PHYSICAL REVIEW A, VOLUME 61, 063407

Nonperturbative quantum electrodynamics theory of high-order harmonic generation

Lianghui Gao, Xiaofeng Li, and Panming Fu
Laboratory of Optical Physics, Institute of Physics and Center for Condensed Matter Physics, Chinese Academy of Sciences, Beijing 100080, China

R. R. Freeman and Dong-Sheng Guo*
Department of Applied Science, University of California, Davis, Davis, California 95616
(Received 12 November 1999; published 9 May 2000)

Using a formal scattering theoretical approach, we develop a nonperturbative quantum electrodynamics theory to describe high-order harmonic generation (HHG). This approach recovers the semiclassical interpretation that HHG results from the recombination of photoelectrons, ionized by the laser field, with the parent ions, and gives the same phenomenological cutoff law. The HHG emission rate is expressed as an analytic closed form. We also discuss the connection between HHG and the above threshold ionization from the formal scattering viewpoint.

PACS number(s): 32.80.Rm, 42.65.Ky, 12.20.Ds, 03.65.Nk

I. INTRODUCTION

Recently, high-order harmonic generation (HHG) has become one of the most rapidly developing topics in multiphoton physics. A general character of a HHG spectrum is that it falls off rapidly at low orders, then exhibits a broad plateau where all the harmonics have the same strength, and ends up with a sharp cutoff at frequency around $E_B + 3.2U_p$, where E_B and U_p are the atomic binding energy and the electron ponderomotive energy in the laser field, respectively. Using a semiclassical approach, Corkum [1] has given very important insight into the physics of the cutoff law. In Corkum's model, the electron, through tunneling, ionizes from the atomic ground state into the continuum. Its subsequent motion can be treated classically and consists of free oscillations driven by the laser field. Once the electron returns to the vicinity of the nucleus, it may recombine with the nucleus and emit a harmonic photon.

There are a large number of theoretical attempts to calculate HHG. A quantum mechanic theory of HHG, with classical-field treatment, has been formulated in terms of the solutions of the time-dependent Schrödinger equation [2] or, equivalently, in terms of the solutions of the time-independent Floquet equation set [3]. These numerical methods are quite computer intensive. Therefore, modeling with simplifications of HHG has become popular. Becker *et al.* [4] approximated the atomic binding potential by a zero-range potential. Lewenstein *et al.* [5] considered an effective dipole model in which a Hamiltonian with just one bound state and an undistorted continuum was introduced. Both models accounted for neither the effects of excited bound states nor the effects of the ionic potential on the electronic motion in the continuum. They both led to comparably manageable integral expressions for the harmonic intensities. Later, Becker *et al.* [6] presented a general framework which treated HHG in strict parallelism to the Keldysh-Faisal-Reiss [7] framework for ionization such that the zero-range potential model and the effective dipole model were special cases.

In a classical-field approach, the key ingredient which determines the HHG spectrum is the ground-state expectation value of the atomic dipole moment. Even though the formal theory of scattering has long been recognized as an adequate tool to treat multiphoton phenomena, because both the initial and final states are the ground state of the atom in a classical field approach, using scattering theory to treat HHG becomes very subtle. Becker *et al.* [6] employed a time-dependent Hamiltonian where the incident laser field was treated classically, while the harmonic mode was quantized. In this way, they were able to define an *S*-matrix element for the emission of one harmonic photon. The *S*-matrix element can be envisioned as the coherent superposition of contributions associated with classical atomic orbits, which describe an electron that starts at the time t'' from some position within the range of the binding potential and, under the influence of the laser field, returns at the later time t' to the position again within the range of the same binding potential. The relation between the *S*-matrix element and the dipole-moment expectation value has been discussed. They also presented a functional relationship between ionization and harmonic generation.

Until now, most theoretical works on HHG have been in the category of classical-field approaches which regards the laser field as a time-dependent external field in addition to its spatial continuity feature. In the classical-field treatment, the atom in the radiation field is not in an isolated system. Thus, the energy and momentum conservation of the entire system consisting of the atom and the radiation field, during sequential transition processes, are hard to track. To provide a more solid basis for the classical-field approaches to HHG and justify some assumptions used in the classical-field modeling, it is necessary to develop a nonperturbative quantum electrodynamics (NPQED) approach to HHG, in addition to its own need to be a complete theory.

Exact solutions for an electron interacting with a quantized, elliptically polarized electromagnetic field have been found [8]. The quantum-field version of Volkov solutions enables us to treat multiphoton ionization (MPI) as a time-

*Permanent address: Department of Physics, Southern University and A&M College, Baton Rouge, Louisiana 70813.

independent scattering problem in an isolated system which consists of photons and an atom [9]. Using NPQED theory, Guo and Drake [10] succeeded in modeling the unusual peak splitting of the photoelectron angular distributions in standing-wave above-threshold ionization (ATI), known as the half Kapitza-Dirac effect in strong radiation fields. It has also been successful in explaining the phase-difference effect in the two-color ATI [11]. Recently, J. Gao *et al.* [12] applied the NPQED theory of ATI directly to treat HHG. They suggested that transitions between the quantized Volkov states contribute dominantly to the HHG spectrum. In their work, the cutoff position is near E_B+2U_p. In the current paper, we develop a time-independent formal scattering theory [13,9] to describe HHG. In our treatment, both the laser field and the harmonic field are quantized. Our theory involves an ionization process of an atomic electron from the ground state to a quantized-field Volkov state due to the interaction of the laser field followed by an electron-absorption process. The harmonic generation is a result of the recombination of the electron with its parent ions. From a physical perspective, our theory is consistent with the semiclassical interpretation made by Corkum and gives the same cutoff law. We also discuss the connection between HHG and ATI from the formal scattering viewpoint.

This paper is organized as follows. In Sec. II we employ the formal scattering theory to derive the transition rates of HHG. The HHG rate is expressed as an analytic closed form. Numerical results are given in Sec. III. We calculate the HHG spectra for various atomic potentials. All the spectra exhibit obvious plateau which end at a harmonic order $E_B/\omega+3.2u_p$. We also study HHG as a function of the ellipticity of the laser field. Section IV discusses the connection between a quantized-field Volkov state and classical-field Volkov state from the viewpoint of Floquet theory. Finally, Sec. V discusses the significance of our results.

