{-Bq} \chi_{-j+q-1}(z_c, z_s) + \epsilon^* \cdot \epsilon^* \frac{U_p-j}{-Bq} \chi_{-j+q+1}(z_c, z_s) \right\}, \quad (15) \]

where $B \equiv 1 - P \cdot k/(m_e\omega)$.

The final transition rate of HHG can be calculated by

\[ \frac{dW}{d\Omega_{k'}/k} = \frac{q\omega^4e^2m_e}{(2\pi)^8} \left| T_1 + T_2 + T_3 \right|^2, \quad (16) \]

where we use $D \equiv |e|g'(2m_ee^2\epsilon)^{3/2}/(2\pi)^3$ to replace the coefficients of $T_1$, $T_2$, and $T_3$.

Fig. 1 HHG spectra generated from Xe atom driven by the laser pulses of wavelength 800 nm and intensities 0.5, 1.0, 1.5 and $2.0\times10^{15}$ W/cm$^2$ from (a) to (d), respectively. The left dashed line and right dashed line in each subgraph refer to the cutoff orders from the cutoff law $q_c\omega = 3.17U_p + 1.32I_p$ and $q_c\omega = 3.34U_p + 1.83I_p$, respectively.

In these spectra, there are no even order harmonics. This feature can be understood directly from the parity property of GPB functions in these three terms $T_1$, $T_2$ and $T_3$ which are shown in Eqs. (13), (14) and (15). The nonvanishing of the integrals requires the integrants to be even functions. Now we change the momentum vector $P$ to $-P$ to test the parity property of the integrands. The phased Bessel and GPB functions have parity property [15]:

\[ X_m(-z) = (-1)^m X_m(z), \]

\[ X_j(-z_c, z_s) = (-1)^j X_j(z_c, z_s), \quad (17) \]

with $z_c = |2|eA/(m_e\omega)|P \cdot \epsilon$ and $z_s = (1/2)u_p \cdot \epsilon \cdot \epsilon$, we get $X_j(-z_c, z_s) = (-1)^j X_j(z_c, z_s)$. In Eq. (13), we get $\Phi_f(P)^* \Phi_i(P) = 4\pi |\Phi_i(P)|^2$, and

\[ X_{-j}(-z_c, z_s) \{ \epsilon^* \cdot (-P) \chi_{-j-q}(-z_c, z_s)^* + |e| |\epsilon^* \cdot \epsilon \chi_{-j-q+1}(-z_c, z_s)^* + \epsilon^* \cdot \epsilon^* \chi_{-j-q-1}(-z_c, z_s)^* \}
\]

\[ = (-1)^{-q+1} X_{-j}(z_c, z_s) \{ \epsilon^* \cdot P \chi_{-j-q}(-z_c, z_s)^* + |e| |\epsilon^* \cdot \epsilon \chi_{-j-q+1}(z_c, z_s)^* + \epsilon^* \cdot \epsilon^* \chi_{-j-q-1}(z_c, z_s)^* \}. \quad (18) \]

In the integration over $P$, the integrand is an odd function when $q$ is even due to the factor $(-1)^{-q+1}$. Noting the denominator $B \equiv 1 - P \cdot k/(m_e\omega) \approx 1$ in the non-relativistic limit, with the same analysis in Eqs. (14) and (15), the integrand is odd when $q$ is even, which leads to the vanishing of the integral.
There are many discussions on the plateau of the spectra in literatures. In this paper, we focus on the discussion of the cutoff property of the spectra.

In a very recent theoretical work [18], we obtained an expression of the cutoff law for HHG spectra, \( q_c \omega = 3.34 U_p + 1.83 I_p \), where we only used the cutoff property of ordinary Bessel functions and the Einstein photoelectric law in the multiphoton case, without any spectra calculation. The data from a very recent experiment [8] support the new cutoff law. Now we have the transition rate formula to calculate the HHG spectra. Those spectra obtained from our rate calculation will offer a strong numerical check to the new cutoff law, also to the previously well-accepted cutoff laws. In Fig. 3, the circle points show the cutoff orders from our spectra calculation. The sample atomic state to ionize is \( \text{Xe} \ 5P_{3/2} \), shined by linearly polarized laser light of wavelength 800 nm. We choose 16 different beam intensities from \( 5 \times 10^{14} \) to \( 2 \times 10^{15} \) W/cm\(^2\). The best fitting line of these scattered points coincides well with the line \( 3.34 U_p + 1.83 I_p \).

![Fig. 3](image)

**Fig. 3** The circle points refer to the cutoff orders obtained from the HHG spectra calculation. The best fitting line to the circle points coincides with the line \( 3.34 U_p + 1.83 I_p \).

Why the cutoff order predicted by our theory is higher than that predicted by the “3.17” law? To answer this question, we need to look into the details in our formulas for the calculation. In a good quantum mechanical theory, all kinds of conservation laws should be automatically derived from the theory. If a transition subprocess is mathematically allowed, it must meet all kinds of conservation laws. Thus, one has to include any possible transition subprocess as an open channel in the calculation. In our formula, the participating photons for HHG are not only the ionization photons, but also some non-ionization photons which participate the photon-mode up-conversion. The process for a photoelectron absorbing non-ionization photons in photon-mode up-conversions may be called the accompanying Raman effect. It is the accompanying Raman effect which makes the cutoff order of HHG greater than the one predicted by semiclassical theories. The laser photons to participate the photon-mode up-conversion are not only those ionization photons which show up in the Einstein’s photo-ionization law, but also some non-ionization photons. In a full quantum mechanical treatment, all possible transition channels must occur if they are not ruled out by mathematical restrictions which embody all kinds of conservation laws in physics. The absorbed laser photons to participate the photon-mode up-conversion are from both, the photo-ionization effect and the accompanying Raman effect.

4 Summary

The transition rate formula for HHG is strictly derived from the NPQED scattering theory. This formula allows us to calculate the HHG rates for different atoms in laser beams of arbitrary intensities and arbitrary wavelengths. Applying the transition rate formula, we calculate the HHG spectra for He, Ne, Ar, Kr, and Xe atoms. These applications can be used for future studies of HHG spectra.

According to the calculated spectra, we study the cutoff properties of HHG. In the spectra calculation, we neither use the commonly accepted cutoff law [3, 4] \( q_c \hbar \omega = 3.17 U_p + 1.32 I_p \), nor the one derived recently [18] \( q_c \hbar \omega = 3.34 U_p + 1.83 I_p \). Thus, the spectra calculation presented here offers an independent reference to different cutoff laws.

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