High-Order Harmonic Generation in the Ionization Process *

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Based on the nonperturbative quantum electrodynamics scattering theory for multiphoton ionization developed recently, high-order harmonic generated in the ionization process is discussed. The influence of the Coulomb potential is treated as a perturbation in the expansion of the transition matrix. It is deduced that the harmonic photons are emitted in the resonant process during ionization and the width of the harmonic peaks is just the ionization rate of the atom.

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High-order harmonic generation (HOHG), as one of the most important photon emission phenomena in multiphoton ionization (MPI) process, has been studied intensively in recent years.[1−4] When an intense laser interacts with an atomic gas, the atoms respond in a nonlinear way and emit coherent radiation at frequencies that are multiples of the laser frequency. Many theoretical works were devoted to the mechanism of HOHG. The prevailing picture of HOHG is the recombination picture for low-frequency laser (the laser frequency is much smaller than the ionization potential).[3,4] In this picture, electrons first tunnel through the barrier formed by the atomic potential and the laser field and appear in the continuum with zero velocity. Their subsequent motion in the field is treated classically. Only those electrons that return to the nucleus can emit harmoics by recombining to the ground state.[3] This picture is obtained by a semiclassical approach. A quantum theory considering one bound state and an undistorted continuum has been developed by Becker *et al.*[5] and Lewenstein *et al.*[6] In spite of the different description of the ionization and motion in field from the semiclassical approach, it actually gives a similar picture of HOHG with the recombination picture.

In view of quantum electrodynamics (QED), all the theories discussed above are confined to semiclassical frame for which the laser fields are all treated as classical fields. The laser field should be considered as quantized field to obtain a complete quantum theory for HOHG. Recently, accompanying the development of the nonperturbative QED scattering theory for MPI,[7−9] two theories of HOHG in frame of quantum field have been presented. The paper by Gao *et al.*[10] gives a physical picture consistent with the recombinaion picture. The calculation of Gao *et al.*[11] shows that the HOHG is dominated by the continuum to continuum transition.

In this paper, we investigate the HOHG in the ionization process by using formal scattering theory. The physical picture shows that the harmonic is generated in the transition from the intermediate Volkov states to the final free electron states through the electron-field interaction and in the influence of Coulomb potential. Since it is difficult to obtain exact wavefunction of electron in the laser field and Coulomb potential, the influence of Coulomb potential has to be considered in formal scattering theory by expanding the transition matrix in which the Coulomb potential is treated as a perturbation.

The total Hamiltonian is written as

$$H = K + V + U, \tag{1}$$

where the "kinetic energy" operator $K = H_\gamma + H_e$ is the sum of the free photon-energy and free-electron-energy operator, U the Coulomb potential and $V = V_L + V_H$ the interaction Hamiltonian between the electron and the electromagnetic field with V_L and V_H labeling the laser mode term and the harmonic mode term, respectively.

The transition matrix element can be written as[8]

$$T_{fi} = \langle \psi_f | V | \psi_i^{(+)} \rangle, \tag{2}$$

where the subscripts i and f refer to the initial and final state, respectively. Here $|\psi_i^{(+)}\rangle$ is a scattering solution that satisfies

$$|\psi_i^{(+)}\rangle = |\psi_i\rangle + \frac{1}{E - K - V - U + i\epsilon} V |\psi_i\rangle. \tag{3}$$

Substituting Eq. (3) into Eq. (2), we have

$$T_{fi} = \langle \psi_f | V | \psi_i \rangle + \langle \psi_f | V G^+ V | \psi_i \rangle, \tag{4}$$

where

$$G^+ = (E - K - V - U + i\epsilon)^{-1}. \tag{5}$$

By expansion, we have

$$G^+ = G_V^+ + G_V^+ U G^+, \tag{6}$$

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where

$$G_V^+ = (E - K - V + \mathrm{i}\epsilon)^{-1}. \tag{7}$$

Then we get

$$T_{\mathrm{fi}} = \langle\psi_{\mathrm{f}}|V|\psi_{\mathrm{i}}\rangle + \langle\psi_{\mathrm{f}}|VG_V^+V|\psi_{\mathrm{i}}\rangle + \langle\psi_{\mathrm{f}}|VG_V^+UG^+V|\psi_{\mathrm{i}}\rangle. \tag{8}$$

The first and second terms in the right-hand side of Eq.(8) are just the considered ones in Ref. 9. The first term gives no contribution, because it is assumed that the electron and laser field are separated in the initial and final states and the process treated here is an MPI process. The second term gives the main contribution to the ionization probability. Then we focus on the third term denoted by T_{c}.