II. TRANSITION RATE FORMULA

We consider a quantized single-mode laser field of frequency ω with a wave vector $\mathbf{k}$ and a high harmonic photon mode of frequency ω' with a wave vector $\mathbf{k}'$. In the Schrödinger picture, the Hamiltonian of the atom-radiation system is [14]

$$H=H_0+U(\mathbf{r})+V_T, \tag{1}$$

where

$$H_0=\frac{(-i\boldsymbol{\nabla})^2}{2m_e}+\omega N_a+\omega' N_a', \tag{2}$$

is the noninteraction part of the Hamiltonian. Here, $N_a=(a^\dagger a+aa^\dagger)/2$, $N_a'=(a'^\dagger a'+a'a'^\dagger)/2$ are photon number operators of the laser and the harmonic photon mode, respectively, with a and a' being the annihilation operators while $a^\dagger$ and $a'^\dagger$ the creation operators. $U(\mathbf{r})$ is the atomic binding potential. The total electron-photon interaction is $V_T=V+V'$ with

$$V=-\frac{e}{m_e}\mathbf{A}(\mathbf{r})\cdot(-i\boldsymbol{\nabla})+\frac{e^2\mathbf{A}^2(\mathbf{r})}{2m_e},$$

$$V'=-\frac{e}{m_e}\mathbf{A}'(\mathbf{r})\cdot(-i\boldsymbol{\nabla})+\frac{e^2\mathbf{A}(\mathbf{r})\cdot\mathbf{A}'(\mathbf{r})}{m_e}. \tag{3}$$

Here, the vector potentials are $\mathbf{A}(\mathbf{r})=g(\hat{\boldsymbol{\epsilon}}ae^{i\mathbf{k}\cdot\mathbf{r}}+\text{c.c.})$ and $\mathbf{A}'(\mathbf{r})=g'(\hat{\boldsymbol{\epsilon}}'a'e^{i\mathbf{k}'\cdot\mathbf{r}}+\text{c.c.})$ for the laser and harmonic mode, respectively; $g=(2\omega V_\gamma)^{-1/2}$, $g'=(2\omega' V_\gamma')^{-1/2}$; V_γ and V_γ' are the normalization volumes of the photon modes. The transverse polarizations are $\hat{\boldsymbol{\epsilon}}=\hat{\boldsymbol{\epsilon}}_x\cos(\xi/2)+i\hat{\boldsymbol{\epsilon}}_y\sin(\xi/2)$ and $\hat{\boldsymbol{\epsilon}}'=\hat{\boldsymbol{\epsilon}}_x'\cos(\xi'/2)+i\hat{\boldsymbol{\epsilon}}_y'\sin(\xi'/2)$. We neglect the $\mathbf{A}'^2$ term for its weak strength. We note that V' is the interaction due to the harmonic mode while V is the interaction due to the laser field.

The initial and final states of HHG are taken as $|\psi_i\rangle\equiv|\Phi_i(\mathbf{r}),n_i,0\rangle\equiv\Phi_i(\mathbf{r})\otimes|n_i\rangle\otimes|0\rangle'$, and $|\psi_f\rangle\equiv|\Phi_i(\mathbf{r}),n_f,0\rangle\equiv\Phi_i(\mathbf{r})\otimes|n_f\rangle\otimes|0\rangle'$ which are the eigenstates of the Hamiltonian $H_0+U(\mathbf{r})$ with eigenenergies $E_i=-E_B+(n_i+\frac{1}{2})\omega+\frac{1}{2}\omega'$ and $E_f=-E_B+(n_f+\frac{1}{2})\omega+\frac{3}{2}\omega'$, respectively, where $\Phi_i(\mathbf{r})$ is the ground-state wave function of the atomic electron with the binding energy E_B, $|n_i\rangle$, and $|n_f\rangle$ are the Fock states of the laser mode with photon number n_i and n_f, while $|0\rangle'$ and $|1\rangle'$ are that of the harmonic mode. The time-independent feature of the fully quantized Hamiltonian enables us to treat HHG as a genuine scattering process in an isolated system that consists of the photons and the atom. Energy is conserved throughout the interaction, resulting in $\omega'=(n_i-n_f)\omega$. The formal scattering theory thus applies.

The *S*-matrix element between the states ψ_i and ψ_f is [13]

$$S_{fi}=\langle\psi_f^-|\psi_i^+\rangle, \tag{4}$$

where

$$\psi_j^\pm=\psi_j+\frac{1}{E_j-H\pm i\epsilon}V_T\psi_j \quad (j=i,f). \tag{5}$$

Physically, ψ_i^+ is the scattering state at $t=0$ which has developed from a precollision state ψ_i in the remote past, while ψ_f^- is the scattering state at $t=0$ which will develop to a postcollision state ψ_f in the remote future. The *S*-matrix element can be expressed as

$$S_{fi}=\delta_{fi}-2\pi i\,\delta(E_f-E_i)T_{fi}, \tag{6}$$

where

$$T_{fi}=\langle\psi_f|V_T|\psi_i^+\rangle \tag{7}$$

is the transition matrix element. By imposing the long-wavelength approximation, $e^{i\mathbf{k}\cdot\mathbf{r}}\simeq 1$ and $e^{i\mathbf{k}'\cdot\mathbf{r}}\simeq 1$, we have $\langle\psi_f|V_T|\psi_i\rangle=0$, which is derived by noticing $\langle\Phi_i(\mathbf{r})|-i\boldsymbol{\nabla}|\Phi_i(\mathbf{r})\rangle=0$, due to the odd parity of the momentum operator, and eliminating the terms not contributing to high-order harmonics. Therefore, we have

$$T_{fi}=\left\langle \psi_f \left| V_T \frac{1}{E_i-H_0-U-V_T+i\epsilon} V_T \right| \psi_i \right\rangle. \quad (8)$$

For the electron-laser-mode subsystem, the eigenstate of the Hamiltonian H_0+V are the quantized-field Volkov states [8]

$$\Psi^0_{\mathbf{P}n}=V_e^{-1/2}\sum_{j=-n}^{\infty} \exp\{i[\mathbf{P}+(u_p-j)\mathbf{k}]\cdot\mathbf{r}\}\mathcal{J}_j(\zeta,\eta,\phi_\xi)^* \times\exp(-ij\phi_\xi)|n+j\rangle, \quad (9)$$

with corresponding energy eigenvalues $E^0_{\mathbf{P}n}=(\mathbf{P}^2/2m_e)+(n+\frac{1}{2}+u_p)\omega$. Here, $u_p=e^2\Lambda^2/m_e\omega$ is the ponderomotive energy in the units of the photon energy of the laser, where Λ, as the limit of $g\sqrt{n}\rightarrow\ (g\rightarrow 0,\ n\rightarrow\infty)$ is the half amplitude of the classical field. The generalized Bessel functions $\mathcal{J}_j$ are defined in terms of ordinary Bessel functions as

$$\mathcal{J}_j(\zeta,\eta,\phi_\xi)=\sum_{m=-\infty}^{\infty} J_{-j-2m}(\zeta)J_m(\eta)(-1)^j e^{2im\phi_\xi}, \quad (10)$$

where

$$\zeta=\frac{2|e|\Lambda}{m_e\omega}|\mathbf{P}\cdot\hat{\epsilon}|,\qquad \eta=\frac{1}{2}u_p\cos\xi,$$

$$\phi_\xi=\tan^{-1}\left(\frac{P_y}{P_x}\tan\frac{\xi}{2}\right)(+\pi).$$

Using the quantized-field Volkov states as unperturbed states, we treat the electron-harmonic interaction V' as a perturbation. Then the eigenstate of the Hamiltonian H_0+V_T, in the first order approximation, can be expressed as [14]

