Coulomb corrections and polarization effects in high-intensity high-harmonic emission

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(Received 5 December 1995)

We describe a quasiclassical approach to correct electron motion following tunneling ionization in intense laser fields, for the presence of the Coulomb potential. The technique is applied to model the dependence of high-harmonic generation on the ellipticity of the incident laser field. We present experimental data on the harmonics with photon energies close to the ionization potential of an atom, identify their source, and explain their unusual dependence on the ellipticity of the incident laser field. Our calculations are in good agreement with experimental data. [S1050-2947(96)04107-8]

PACS number(s): 42.50.Hz, 42.65.Ky, 32.80.Rm

Analytical theories are generally performed within the Keldysh approximation [1], i.e., the effect of the Coulomb potential is neglected. However, when it comes to more subtle aspects and to quantitative predictions, even in the intense field limit, Coulomb effects can play an important role, as has been found with tunneling ionization [2] or above threshold ionization [3]. Ionization occurs via tunneling when the energy of the electron-laser interaction $U_p=E^2/4\omega^2$ exceeds the atomic ionization potential I_p. Currently, the only analytical technique introducing Coulomb modifications to Keldysh theories in intense field or tunneling limit [2] corrects the *probability of tunneling ionization* in the constant electric-field approximation and does not address subsequent electron motion, which is crucial, e.g., for high-harmonic emission. Such a correction is introduced in this paper.

Our theoretical analysis is based on the following concept. First, the intense laser-atom interaction is analyzed in the absence of the Coulomb potential. In the tunneling limit the Keldysh solution can be viewed in three separate steps: tunnel ionization, electron propagation in the continuum, and the interaction of the returning electron with the parent ion [3–7]; see Fig. 1. Standard perturbation techniques applied to Keldysh theories fail mainly because it is difficult to find approximations that remain valid *for all three stages* of the electron evolution. Hence each of the three steps should be corrected individually utilizing different approximations. Both tunneling to the continuum and reencounter with the parent ion occur on a time scale much faster than one optical laser cycle. Therefore, the quasistationary field approximation can be used to correct for the Coulomb potential, as done for tunneling ionization in Ref. [2]. For the electron propagation in the continuum the quasistationary approximation cannot be used. We use a semiclassical perturbation theory similar to that introduced in [2], but applied to the *time-dependent* Schrödinger equation.

This approach can be applied to various intense field problems, such as above-threshold ionization and high-harmonic generation. High-harmonic generation can be corrected most easily since the returning electron recombines with the parent ion. Above-threshold ionization is more complicated to correct, as the ionized electron does not recombine, but can undergo complex rescattering processes at the parent nucleus [8].

In this paper we apply our approach to model the emission of high harmonics with photon energies close to the ionization potential of an atom $N\omega\sim I_p$. These "threshold harmonics" were recently found to have unusual properties [9,10]. In particular, their ellipticity dependence strongly deviates [9] from Gaussian, which is typical [11] for high harmonics with photon energies well in excess of the ionization potential. As seen in Fig. 2, quite unexpected minima in the yield of some threshold harmonics are observed for linear laser polarization, while slightly elliptical light provides higher intensity of these harmonics. These surprising experimental data have raised the question of the source of the threshold harmonics [9] (bound-bound, bound-free, or free-free transitions).

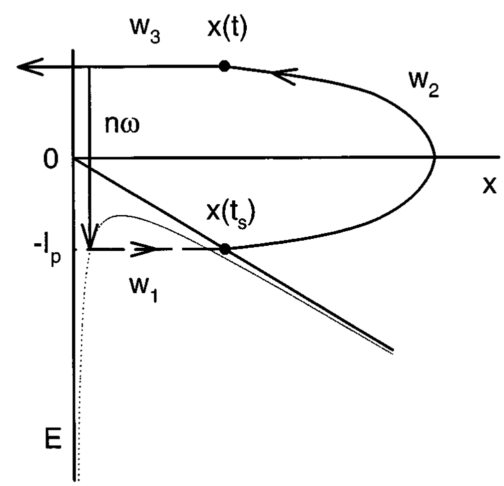

FIG. 1. Schematic of high-harmonic generation; the x and E axes denote the space and energy coordinates. The dotted line and its asymptote are the combined potential of nucleus and electric field and the potential of the electric field, respectively; the terms W_1, W_2, and W_3 represent the different stages of electron evolution: tunnel ionization, propagation in the electric field, and recombination with the parent ion.