Expanding G^+ in terms of V, we have

$$G^+ = G_U^+ + G_U^+VG^+, \tag{9}$$

where

$$G_U^+ = (E - K - U + \mathrm{i}\epsilon)^{-1}. \tag{10}$$

T_{c} can be expanded to

$$\begin{aligned} T_{\mathrm{c}} &= \langle\psi_{\mathrm{f}}|VG_V^+UG_U^+V|\psi_{\mathrm{i}}\rangle + \langle\psi_{\mathrm{f}}|VG_V^+UG_U^+VG^+V|\psi_{\mathrm{i}}\rangle \\ &= T_1 + T_2. \end{aligned} \tag{11}$$

G_V^+ can be expressed as

$$G_V^+ = \sum_\mu \frac{|\psi_\mu\rangle\langle\psi_\mu|}{E - E_\mu}, \tag{12}$$

where ψ_μ is the solution of the eigenequation

$$(K + V)|\psi_\mu\rangle = E_\mu|\psi_\mu\rangle, \tag{13}$$

and G_U^+ can be expanded as

$$G_U^+ = \sum_j \frac{|\phi_j\rangle\langle\phi_j|}{E - E_j}, \tag{14}$$

where ϕ_j is the solution of the eigenequation

$$(K + U)|\phi_j\rangle = E_j|\phi_j\rangle. \tag{15}$$

To simplify the problem, it is assumed that only one bound state exist with other states ignored. So we write G_U^+ as

$$G_U^+ = \sum_j \frac{|\phi_j\rangle\langle\phi_j|}{E - E_j} = \frac{|\phi_j\rangle\langle\phi_j|}{E - E_j}. \tag{16}$$

Then T_1 can be written as

$$T_1 = \langle\phi_{\mathrm{f}}, n_{\mathrm{f}}|V\sum_\mu \frac{|\psi_\mu\rangle\langle\psi_\mu|}{E - E_\mu}U\frac{|\phi_{\mathrm{i}}\rangle\langle\phi_{\mathrm{i}}|}{E - E_{\mathrm{i}}}V|\phi_{\mathrm{i}}, n_{\mathrm{i}}\rangle, \tag{17}$$

where we write the electron wavefunction and photon wavefunction separately in the initial and final states, ϕ_{f} is a free electron state and ϕ_{i} is a bound state, $\langle\phi_{\mathrm{f}}, n_{\mathrm{f}}|V|\psi_\mu\rangle$ is the transition matrix between the Volkov state and the free electron state, and has been discussed in Ref. 9. It gives no contribution to HOHG. The transition matrix $\langle\phi_{\mathrm{i}}|V|\phi_{\mathrm{i}}, n_{\mathrm{i}}\rangle$ containing harmonic generation is, if written exactly,

$$M = \langle\phi_{\mathrm{i}}, n_{\mathrm{f}}, 1|V_{\mathrm{H}}|\phi_{\mathrm{i}}, n_{\mathrm{i}}, 0\rangle, \tag{18}$$

where V_{H} is written as

$$V_{\mathrm{H}} = -\frac{e}{m_{\mathrm{e}}}\boldsymbol{A}_{\mathrm{H}}(\boldsymbol{r})\cdot(-\mathrm{i}\nabla) + \frac{e^2\boldsymbol{A}_0(\boldsymbol{r})\cdot\boldsymbol{A}_{\mathrm{H}}(\boldsymbol{r})}{m_{\mathrm{e}}}. \tag{19}$$