$$\Psi_{\mathbf{P}n,n'}=\Psi^0_{\mathbf{P}n,n'}+\Psi'_{\mathbf{P}n,n'} \quad (11)$$

with

$$\Psi'_{\mathbf{P}n,n'}=\sum_{\mathbf{P}_1 n_1,n_1'} |\Psi^0_{\mathbf{P}_1 n_1,n_1'}\rangle \frac{\langle\Psi^0_{\mathbf{P}_1 n_1,n_1'}|V'|\Psi^0_{\mathbf{P}n,n'}\rangle}{E(\mathbf{P}n,n')-E(\mathbf{P}_1 n_1,n_1')}, \quad (12)$$

where $\Psi^0_{\mathbf{P}n,n'}=\Psi^0_{\mathbf{P}n}|n'\rangle$ is the direct product of a quantized Volkov state and a Fock state $|n'\rangle$ of the harmonic mode with eigenenergy $E(\mathbf{P}n,n')=E^0_{\mathbf{P}n}+(n'+\frac{1}{2})\omega'$. Note that there is no energy shift up to first order.

The Volkov states $|\Psi_{\mathbf{P}n,n'}\rangle$ form a complete set [8] and can be used as a basis set to expand Eq. (8),

$$T_{fi}=\sum_{\mathbf{P}n,n'} \left\langle \psi_f \left| V_T \frac{1}{E_i-H_0-U-V_T+i\epsilon} \right| \Psi_{\mathbf{P}n,n'} \right\rangle \times\langle\Psi_{\mathbf{P}n,n'}|V_T|\psi_i\rangle. \quad (13)$$

We then assume that the effect of the binding potential U to HHG can be neglected when the electron is in the continuum. Keeping only the leading term in the Taylor expansion of the inverse operator Eq. (13), in terms of powers of U, we drop U in the denominator and obtain

$$T_{fi}=i\pi\sum_{\mathbf{P}n,n'} \langle\psi_f|V_T|\Psi_{\mathbf{P}n,n'}\rangle\langle\Psi_{\mathbf{P}n,n'}|V_T|\psi_i\rangle \times\delta(E_i-E(\mathbf{P}n,n')). \quad (14)$$

In the derivation of Eq. (14), the relation

$$\lim_{\epsilon\rightarrow 0^+}\frac{\epsilon}{[(E_i-E_\mu)^2+\epsilon^2]}=\pi\delta(E_i-E_\mu)$$

has been used. We expand T_{fi} to the first-order of V'

$$T_{fi}=i\pi\sum_{\mathbf{P}n,n'} [\langle\psi_f|V|\Psi'_{\mathbf{P}n,n'}\rangle\langle\Psi^0_{\mathbf{P}n,n'}|V|\psi_i\rangle +\langle\psi_f|V|\Psi^0_{\mathbf{P}n,n'}\rangle\langle\Psi'_{\mathbf{P}n,n'}|V|\psi_i\rangle+\langle\psi_f|V'|\Psi^0_{\mathbf{P}n,n'}\rangle \times\langle\Psi^0_{\mathbf{P}n,n'}|V|\psi_i\rangle+\langle\psi_f|V|\Psi^0_{\mathbf{P}n,n'}\rangle\langle\Psi^0_{\mathbf{P}n,n'}|V'|\psi_i\rangle] \times\delta(E_i-E(\mathbf{P}n,n')). \quad (15)$$

Since only the harmonics in the direction of the laser light are coherent, we set the wave vector of the harmonic mode to be parallel to that of the laser mode. With this condition, a direct evaluation shows $\langle\psi_f|V|\Psi'_{\mathbf{P}n,0}\rangle=\langle\Psi'_{\mathbf{P}n,1}|V|\psi_i\rangle=0$ [14]. We thus have

$$T_{fi}=i\pi\sum_{\mathbf{P}n} [\langle\psi_f|V'|\Psi^0_{\mathbf{P}n,0}\rangle\langle\Psi^0_{\mathbf{P}n,0}|V|\psi_i\rangle\delta(E_i-E(\mathbf{P}n,0)) +\langle\psi_f|V|\Psi^0_{\mathbf{P}n,1}\rangle\langle\Psi^0_{\mathbf{P}n,1}|V'|\psi_i\rangle\delta(E_i-E(\mathbf{P}n,1))]. \quad (16)$$

Equation (16) suggests the following interpretation. The first term describes the transition of the electron from the ground state to a Volkov state under the interaction of the laser field. The electron then returns to the ground state and emits a harmonic photon under the electron-harmonic-mode interaction V'. The second term is the event unfolding in the opposite order.

The matrix element $\langle\Psi^0_{\mathbf{P}n,0}|V|\psi_i\rangle$ is

$$\langle\Psi^0_{\mathbf{P}n,0}|V|\psi_i\rangle=V_e^{-1/2}\omega(u_p-j)\Phi(\mathbf{P}+(u_p-j)\mathbf{k}) \times\mathcal{J}_j(\zeta,\eta,\phi_\xi)e^{ij\phi_\xi}, \quad (17)$$

with $j\equiv n_i-n$; while

$$\langle \psi_f|V'|\Psi^0_{\mathbf{P}n,0}\rangle = V_e^{-1/2}\left(\frac{eg'}{m_e}\right)\Phi(\mathbf{P}+(u_p-j)\mathbf{k})^*\hat{\boldsymbol{\epsilon}}'^* \times\{[\mathbf{P}+(u_p-j)\mathbf{k}]\mathcal{J}_{j'}(\zeta,\eta,\phi_\xi)^* e^{-j'\phi_\xi} + \hat{\boldsymbol{\epsilon}} e\Lambda \mathcal{J}_{j'+1}(\zeta,\eta,\phi_\xi)^* e^{-(j'+1)\phi_\xi} + \hat{\boldsymbol{\epsilon}}^* e\Lambda \mathcal{J}_{j'-1}(\zeta,\eta,\phi_\xi)^* e^{-(j'-1)\phi_\xi}\}, \quad (18)$$

with $j'\equiv n_f-n$. Here, $\Phi(\mathbf{P}+(u_p-j)\mathbf{k})$ is the Fourier transform of the initial wave function $\Phi_i(\mathbf{r})$. The matrix elements $\langle\psi_f|V|\Psi^0_{\mathbf{P}n,1}\rangle$ and $\langle\Psi^0_{\mathbf{P}n,1}|V'|\psi_i\rangle$ are calculated in a similar way. The δ functions in Eq. (16) is factorized as

$$\delta(E_i-E(\mathbf{P}n,0)) = \left(\frac{m_e}{2\omega}\right)^{1/2}(j-u_p-E_B/\omega)^{-1/2} \times\delta(|\mathbf{P}|-(2m_e\omega)^{1/2}(j-u_p-E_B/\omega)^{1/2})$$

and

$$\delta(E_i-E(\mathbf{P}n,1)) = \left(\frac{m_e}{2\omega}\right)^{1/2}(j'-u_p-E_B/\omega)^{-1/2} \times\delta(|\mathbf{P}|-(2m_e\omega)^{1/2}(j'-u_p-E_B/\omega)^{1/2}).$$