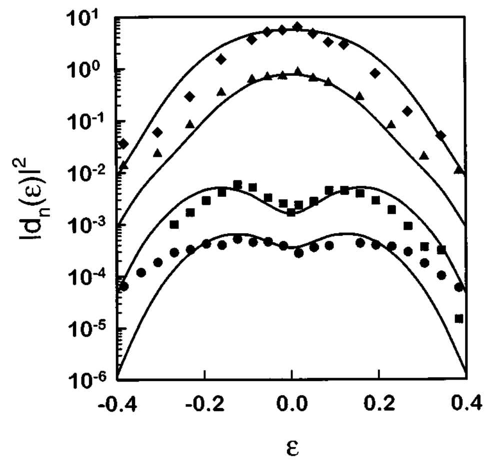

FIG. 2. Absolute magnitude of the dipole moment for harmonic orders $N=13$–19 versus the ellipticity of the incident laser light. The solid lines represent the numerical results; the closed circles, squares, triangles, and diamonds denote experimental results for $N=13$–19 generated in neon, respectively.

We identify the threshold harmonics $N\omega \geq I_p$, obtained in the tunneling limit, as being due to bound-free transitions and find that their surprising ellipticity behavior is due to an interference between the first and second return of the electron to its parent ion. Qualitatively, the modified ellipticity behavior can also be found for a δ-function potential [6]. However, our analysis demonstrates that, in order to obtain quantitative agreement with experimental data, the inclusion of the Coulomb corrections to electron propagation in the continuum is essential. Indeed, the interference effects between the two returns are sensitive to the phase of the electron wave function, which is mainly accumulated during the electron motion in the continuum and is significantly modified by the Coulomb potential. Our conclusions are supported by a comparison to recent experimental data on the threshold harmonics, which we now briefly describe.

The details of our experimental setup and measurement procedure are published elsewhere [9a]. In brief, we used 150-fsec Ti:sapphire laser, operating at 754 nm, focused down with an $f/30$ lens to $I \sim 1\times10^{15}$ W/cm^2 into a Ne gas at 100 Torr. The polarization was linear to better than 500:1 in intensity. Ellipticity was introduced by rotating a zeroth-order quarter wave plate placed just before the focusing lens. Harmonics were observed by placing a channeltron electron multiplier in the focal plane of a flat field variably spaced grating spectrograph. Odd harmonics extending up to about $N=65$ were observed from Ne.

Present data differ from those previously published [9a] for the ellipticity dependence of the near-threshold harmonics in two respects. First, care has been taken to remove a small third-order contribution from higher harmonics by inserting thin Al and Mg foil filters between the grating and electron multiplier. Second, the spectrograph has been carefully recalibrated using the 49.5 and 72.8-eV absorption edges of Mg and Al, respectively. This recalibration of the spectrograph leads to the conclusion that the harmonic lines attributed as $H13$, $H15$, and $H17$ in Ref. [9a] were in fact $H15$, $H17$, and $H19$. The data for the ellipticity dependence of $H13$–$H19$ is shown in Fig. 2. It should be noted that at the wavelength used in this experiment, $H13$ at 21.38 eV is just below the field-free ionization potential of neon $I_p=21.565$ eV.

Harmonics from the 21st up to the 51st (the highest measured) exhibit a nearly Gaussian ellipticity dependence with approximately constant width and are well described by the models of Refs. [5,6], as published previously. The ϵ-dependences of the 17th and 19th harmonics are qualitatively similar to each other and show a clear difference from that of higher harmonics: they are broader and deviate consistently from Gaussian. Particularly striking is the ellipticity dependence of the threshold 13th and 15th harmonics. The total yield has a pronounced minimum at zero ellipticity and peaks at $\epsilon \approx 0.1$.

We now proceed to modeling these experimental data. Consider a hydrogenlike atom in an elliptically polarized laser field $E(t)=[E_x \cos(\omega t), E_y \sin(\omega t), 0]$. Initially, the electron is in the ground state $|g\rangle$. In the presence of an electric field the wave function becomes $|\Psi\rangle=|g\rangle+|\Phi\rangle$. For the sake of simplicity the ground-state depletion is neglected. Since we are interested in the limit where the ponderomotive potential $U_p=E^2/(4\omega^2)$ is larger than I_p, ionization proceeds via tunneling and coupling to other bound states can also be neglected; then, the function Φ traces the evolution in the continuous spectrum.

The high-harmonic signal is determined by the quantum-mechanical expectation value of the acceleration operator. The intensity of the nth harmonic from a single atom is proportional to $(n\omega)^4|\mathbf{d}_n|^2$, where $\mathbf{d}_n$ is the nth Fourier component of the field-induced dipole moment, i.e., $\mathbf{d}(t) \approx \langle g|\mathbf{d}|\Phi(t)\rangle + \text{c.c.} = \Sigma_n \mathbf{d}_n \exp(in\omega t)$.