If we write the denominator $E - E_{\mathrm{i}}$ in energy conversation case,

$$E_{\mathrm{b}} + n\omega_0 + \omega_{\mathrm{H}} = E_{\mathrm{b}} + n_{\mathrm{i}}\omega_0. \tag{20}$$

where ω_0 is the energy of the laser mode photon(we set $\hbar = c = 1$) and ω_{H} is the energy of the harmonic photon. Since V_{H} only includes the first power term of $a_{k_0}^\dagger$, we have

$$\omega_{\mathrm{H}} = \omega_0. \tag{21}$$

So it is concluded that T_1 gives no contribution to HOHG. Substituting Eqs. (12) and (16) into the expression of T_2 and only keeping the first term in the expansion, we have

$$\begin{aligned} T_2 = \langle\phi_{\mathrm{f}}, n_{\mathrm{f}}|V\sum_{\mu'}\frac{|\psi'_\mu\rangle\langle\psi'_\mu|}{E - E_{\mu'}}U\frac{|\phi_{\mathrm{i}}\rangle\langle\phi_{\mathrm{i}}|}{E - E_{\mathrm{i}}}V \\ \cdot\sum_\mu\frac{|\psi_\mu\rangle\langle\psi_\mu|}{E - E_\mu}V|\phi_{\mathrm{i}}, n_{\mathrm{i}}\rangle, \end{aligned} \tag{22}$$

where Eq. (22) contains the matrix element

$$T_{\mathrm{H}} = \langle\phi_{\mathrm{i}}|V_{\mathrm{H}}\sum_\mu\frac{|\psi_\mu\rangle\langle\psi_\mu|}{E - E_\mu}V_{\mathrm{L}}|\phi_{\mathrm{i}}, n_{\mathrm{i}}\rangle, \tag{23}$$

which is just the same as that considered in Ref. 10.

This term gives contribution to HOHG. We write the term $E - E_{\mathrm{i}}$ in Eq. (22) as

$$E - E_{\mathrm{i}} = E_{\mathrm{b}} + n_{\mathrm{i}}\omega_0 - (E_{\mathrm{b}} + n\omega_0 + \omega_{\mathrm{H}}). \tag{24}$$

Clearly, if Eq. (24) is equal to zero, then

$$\omega_{\mathrm{H}} = (n - n_{\mathrm{i}})\omega_0 = q\omega_0, \tag{25}$$

meaning that the q-th harmonic photon is emitted in the resonant process. On the other hand, non-harmonic photons can also be emitted in the case $E - E_{\mathrm{i}} \neq 0$, i.e. in the nonresonant process.

We define

$$T_i = \langle\phi_{\mathrm{f}}, n_{\mathrm{f}}|V\sum_{\mu'}\frac{|\psi'_\mu\rangle\langle\psi'_\mu|}{E - E_{\mu'}}U|\phi_{\mathrm{i}}\rangle. \tag{26}$$

Comparing Eq. (26) with the second term in Eq. (8), i.e., the transition matrix calculated in Ref. 9, it is clear that T_{i} is almost the same as the ionization transition matrix except that the part $\langle\psi'_\mu|V|\phi_{\mathrm{i}}\rangle$ is replaced by $\langle\psi'_\mu|U|\phi_{\mathrm{i}}\rangle$. To estimate the magnitude of

the transiton probability of this kind of HOHG, we can approximately assume that $T_{\rm i}$ is equal to the ionization transiton matrix. Then Eq. (22) can be rewritten as

$$T_2 = T_{\rm i}\frac{1}{E-E_{\rm i}}T_{\rm H}. \tag{27}$$

So

$$|T_2|^2 = |T_{\rm i}\frac{1}{E-E_{\rm i}}T_{\rm H}|^2 \simeq |T_{\rm i}|^2|\frac{1}{E-E_{\rm i}}|^2|T_{\rm H}|^2. \tag{28}$$

The transition rate of the whole process is

$$W = \frac{V_\gamma^2 V_{\rm e}}{(2\pi)^9}\int {\rm d}\boldsymbol{p}_{\rm f}{\rm d}\boldsymbol{k}'{\rm d}\boldsymbol{k}_{\rm H}|T_2|^2 2\pi\delta(E_{\rm f}-E_{\rm i}). \tag{29}$$

To obtain Eq. (29), we have used the conclusion of Ref. 9 that resonant spontaneous emission in the transition from the intermediate Volkov state to the final state gives the main contribution to ionization probability. The spontaneous emitted photon is labeled by $\boldsymbol{k}'$ in Eq. (29).