After integrating over the radial part of the momentum space, the transition matrix element T^q_{fi} for the qth order harmonic, with the frequency $\omega'=q\omega$ where $q=j-j'$, reads

$$T^q_{fi} = \frac{i m_e e g'\omega^2}{2\pi}\hat{\boldsymbol{\epsilon}}'^*\cdot\sum_{j,j'(j-j'=q)}(u_p-j) \times\sqrt{j-u_p-E_B/\omega}\,|\Phi(|\mathbf{P}|)|^2 \times\int \sin\theta d\theta d\phi\Big\{[2\mathbf{e_p}\sqrt{j-u_p-E_B/\omega}\,\mathcal{J}_j(\zeta,\eta,\phi_\xi) \times\mathcal{J}_{j'}(\zeta,\eta,\phi_\xi)^* e^{i(j-j')\phi_\xi}-\text{c.c.}] +[\hat{\boldsymbol{\epsilon}}\sqrt{2u_p}\mathcal{J}_j(\zeta,\eta,\phi_\xi)\mathcal{J}_{j'+1}(\zeta,\eta,\phi_\xi)^* e^{i(j-j'-1)\phi_\xi} +\hat{\boldsymbol{\epsilon}}^*\sqrt{2u_p}\mathcal{J}_j(\zeta,\eta,\phi_\xi)\mathcal{J}_{j'-1}(\zeta,\eta,\phi_\xi)^* e^{i(j-j'+1)\phi_\xi} +\text{c.c.}]\Big\}. \quad (19)$$

Here,

$$|\mathbf{P}| = \sqrt{2m_e[(j-u_p)\omega-E_B]},$$

$$\mathbf{e_p} = (\sin\theta\cos\phi,\ \sin\theta\sin\phi,\cos\theta)$$

is the unit vector along **P**, and θ and ϕ are polar angles of **P**. In deriving Eq. (19), we use the relation $|\Phi(\mathbf{P}+(u_p-j)\mathbf{k})|^2_{\text{av}}\simeq 4\pi|\Phi(|\mathbf{P}|)|^2$ [8]. The subscript ''av'' means the average value among the different magnetic quantum numbers of the atomic state. The differential rate of emission of a harmonic photon is obtained by squaring the transition matrix element, i.e.,

$$\frac{dw}{d\Omega} = \frac{V'_\gamma}{(2\pi)^2}\omega'^2|T_{fi}|^2. \quad (20)$$

We consider the case where the incident light is linearly polarized (i.e., $\xi=0$). In this case, the emission rate per solid angle in the propagation direction of laser light is expressed as an analytic form as follows:

$$\left.\frac{dw}{d\Omega}\right|_{k'\parallel k} = \frac{4qe^2m_e^2\omega^5}{\pi^2}\left|\sum_{j,j'(j-j'=q)}(u_p-j)\sqrt{j-u_p-E_B/\omega} \times|\Phi(|\mathbf{P}|)|^2(\sqrt{2u_p}D_{j,j'+1}+\sqrt{2u_p}D_{j,j'-1})\right|^2. \quad (21)$$

Here

$$D_{j,j'\pm1} = \sum_{k_1=-\infty}^{\infty}\sum_{k_2=-\infty}^{\infty}\sum_{k_3=0}^{\infty}(-1)^{k_3}\left(\frac{\zeta}{2}\right)^{j+2k_1+j'\pm1+2k_2+2k_3} \times\frac{C^{k_3}_{j+2k_1+j'\pm1+2k_2+2k_3}J_{k_1}(\eta)J_{k_2}(\eta)}{(j+2k_1+k_3)!(j'\pm1+2k_2+k_3)!} \times\frac{1}{(j+2k_1+j'\pm1+2k_2+2k_3+1)}, \quad (22)$$

with $\zeta=2\sqrt{2u_p(j-u_p-E_B/\omega)}$, $\eta=\frac{1}{2}u_p$, and $C^m_n=n!/[m!(n-m)!]$.

The momentum space integration over the azimuthal angle of **P** results in the oddness condition for the high-order harmonics, a manifestation of the parity conservation in HHG. One harmonic photon has an odd parity. It can only be converted by an odd number of laser photons since one laser photon also has an odd parity.

We then consider the case where the incident light is elliptically polarized. The emission rate per solid angle can be written as

$$\left.\frac{dw}{d\Omega}\right|_{k'\parallel k} = \frac{qe^2m_e^2\omega^5}{(2\pi)^4}|\hat{\boldsymbol{\epsilon}}'^*\cdot(P\hat{\boldsymbol{\epsilon}}_x+Q\hat{\boldsymbol{\epsilon}}_y)|^2. \quad (23)$$

Here,

$$P = 2i\,\text{Im}(A)+2\cos\frac{\xi}{2}\text{Re}(C)+2\cos\frac{\xi}{2}\text{Re}(D),$$

$$Q = 2i\,\text{Im}(B)-2\sin\frac{\xi}{2}\text{Re}(C)+2\sin\frac{\xi}{2}\text{Re}(D). \quad (24)$$

The parameters *A*, *B*, *C*, and *D* are defined as

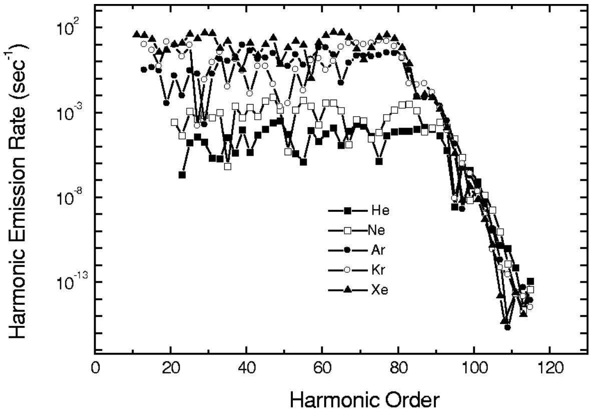

FIG. 1. Harmonic emission rate as a function of harmonic order for He, Ne, Ar, Kr, and Xe in YAG of intensity 2.2×10^{14} W/cm^2 for a hydrogenlike atomic model. The corresponding parameters are $u_p=20$ and $E_B/\omega=10.4$, 12.0, 13.4, 18.5, and 21.1 for He, Ne, Ar, Kr, and Xe, respectively.