In the absence of the Coulomb potential the analytical expression for the dipole moment was derived in [5]. Using the stationary phase method to perform integration over the initial momenta $\mathbf{p}$ of the ionized electron, one obtains the following expression, also given in [5]:

$$\mathbf{d}(t)=\frac{(2\pi)^{3/2}}{\sqrt{i}}\int^{t} dt'\,\mathbf{E}(t')\,\mathbf{d}[\mathbf{p}(t,t')-\mathbf{A}(t')] \times\frac{\hat{U}[t,t',\mathbf{p}(t,t')]}{(t-t')^{3/2}}\,\mathbf{d}[\mathbf{p}(t,t')-\mathbf{A}(t)]+\text{c.c.}, \quad (1)$$

where the Volkov propagator $\hat{U}$ is

$$\hat{U}(t,t',\mathbf{p})=\exp\left[-i\int_{t'}^{t}[\mathbf{p}-\mathbf{A}(t'')]^2/2dt''-iI_p(t-t')\right]. \quad (2)$$

Here $\mathbf{A}(t)$ is the vector potential of the electric field and $\mathbf{d}(\mathbf{v})$ is the dipole matrix element of the transition from the ground state to the continuum state characterized by the electron velocity $\mathbf{v}$ at the infinity. The canonical momentum $\mathbf{p}(t,t')$ is given by the condition that an electron ionized at the moment t' returns to its initial position at the moment t, $\mathbf{r}(t)=\mathbf{r}(t')$. For elliptically polarized light

$$\mathbf{p}(t,t')=\left(E_x\,\frac{\cos\omega t-\cos\omega t'}{\omega^2(t-t')},E_y\,\frac{\sin\omega t-\sin\omega t'}{\omega^2(t-t')},0\right). \quad (3)$$

Before the effects of the Coulomb potential can be included, the integration in Eq. (1) has to be performed analytically, to extract the ''classical'' stage of electron evolution between tunneling and recombination. This is achieved by expanding fast oscillating classical action $S(\mathbf{p},t,t')=\int_{t'}^{t}dt''[\mathbf{p}(t,t')-\mathbf{A}(t'')]^2/2$ in Eq. (2) in a Taylor series (up to the *third* order) near its stationary phase points. This method is applicable when $I_p<U_p$, i.e., when the saddle points of the integral Eq. (1) are close to the stationary phase points of the classical action, so that the Taylor expansion around the stationary phase points is accurate at the saddle points. Integral Eq. (1) has a pole at the saddle point. Taking into account its contribution, for low elipticity of the incident light, $E_y^2 \ll E_x^2$, we obtain

$$\mathbf{d}(t,t_s)=\sum_{t_s}\frac{1}{\sqrt{i}}\,a_{\mathrm{ion}}(t_s)a_{\mathrm{pr}}(t,t_s)a_{\mathrm{rec}}(t)+\mathrm{c.c.} \tag{4}$$

Here a_{ion}, a_{pr}, and $a_{\mathrm{rec}}(t)$ are the amplitudes of tunneling ionization, propagation after tunneling and spontaneous recombination, and $t_s=t_s(t)$ are determined by the solution of the stationary phase equation for the major component of the classical action: $1/2[\mathbf{p}_x(t,t'=t_s)-\mathbf{A}_x(t'=t_s)]^2=0$. There is more than one solution of this equation, corresponding to different moments of birth t_s yielding the same moment of return t. However, due to fast spreading of the electronic wave packet only those t_s that are within few laser cycles from the moment t are essential. Up to numerical constants $O(1)$ the three amplitudes in Eq. (4) are

$$a_{\mathrm{ion}}(t_s)=\left(\frac{1}{1+v_y^2(t_s)/2I_p}\right)^{1/2}(2I_p)^{1/2}\times\exp\left[-\frac{1}{3}\frac{[2I_p+v_y^2(t_s)]^{3/2}}{E_x(t_s)}\right], \tag{5}$$

$$a_{\mathrm{pr}}(t,t_s)=\left(\frac{2\pi}{t-t_s}\right)^{3/2}\frac{(2I_p)^{1/4}}{E_x(t_s)}\times\exp\left[-i\int_{t_s}^{t}dt'\{\tfrac{1}{2}[\mathbf{p}(t,t')-\mathbf{A}(t')]^2+I_p\}\right], \tag{6}$$