Since the ionization rate is

$$W_{\rm i} \simeq \frac{V_\gamma V_{\rm e}}{(2\pi)^6}\int {\rm d}\boldsymbol{p}{\rm d}\boldsymbol{k}'|T_{\rm i}|^2 2\pi\delta(E_{\rm f}-E_{\rm i}), \tag{30}$$

so the differential transition rate can be expressed as

$$\frac{{\rm d}W}{{\rm d}\Omega} \simeq \frac{V_\gamma}{(2\pi)^3}W_{\rm i}\int \omega_{\rm H}^2{\rm d}\omega_{\rm H}|T_{\rm H}|^2|\frac{1}{E-E_{\rm i}}|^2. \tag{31}$$

It is clear that Eq. (31) will diverge because of the denominator $E-E_{\rm i}$. This difficulty can also be solved by introducing an imaginary part in the energy, the same as that adopted in Ref. 9. It should be noted that the atom is in the laser field and will undergo ionization. So an imaginary part $\Gamma_{\rm i}$ should be included in $E_{\rm i}$ and $\Gamma_{\rm i}$ is just the ionization rate of the atom. Then Eq. (24) is written as

$$E - E_{\rm i} = (n_{\rm i}-n)\omega_0 - \omega_{\rm H} + {\rm i}\Gamma_{\rm i}, \tag{32}$$

where $\Gamma_{\rm i}$ is just equal to $W_{\rm i}$. Then we get

$$\frac{{\rm d}W}{{\rm d}\Omega{\rm d}\omega_{\rm H}} \simeq \frac{V_\gamma}{(2\pi)^3}W_{\rm i}\omega_H^2|T_{\rm H}|^2\frac{1}{(q\omega_0-\omega_{\rm H})^2+\Gamma_{\rm i}^2}. \tag{33}$$

Using the differential rate of HOHG obtained in Ref. 10,

$$\frac{{\rm d}W_{\rm h}}{{\rm d}\Omega} = \frac{V_\gamma}{(2\pi)^3}\int k_{\rm H}^2{\rm d}k_{\rm H}|T_{\rm H}|^2 2\pi\delta(E_{\rm f}-E_{\rm i}), \tag{34}$$

the differential rate of HOHG in the ionization process can be finally approximately written as

$$\frac{{\rm d}W}{{\rm d}\Omega} \simeq \frac{1}{2}\frac{{\rm d}W_{\rm h}}{{\rm d}\Omega},$$

where we have used $\lim_{\epsilon\to 0^+}\frac{\epsilon}{x^2+\epsilon^2} = \pi\delta(x)$. So the probability of the two kinds of HOHG are of the same order. From Eq. (33), it can also be concluded that the width of the harmonic peaks is just the ionization rate of the atom.[12] Moreover, after integrating over the momentum in the intermediate states ψ_μ, it is also deduced that only the odd order harmonic can be generated but the even orders vanish.

The matrix element T_2 describes the transition process from initial bound state to final free electron state and high-order harmonic is generated during the transition. So it can be called HOHG in the ionization process. Physically, the key point is that the electron bound state, i.e. the Coulomb potential participates in the transition process. The physical picture of the HOHG in the ionization process can be expressed as: through the interaction $V_{\rm L}$ between the electron and laser field, the electron firstly transits from the initial bound state to the intermediate Volkov state; secondly, the electron transits to bound state again through the spontaneous term $V_{\rm H}$ and emits a harmonic photon; finally, the electron transits to final state through Coulomb potential U and electron-electromagnetic field interaction V.

Using formal scattering theory, the high-order harmonic generated in the ionization process is investigated. The transition matrix element obtained indicates that the Coulomb potential participates in the process by the same way as in continuum-bound high-order harmonic generation. So the two kinds of harmonics have the same property that only the odd order harmonic photons can be emitted. Moreover, we find that the probabilities of them are of the same order by estimating the magnitude of the transition probability. In the ionization process, the harmonic photons are emitted in the resonant process while the non-harmonic photons are emitted in the nonresonant process and the width of the harmonic peaks is just the ionization rate of the atom.

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