$$A=\sum_{j,j'(q=j-j')}(u_p-j)(j-u_p-E_B/\omega)|\Phi(|\mathbf{P}|)|^2\times\int 2\sin^2\theta\cos\phi d\theta d\phi\mathcal{J}_j(\zeta,\eta,\phi_\xi)\times\mathcal{J}_{j'}(\zeta,\eta,\phi_\xi)^*e^{i(j-j')\phi_\xi}, \tag{25}$$

$$B=\sum_{j,j'(q=j-j')}(u_p-j)(j-u_p-E_B/\omega)|\Phi(|\mathbf{P}|)|^2\times\int 2\sin^2\theta\sin\phi d\theta d\phi\mathcal{J}_j(\zeta,\eta,\phi_\xi)\times\mathcal{J}_{j'}(\zeta,\eta,\phi_\xi)^*e^{i(j-j')\phi_\xi}, \tag{26}$$

$$C=\sum_{j,j'(q=j-j')}(u_p-j)\sqrt{j-u_p-E_B/\omega}\sqrt{2u_p}|\Phi(|\mathbf{P}|)|^2\times\int\sin\theta d\theta d\phi\mathcal{J}_j(\zeta,\eta,\phi_\xi)\times\mathcal{J}_{j'+1}(\zeta,\eta,\phi_\xi)^*e^{i(j-j'-1)\phi_\xi}, \tag{27}$$

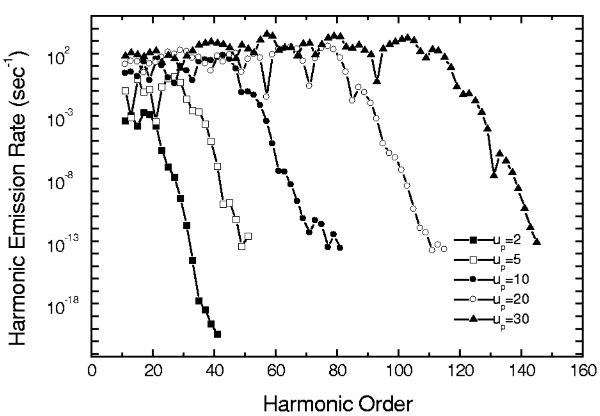

FIG. 2. Harmonic emission rate as a function of harmonic order for Xe in YAG for five different intensities.

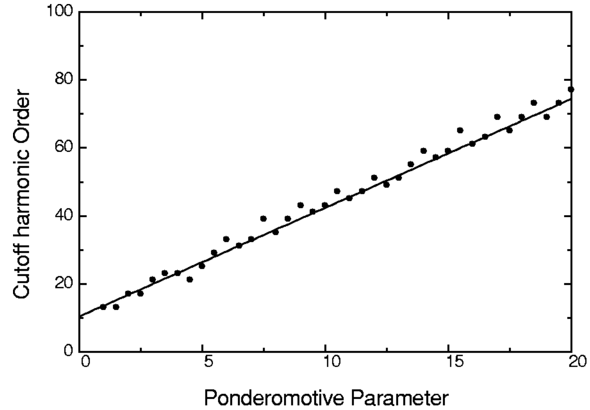

FIG. 3. The cutoff harmonic order as a function of ponderomotive parameter u_p. The straight line $q_c=E_B/\omega+3.2u_p$ corresponds to the cutoff law.

and

$$D=\sum_{j,j'(q=j-j')}(u_p-j)\sqrt{j-u_p-E_B/\omega}\sqrt{2u_p}|\Phi(|\mathbf{P}|)|^2\times\int\sin\theta d\theta d\phi\mathcal{J}_j(\zeta,\eta,\phi_\xi)\mathcal{J}_{j'-1}(\zeta,\eta,\phi_\xi)^*\times e^{i(j-j'+1)\phi_\xi}, \tag{28}$$

with $\zeta=2\sin\theta\sqrt{u_p(j-u_p-E_B/\omega)(1+\cos\xi\cos 2\phi)}$, $\eta=\frac{1}{2}u_p\cos\xi$, and $\phi_\xi=\tan^{-1}[\tan\phi\tan(\xi/2)](+\pi)$.

III. NUMERICAL RESULTS

We first consider the case where the incident light is linearly polarized. Figure 1 presents the HHG spectra of He, Ne, Ar, Kr, and Xe produced by a Nd:YAG laser ($\hbar\omega=1.165$ eV) of intensity 2.2×10^{14} W/cm^2. The corresponding parameters are $u_p=20$ and $E_B/\omega=10.4$, 12.0, 13.4, 18.5, and 21.1 for He, Ne, Ar, Kr, and Xe, respectively. We use the hydrogenic atomic model $\Phi(\mathbf{P})=2^3\pi^{1/2}\alpha^{5/4}/(\alpha+P^2)^2$, where $\alpha=2m_eE_B$. All the HHG spectra exhibit a clear plateau, which ends at a harmonic order near $E_B/\omega+3.2u_p$. We then study the dependence of the HHG on the incident intensity. Figure 2 presents the HHG spectra of Xe with $u_p=2$, 5, 10, 20, and 30; while Fig. 3 presents the cutoff harmonic order q_c as a function of u_p. The straight line $q_c=E_B/\omega+3.2u_p$ in Fig. 3 exhibits an almost perfect fit. We also compare HHG spectra obtained from the calculations using various atomic model potentials and ground state wave functions (Fig. 4). The black circles are the results where the ground-state *s*-wave function in the momentum space takes the Gaussian form $\Phi(|\mathbf{P}|)=(4\pi/\alpha)^{3/4}\exp[-(P^2/2\alpha)]$ with $\alpha=2m_eE_B$; while the black squares are the results for the hydrogenlike atoms. The NPQED approach to HHG also enable us to calculate HHG spectra even when the analytic form of the ground state wave function is unknown. The open circles present the results using a Hartree-Fock wave function. All the HHG spectra are similar and end up at the same cutoff harmonic order. Figure

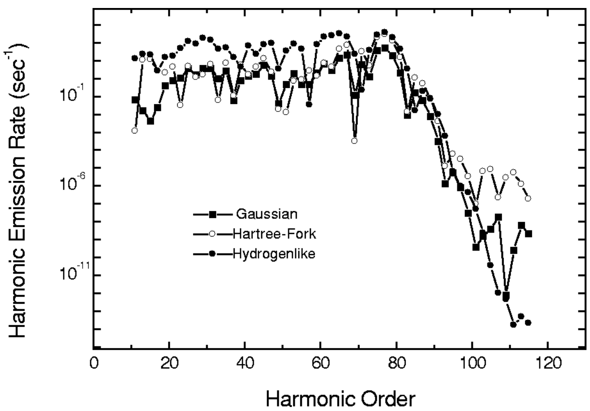

FIG. 4. Comparison of harmonic spectra obtained with Gaussian, hydrogenlike and Hartree-Fock atomic models. Parameters $E_B/\omega = 10.4$ and $u_p = 20$.