$$a_{\mathrm{rec}}(t)=\mathbf{d}^*[\mathbf{v}]=\frac{C\mathbf{v}(t)}{[I_p+v(t)^2/2]^3}, \tag{7}$$

where C is a numerical constant (see, e.g., [5]). The elipticity dependence of $\mathbf{d}(t)$ enters Eqs. (5)–(7) through the velocity of the electron at the moment of return $\mathbf{v}(t)=\mathbf{p}(t,t_s)-\mathbf{A}(t)$ and the initial transversal energy $v_y^2(t_s)/2$, which the electron must have at the moment of birth t_s in order to return to the parent ion at the moment t:

$$\frac{v_y^2(t_s)}{2}=\frac{E_y^2}{2\omega^2}\left(\cos\omega t_s-\frac{\sin\omega t-\sin\omega t_s}{\omega(t-t_s)}\right)^2. \tag{8}$$

The whole process described by Eqs. (5)–(7) is depicted schematically in Fig. 1. In linearly polarized field the first term Eq. (5) coincides with the tunneling amplitude derived by Keldysh [1]. It decreases with increasing elipticity, since the electron must now be born with substantial transversal energy to ensure its return to the nucleus.

The second term describes the electron propagation in the electric field. It is governed by the exponential phase factor, which traces the electron energy along the path given by classical mechanics. The classical part of the electron trajectory is determined by the conditions $\mathbf{v}_x(t_s)=0$ and $\mathbf{r}(t)=\mathbf{r}(t_s)$. The requirement for zero initial velocity v_x defines the initial position $x(t_s)=I_p/E_x$ and the initial energy $-I_p$ of the ionized electron; see Fig. 1. [Quantum-mechanical propagation under the potential barrier up to the point $x(t_s)$ is contained in Eq. (5)]. The preexponential factor in Eq. (6) describes transversal *spreading* (proportional to $E^{1/2}\tau/I_p^{1/4}$) of the electron wave packet and its longitudinal *stretching* [proportional to $(E\tau)^{1/2}$] by the electric field. Scaling for the transversal spreading is exactly what one obtains using tunneling time and the energy-time uncertainty relation.

We can now use these results to include the modifications to high-harmonic generation in the presence of the Coulomb potential. We first discuss the corrections to the quantum-mechanical processes of ionization and recombination [Eqs. (5) and (7), respectively] and then to the propagation [Eq. (6)].

Tunneling ionization in strong fields has been extensively discussed in literature [2]. The inclusion of the Coulomb potential results in multiplying the tunneling amplitude $a_{\mathrm{ion}}(t_s)$, Eq. (5), by a preexponential factor

$$\sqrt{C_{n^*lm}\{4[2I_p+v_y^2(t_s)]^{3/2}/E_x(t_s)\}^{2n^*-m-l}},$$

where n^* is the effective principal quantum number and C_{n^*lm} is the numerical factor given in Ref. [2].

The recombination amplitude, Eq. (7) in the Keldysh approximation, is given by the dipole matrix element $\mathbf{d}(\mathbf{v})$ between the atomic ground state and a plane wave (not scattered by the Coulomb potential). The main aspects of the Coulomb corrected matrix element can be derived based on symmetry considerations. The matrix element $\mathbf{d}(\mathbf{v})$ is a vector, and due to the spherical symmetry of the ground-state wave function its direction is determined by the velocity direction of the incoming wave, which at infinite distance from the Coulomb center is a plane wave $|\mathbf{v}\rangle$. Consequently, $\mathbf{d}(\mathbf{v})$ must reverse the sign for left- and right-incoming waves $\mathbf{d}(-\mathbf{v})=-\mathbf{d}(\mathbf{v})$ and can be casted in a general form $\mathbf{d}(\mathbf{v})=\mathbf{v}f(v)$, where the scalar $f(v)$ depends on $|\mathbf{v}|$ and contains the Coulomb corrections.

The *absolute yield* of any given harmonic depends on the exact form of $f(v)$. However, in first approximation, the *ellipticity dependence* is independent of the particular form of f and is due to the vector part of the dipole moment, as discussed below. For every given harmonic $N\omega$ the stationary phase (saddle) point of the corresponding Forier integral of Eq. (4) is realized when $I_p+v^2(t)/2\approx N\omega$ [5] and hence $f(v)$ can be taken out of the integral at this stationary phase (saddle) point. We performed calculations with $f(v)=1$ and $f(v)=C/(2I_p+v^2)^3$ and found a negligible difference between the two models for the ellipticity dependence, while in the absolute yield the difference was quite significant.