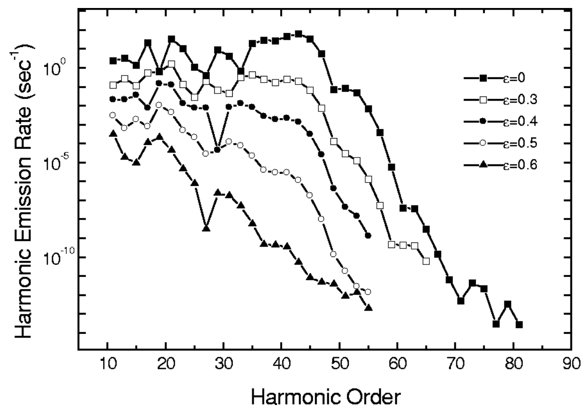

FIG. 6. Harmonic emission rate as a function of harmonic order for Xe for five values of ellipticities. Parameters $E_B/\omega = 10.4$ and $u_p = 10$.

5 presents the 35th harmonic strengths as a function of intensity in a logarithmic scale for hydrogenic, Gaussian and Hartree-Fock atomic model. Here again all curves follow a similar intensity dependence and interference pattern. We find that the slope changes as the harmonic enters the plateau region [i.e., for $q = 35$, $u_p = (q - E_B/\omega)/3.2 = 7.8$, which is just the cutoff point].

We then consider the case where the incident light is elliptically polarized. Figure 6 presents the HHG spectra of Xe for $u_p = 10$ and $\varepsilon = 0.3$, 0.4, 0.5, and 0.6. Here, $\varepsilon = \tan(\xi/2)$ is the ellipticity of the laser field. For comparison, HHG spectrum for $\varepsilon = 0$ is also given. With the increase of the ellipticity, the harmonic intensity decreases whereas the cutoff shifts toward the lower harmonic order. The cutoff harmonic order is approximately given by $E_B/\omega + 3.2u_p(1 - \varepsilon^2)/(1 + \varepsilon^2)$ [15]. On the other hand, for $\varepsilon = 0.6$ we observe, instead of a plateau structure, a decrease of the harmonic intensity. In Fig. 7, we plot the harmonic strengths as a function of the laser ellipticity for 17th, 33rd, and 43rd harmonics. The curves are normalized such that the harmonic strengths for $\varepsilon = 0$ are set to 1. The harmonic strength decreases drastically as the laser ellipticity increases from 0 to 0.5. Furthermore, interference effects are clearly observed. For example, there exists a dip for the 17th harmonic, while a local minimum is induced at $\varepsilon = 0$ for the 33rd harmonic.

IV. RELATION BETWEEN QUANTIZED-FIELD AND CLASSICAL-FIELD VOLKOV STATES

The NPQED approach to HHG involves an ionization of the electron from the ground state to a quantized-field Volkov state under the interaction of the laser field followed by a returning of the electron to the ground state with a harmonic photon emission. This is consistent with the semiclassical model that HHG results from the recombination of the electrons, excited into the continuum by the laser field, with the parent ions. Other quantum theories which recover the semiclassical interpretation are the zero-range potential model [4,6] and the effect dipole model [5]. They use time-dependent classical-field Volkov states [16] as intermediate states, by which HHG can be interpreted in terms of classical orbits departing from and returning to the ion. As is well known, a classical-field Volkov state represents an otherwise-free electron moving in a time-dependent classical em plane wave. In contrast, a quantized-field Volkov state is a coherent superposition of photonic Fock states in addition

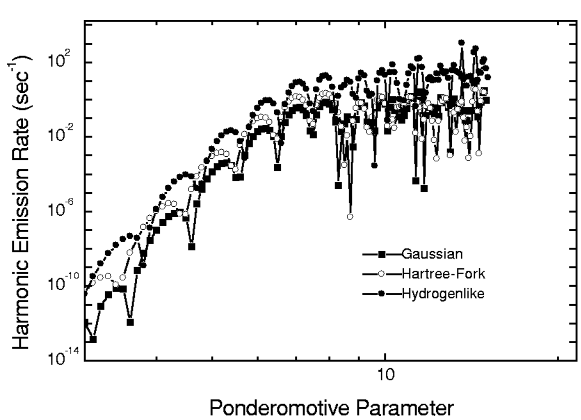

FIG. 5. Comparison of intensity dependences of the 35th order harmonic for Gaussian, hydrogenlike, and Hartree-Fock atomic models with parameter $E_B/\omega = 10.4$.

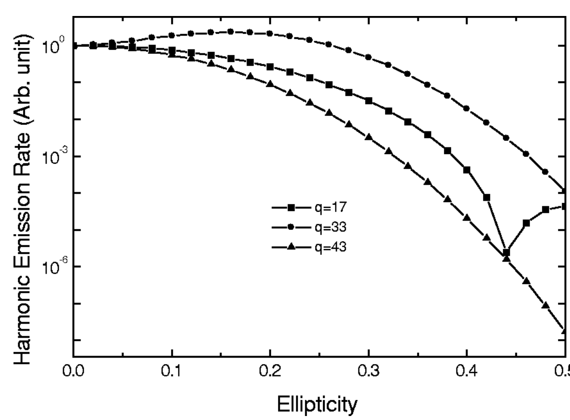

FIG. 7. Harmonic emission rate of various harmonics as the function of ellipticity for Xe. Parameters $E_B/\omega = 10.4$ and $u_p = 10$.

to the description of the electron motion. It represents the stimulated emission and absorption of photons in the quantized field by the electron. In order to clarify the intrinsic connection between the NPQED approach and the classical-field approach, we show below the relation between a quantized-field Volkov state and a corresponding classical-field Volkov state.

An original classical-field Volkov wave function [16] $\Psi_C(\mathbf{r},t)$ is a solution of the time-dependent Dirac equation. The classical-field nonrelativistic (NR) Volkov wave function should be considered as a solution of the following Schrödinger equation. (But the problem has never been simple. The subtlety due to the NR electron and relativistic light was not solved until a recent work [17].)

$$H_C(t)\Psi_C(\mathbf{r},t)=i\frac{\partial\Psi_C(\mathbf{r},t)}{\partial t}, \tag{29}$$

with Hamiltonian

$$H_C(t)=\frac{(-i\boldsymbol{\nabla})^2}{2m_e}-\frac{e}{m_e}\mathbf{A}(t)\cdot(-i\boldsymbol{\nabla})+\frac{e^2\mathbf{A}^2(t)}{2m_e}, \tag{30}$$

in the long-wavelength approximation, the classical field vector potential is $\mathbf{A}(t)=\Lambda(\hat{\epsilon}e^{-iwt}+\text{c.c.})$ with Λ the amplitude of the field. The Volkov wave function $\Psi_C(\mathbf{r},t)$ can be approximately expressed as [7]

$$\Psi_C(\mathbf{r},t)=V_e^{-1/2}\exp\left[i\mathbf{P}\cdot\mathbf{r}-i\frac{P^2}{2m_e}t-i\int_{-\infty}^{t}d\tau V_C(\mathbf{P},\tau)\right], \tag{31}$$

where

$$V_C(\mathbf{P},t)=-\frac{e}{m_e}\mathbf{A}(t)\cdot\mathbf{P}+\frac{e^2\mathbf{A}^2(t)}{2m_e}. \tag{32}$$

On the other hand, the Hamiltonian $H_C(t)$ in the long-wavelength approximation is a periodic function of t with frequency ω. According to Floquet's theorem, $\Psi_C(\mathbf{r},t)$ can also be written as [18]

$$\Psi_C(\mathbf{r},t)=\exp[i(\mathbf{P}\cdot\mathbf{r}-Et)]\sum_{j=-\infty}^{\infty}\psi_j\exp(ij\omega t). \tag{33}$$

Here, E is the quasienergy of the electron. Substituting Eq. (33) into Eq. (29) and identifying the Fourier coefficients of the like terms on both sides of the equation, we obtain

$$\left(\frac{\mathbf{P}^2}{2m_e}+u_p\omega+j\omega\right)\psi_j-\frac{\zeta\omega}{2}(e^{i\phi_\xi}\psi_{j+1}+e^{-i\phi_\xi}\psi_{j-1}) +\eta\omega(\psi_{j-2}+\psi_{j+2})=E\psi_j. \tag{34}$$

By defining a vector with an infinite dimension

$$\psi=\begin{pmatrix}\vdots\\ \psi_{j-1}\\ \psi_j\\ \psi_{j+1}\\ \vdots\end{pmatrix}, \tag{35}$$

Eq. (34) can be written as a secular equation, with the quasienergy as the eigenvalue

$$H_F\psi=E\psi. \tag{36}$$

Here the Floquet Hamiltonian H_F is a time-independent infinite dimensional Hermitian matrix with nonvanishing elements

$$(H_F)_{j,j}=\frac{\mathbf{P}^2}{2m_e}+u_p\omega+j\omega,$$

$$(H_F)_{j,j\pm1}=-\frac{\zeta}{2}\omega e^{\pm i\phi_\xi},$$

$$(H_F)_{j,j\pm2}=\eta\omega. \tag{37}$$

In most cases, the time-independent Floquet equations are solved by numerical methods. However, in our case, we have well-known classical-field Volkov solutions

$$\Psi_C(\mathbf{r},t)=V_e^{-1/2}\exp\left\{i\left[\mathbf{P}\cdot\mathbf{r}-\left(\frac{\mathbf{P}^2}{2m_e}+u_p\omega\right)t\right]\right\} \times\sum_{j=-\infty}^{\infty}\mathcal{J}_j(\zeta,\eta,\phi_\xi)^*e^{-ij\phi_\xi}\exp(ij\omega t). \tag{38}$$

Comparing Eq. (38) with Eq. (33), we obtain

$$\psi_j=\mathcal{J}_j(\zeta,\eta,\phi_\xi)^*e^{-ij\phi_\xi},$$

$$E=\frac{\mathbf{P}^2}{2m_e}+u_p\omega. \tag{39}$$

Although the Floquet theory is a semiclassical theory, Shirley [18] has pointed out that the Floquet states can be interpreted physically as quantum field states. Actually, we find that, based on the Floquet equation, a quantized-field Volkov state can be identified as a correspondence from a classical field Volkov state. A NR quantized-field Volkov wave function $\Psi_\mu(\mathbf{r})$ can be regarded as a solution of the time-independent Schrödinger eigenvalue equation [17]

$$\left[-\frac{1}{2m_e}\boldsymbol{\nabla}^2+V+\omega N_a\right]\Psi_\mu(\mathbf{r})=E_\mu\Psi_\mu(\mathbf{r}), \tag{40}$$

where V is defined by Eq. (3). Let

$$\Psi_\mu(\mathbf{r})=V_e^{-1/2}e^{i\mathbf{P}\cdot\mathbf{r}}\sum_k\phi_k|k\rangle, \tag{41}$$

then, in the long-wavelength approximation, ϕ_k satisfies

$$\left[\frac{\mathbf{P}^2}{2m_e}+(k+1/2)\left(\omega+\frac{e^2g^2}{m_e}\right)\right]\phi_k-\frac{eg|\mathbf{P}\cdot\hat{\boldsymbol{\epsilon}}|}{m_e}[e^{i\phi_\xi}\sqrt{k+1}\phi_{k+1}+e^{-i\phi_\xi}\sqrt{k}\phi_{k-1}]+\frac{e^2g^2}{2m_e}\cos\xi[\sqrt{(k-1)(k-2)}\phi_{k-2}+\sqrt{(k+1)(k+2)}\phi_{k+2}]=E_\mu\phi_k. \tag{42}$$

We consider the large-photon-number limit by letting $k=n+j$ with $n\gg j$ and $g\sqrt{n}\to\Lambda$ $(g\to 0,\ n\to\infty)$. By defining $\varphi_j=\phi_{n+j}$, then Eq. (42) becomes

$$\left[\frac{\mathbf{P}^2}{2m_e}+u_p\omega+j\omega\right]\varphi_j-\frac{\zeta\omega}{2}(e^{i\phi_\xi}\varphi_{j+1}+e^{-i\phi_\xi}\varphi_{j-1})+\eta\omega(\varphi_{j-2}+\varphi_{j+2})=(E_\mu-n\omega)\varphi_j. \tag{43}$$

Here j runs from $-n$ to infinite. Since Eq. (43) is exactly the same with Eq. (34) in the limit of $n\to\infty$ with identifying $E_\mu=E+n\omega$, we obtain

$$\varphi_j=\mathcal{J}_j(\zeta,\eta,\phi_\xi)^*e^{-ij\phi_\xi}. \tag{44}$$

Therefore, we reobtained the quantized-field Volkov state

$$\Psi_\mu(\mathbf{r})=V_e^{-1/2}\exp(i\mathbf{P}\cdot\mathbf{r})\sum_{j=-n}^{\infty}\mathcal{J}_j(\zeta,\eta,\phi_\xi)^*\times\exp(-ij\phi_\xi)|n+j\rangle, \tag{45}$$

which agrees with Eq. (9) in the long-wavelength approximation.

V. DISCUSSION AND CONCLUSION

To understand the difference between our approach and classical-field approaches, we briefly review the latter as follows. In classical-field approaches, most theories evaluate the time-dependent dipole moment expectation value $\mathbf{D}(t)$ of the dressed ground state. The harmonic generations are related to the Fourier components of $\mathbf{D}(t)$. Becker *et al.* [6] adopted a different treatment by calculating the *S*-matrix element for harmonic emission rather than the dipole moment expectation value. The *S*-matrix treatment applies for transitions from an initial to a different final state under the interaction. While for the case where the initial and the final states are the same, the probability and the rate of the transition means the leaving probability and the leaving rate. However, in a classical-field treatment, the initial and the final states in HHG are both the atomic ground state $\Phi_i(\mathbf{r})$. If the laser and the harmonic fields are both treated as external field, one will not get a correct *S*-matrix element as well as a correct transition matrix element. To treat HHG as a scattering process, Becker *et al.* used quantized harmonic mode while treated the incident laser field classically. The initial and the final states then became $\psi_i=|\Phi_i(\mathbf{r}),0\rangle$ and $\psi_f=|\Phi_i(\mathbf{r}),1\rangle$, respectively, where $|0\rangle$ and $|1\rangle$ were the Fock states of the harmonic mode. The *S*-matrix element for spontaneous emission of one harmonic photon took the form

$$S=\langle\psi_f|U_I(\infty,-\infty)|\psi_i\rangle \tag{46}$$

with

$$U_I(t,t')=T\exp\left[-i\int_{t'}^{t}d\tau H_I(\tau)\right] \tag{47}$$

in the interaction representation. The Hamiltonian $H_I(t)$ was defined in Eq. (A5) in Ref. [6] and T was the Dyson's ordering operator. Since the laser field was treated as an external and classical field, the electron was not in an isolated system; hence, formal scattering theory cannot be applied directly. Strictly speaking, the ''interaction Hamiltonian'' adopted by Becker *et al.* is not a true interaction Hamiltonian due to the time dependence of the external em field. There is no a transformation which can remove the time dependence in the Hamiltonian to find the true Schrödinger picture and the true Heisenberg picture.