The propagation of the electron in the presence of the Coulomb potential is corrected in the following way. The exponent in a_{pr} contains the classical expression for the energy of a free electron in an electric field $\exp[iS(t_s,t)]$. Here $S=\int_{t_s}^{t}dt'H(t')$ is the classical action and H is the Hamiltonian. This correspondence suggests the use of time-dependent quasiclassical perturbation theory to include the effect of the Coulomb potential. The exponent in Eq. (6) is modified to $S+\Delta S=\int_{t_s}^{t}dt'\{1/2[\mathbf{p}(t,t_s)-\mathbf{A}(t')]^2+I_p-1/r(t')\}$. The energy change due to the Coulomb potential is evaluated along the electron trajectory $r(t')$ in the electric field; Coulomb modifications to the electron path are small and can be neglected to first approximation. To evaluate the role of this correction for threshold harmonics let us find the derivative of the potential-energy contribution to the classical action. Using the relation $r(t)=r(t_s)$, the derivative is $[I_p-1/r(t_s)](1-dt_s/dt)$, where $1/r(t_s)\approx 1/x(t_s)=E_x(t_s)/I_p$. For threshold harmonics the moments of birth correspond to $\cos\omega t_s\approx 1$, $E_x(t_s)\approx E_x$. Consequently, the presence of the Coulomb potential results in an effective reduction of the ionization potential in the exponent of Eq. (6) to $I_c=I_p-E_x/I_p$. Note that the Stark shift of the ground state also has to be included. In the quasistatic approximation $\Delta I_p\approx 0.5\alpha E_x^2\cos^2\omega t$, where α is the static polarizability, $\alpha\approx 2.7$ for Ne.

Applying the corrected Coulomb potential in Eq. (6) and replacing $f(v)$ with a constant, numerical Fourier transformation of Eq. (4) yields the dipole moment of the nth harmonic $|\mathbf{d}_n(\epsilon)|^2$. The parameter $\epsilon=|E_y/E_x|$ denotes the ellipticity of the laser field. In the following, we compare the theoretical results to the experimental data described earlier in the paper. The parameters for the numerical evaluation are $I_p=21.56$ eV, 754-nm laser wavelength, and laser intensity $I=4\times10^{14}$ W/cm^2. According to the propagation results of Ref. [12], in our experimental conditions this intensity optimizes the single-atom response and the phase-matching conditions in laser-produced plasma with rapidly changing density (propagating plasma front).

In terms of harmonic orders I_p lies between $N=13$ and 15. Figure 2 shows the comparison of theoretical and experimental data for the total emission of harmonic orders $N=13$–19. For each harmonic an individual scaling factor has been introduced to match the overall magnitude of the theoretical and the experimental data. Good agreement is obtained for all curves. The ellipticity dependence of the 13th and the 15th harmonic provides an important test for the Coulomb correction we introduce. It is found that inclusion of the Coulomb correction is essential to obtain the dip in the ellipticity dependence for $N=13,15$. Repetition of the calculation with I_p instead of I_c shifts the dip to $N=17$.

The different ellipticity behavior for $N=13,15$ results from an interference effect. Classically, these harmonics are generated by an electron born close to the peak of the electric field, which returns to its parent ion with low kinetic energy. The first and the second return succeed immediately. During the second return the sign of the electron momentum and of the dipole moment is reversed. Superposition of the two returns results in a negative interference of the dipole moments and therewith, in a suppression of high-harmonic emission in the direction parallel to the main polarization axis of the laser field. Hence, for increasing ellipticity, the perpendicular component can dominate the parallel component of harmonic emission resulting in an increase of the total high-harmonic emission.

Concluding, we have introduced an approach to account for the effect of the Coulomb potential on the electron evolution in intense laser fields after tunneling ionization. This formalism has been applied to study the ellipticity dependence of high-harmonic generation. We have presented experimental data on the ellipticity dependence of the threshold harmonics $N=13$–19, generated in neon with a 754-nm, 150-fsec laser pulse. Our approximate approach has been supported by a comparison to these experimental data. We explained the physics responsible for unusual ellipticity dependence of the threshold harmonics and found that inclusion of the Coulomb potential is essential in order to obtain quantitative agreement with experimental data.

The authors are grateful for helpful discussions with P. B. Corkum. This work was partially supported by the Österreichische Nationalbank Jubiläumsfondprojekt No. 5124. T. B. is supported by the Österreichische Akademie der Wissenschaften, APART Grant. M. Yu. I. is supported by a NSERC collaborative research grant.

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