Unlike the work of Becker *et al.* [6], we use quantized-field method to both laser and harmonic fields, so that HHG can be treated as a time-independent scattering process in an isolated system which consists of photons and an atom. One can readily find the well-defined Schrödinger picture, Heisenberg picture, and interaction picture in our approach. Step-by-step energy conservation is achieved in all subprocesses by NPQED theory. The processes underlying HHG can be expressed as a concise formal expression [see Eq. (16)]. In comparison to the zero-range potential model [4] and the effective dipole model [5], the advantage of our approach is that it gives more freedom for the choice of the binding potential $U(\mathbf{r})$, since it allows numerical solution of $\Phi(|\mathbf{P}|)$ in the calculation of the HHG spectra.

We now consider the connection between HHG and ATI from the viewpoint of formal scattering theory. Both ATI and HHG are scattering processes that involve the electron-atomic Coulomb interaction U and the electron-photon interaction V_T. We consider the S matrix defined in Eq. (4). Before the laser pulse comes in, the electron is bound by atomic potential U. The initial condition of the interaction can be considered as that U is on and V_T is off in the remote past. The full interaction takes place in the presence of the time-independent potentials U and V_T. Since ATI and HHG have the same precollision state, the scattering wave function ψ_i^+ are the same for both cases. The difference between ATI and HHG is that the wave functions of the electron after collision with the light are subjected to different boundary conditions. HHG is a single-potential scattering process with the final condition that U is on and V_T is off after the collision. The final-state scattering wave function ψ_f^- is given by Eq. (5). In contrast, ATI is a breakup process with the final condition U off and V_T off after the collision. The corresponding final-state scattering wave function is [13]

$$\psi_f^-=\psi_f+\frac{1}{E_j-H\pm i\epsilon}(U+V_T)\psi_f, \tag{48}$$

and the transition matrix element then becomes [9]

$$T_{fi}=\sum_\mu\langle\psi_f|\Psi_\mu\rangle\langle\Psi_\mu|V_T|\psi_i\rangle. \tag{49}$$

$$(\mathcal{E}_\mu=\mathcal{E}_i=\mathcal{E}_f)$$

Here, the final state ψ_f is the free-electron and free-photon state.

Recently, J. Gao *et al.* [12] showed a NPQED scattering theory of HHG. They extended the NPQED description of ATI to HHG. By replacing the final state ψ_f with a bound electron and free-photon state, i.e., $\psi_f = |\Phi_i(\mathbf{r}), n_f, 1\rangle$, they applied Eq. (49) directly to calculate the HHG spectra. In their theory an extra phase shift was introduced in the wave functions of intermediate states to accompany the transition. The scattering process in our treatment is different from the one of J. Gao *et al.*, that shows the process discussed by them is not the only possible process to generate high harmonics. In contrast, in our treatment the high harmonics are generated without assumption on phase shift in the wave functions. The relation between this work and J. Gao *et al.*'s work will be further discussed in future publications.

In conclusion, we develop a time-independent formal scattering theory to describe HHG. This theory recovers the semiclassical interpretation that HHG results from the recombination of the photoelectrons, excited into the continuum by the laser field, with the parent ions, and gives the same phenomenological cutoff law. The HHG emission rate can be expressed as an analytic closed form when the incident light is linearly polarized.

ACKNOWLEDGMENTS

The authors gratefully acknowledge support from the Chinese Academy of Sciences, the National Natural Science Foundation of China, and the Climbing Program from the Chinese Commission of Science and Technology. We thank Y.-S. Wu and J. Gao for stimulating discussions. One of us, D.-S.G., was supported in part by NSF Grant No. PHY-9603083.

[1] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[2] J. L. Krause, K. J. Schafer, and K. C. Kulander, Phys. Rev. Lett. **68**, 3535 (1992).
[3] R. M. Potvliege and Robin Shakeshaft, Phys. Rev. A **40**, 3061 (1989).
[4] W. Becker, S. Long, and J. K. McIver, Phys. Rev. A **50**, 1540 (1994).
[5] M. Lewenstein, Ph. Balcou, M. Yu. Ivanov, A. L'Huillier, and P. B. Corkum, Phys. Rev. A **49**, 2117 (1994).
[6] W. Becker, A. Lohr, M. Kleber, and M. Lewenstein, Phys. Rev. A **56**, 645 (1997).
[7] L. V. Keldysh, Zh. Éksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys. JETP **20**, 1307 (1965)]; F. H. M. Faisal, J. Phys. B **6**, L89 (1973); H. R. Reiss, Phys. Rev. A **22**, 1786 (1980).
[8] D.-S. Guo and T. Åberg, J. Phys. A **21**, 4577 (1988); D.-S. Guo and T. Åberg, J. Phys. B **24**, 359 (1991); D.-S. Guo and G. W. F. Drake, J. Phys. A **25**, 3383 (1992).
[9] D.-S. Guo, T. Åberg, and B. Crasemann, Phys. Rev. A **40**, 4997 (1989).
[10] D.-S. Guo and G. W. F. Drake, Phys. Rev. A **45**, 6622 (1992).
[11] L. Gao, X. Li, P. Fu, and D.-S. Guo, Phys. Rev. A **58**, 3807 (1998).
[12] J. Gao, F. Shen, and J. G. Eden, Phys. Rev. Lett. **81**, 1833 (1998).
[13] M. Gell-Mann and M. L. Goldberger, Phys. Rev. **91**, 398 (1953); M. L. Goldberger and K. M. Watson, *Collision Theory* (Wiley, New York, 1964).
[14] J. Gao, D.-S. Guo, and Y.-S. Wu, Phys. Rev. A **61**, 043406 (2000).
[15] B. Wang, X. Li, and P. Fu, Phys. Rev. A **59**, 2894 (1999).
[16] D. M. Volkov, Z. Phys. **94**, 250 (1935).
[17] D.-S. Guo, R. R. Freeman, and Y.-S. Wu, Phys. Rev. A **58**, 521 (1998).
[18] J. H. Shirley, Phys. Rev. **138**, B979 (1965).