High Harmonic Generation Beyond the Electric Dipole Approximation

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A generalization of the analytical theory of high harmonic generation in the long wavelength limit and in the single active electron approximation is developed taking into account the magnetic dipole and electric quadrupole interaction. Quantum mechanical and classical theories are found to be in excellent agreement, which allows one to explain the influence of multipole effects in terms of an intuitive picture. For Ti:S lasers (0.8 μm) multipole contributions are found to be small below an intensity of about 10^{17} W/cm^2, at which harmonic radiation with photon energies of several keV is generated. This promises the extension of high harmonic generation well into the sub-nm wavelength regime.

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High harmonic generation (HHG) in noble gases [1] is a promising source for the generation of coherent XUV (extreme ultraviolet) radiation that is about to open new research fields, such as XUV nonlinear optics [2] and attosecond pump-probe spectroscopy [3]. The shortest harmonics generated so far have a photon energy of 0.5 keV [4,5]. Currently, great effort is invested to extend HHG into the x-ray regime with photon energies above 1 keV, where further ground breaking applications would become possible. For example, the short pulse duration and high coherence of a few-keV harmonic source would make time resolved x-ray spectroscopy experiments [6] on an unprecedented time scale possible, allowing the observation of fundamental dynamical processes in matter, as, for example, the making and breaking of chemical bonds.

At present the main limitation for HHG at wavelengths below 10 nm is due to free electron induced dephasing between fundamental and harmonic field [2]. However, there are indications that the limitations introduced by dephasing can be overcome. Phase matched harmonic growth was already demonstrated experimentally at low harmonic orders [7] and, recently, numerical calculations revealed a phase matching mechanism that is predicted to work efficiently in the above keV photon energy range [8]. Assuming that the phase matching problem can be solved, the question arises of which is the shortest wavelength that can be generated by HHG in principle. The ultimate limitation of HHG arises from the large velocity and therewith from the large electron excursion amplitude required for the generation of extremely high harmonics. At a laser intensity of 2×10^{16} W/cm^2, necessary for the generation of above keV harmonic radiation, the variation of the laser field over the excursion amplitude α_0 becomes of the order of $k\alpha_0 \approx 0.1$, with k being the absolute value of the laser wave vector. Then multipole contributions have to be taken into account [9]. While in the electric dipole approximation used commonly for the analysis of HHG [10] the ionized electron performs a quiver motion exclusively in the direction of the laser polarization, multipole effects introduce an additional force in the direction of the wave vector of the laser field. As a result, the center of the wave packet tends to miss the parent ion, which can lead to a drastic reduction of the efficiency of HHG.

In the present paper we develop an analytical solution of the 3D Schrödinger equation for a single atom (ion) in a strong laser pulse, including lowest order multipole effects, which comprise the magnetic dipole and the electric quadrupole contribution. The solution is used to quantify the limitations of HHG introduced by multipole effects. Our theory generalizes previous theories of HHG relying on the electric dipole approximation [11,12]. The results are found to be in excellent agreement with a perturbative solution of the classical equation of motion, which corroborates the validity of our analysis and makes a simple interpretation of the quantum mechanical theory possible. The quantum mechanical calculation yields a simple formula that determines the onset of multipole contributions to HHG. For example, for HHG with a Ti:sapphire laser in He$^+$ ions multipole effects are small for intensities below 10^{17} W/cm^2 allowing in principle the generation of harmonic radiation with photon energies of several keV.

The starting point of our derivation is the Schrödinger equation for a single electron atom in cgs units and in the velocity gauge given by

$$i\hbar\partial_t\Psi = \frac{1}{2m}\left[i\hbar\nabla + \frac{e}{c}\mathbf{A}(\mathbf{r},t)\right]^2\Psi - \frac{Ze^2}{r}\Psi, \quad (1)$$

where m, e, and Z are the electron mass, electron charge, and nuclear charge, respectively, Ψ is the wave function, $\nabla = (\partial_x, \partial_y, \partial_z)$, and $\partial_{t,x,y,z}$ refer to the partial time and space derivatives, respectively. The vector potential is related to the electric and magnetic fields by $\mathbf{E}(\mathbf{r},t) = -(1/c)\partial_t\mathbf{A}(\mathbf{r},t)$ and $\mathbf{B}(\mathbf{r},t) = \nabla \times \mathbf{A}(\mathbf{r},\mathbf{t})$, respectively. We assume that the laser field is polarized in the x direction and propagates in the z direction, $\mathbf{A}(\mathbf{r},t) = (A(z,t),0,0)$. This gives for the electric and magnetic field $\mathbf{E}(\mathbf{r},t) = (E(z,t),0,0)$ and $\mathbf{B}(\mathbf{r},t) = (0,B(z,t),0)$.

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The Schrödinger equation (1) is solved by expanding the vector potential to first order in the space coordinate, $\mathbf{r} = (x, y, z)$, which results in $A(z, t) = A(t) + z\partial_z A(t)$. Throughout the paper we use the notation $F(z = 0, t) = F(t)$ for $F = A, \partial_z A, E, B$. The first order expansion of A corresponds to taking into account the magnetic dipole and the electric quadrupole interaction. Relativistic effects, such as the mass correction, can be neglected up to $v/c \approx 0.2$, which is realized at a Ti:sapphire peak intensity of $\approx 10^{17}$ W/cm^2. Here c is the vacuum light velocity, $v \approx E_0/\omega$ is the electron velocity, E_0 is the peak electric field, and ω is the circular frequency.

For the solution of Eq. (1) we use the ansatz

$$\Psi(\mathbf{r}, t) = \exp\left(\frac{iW_b t}{\hbar}\right)\left(|\tilde{0}\rangle + \int d^3\mathbf{P}\, b(\mathbf{P}, t)\, |P\rangle\right), \quad (2)$$

where $-W_b$ is the energy of the ground state and $b(\mathbf{P}, t)$ is the amplitude of the (plane wave) continuum state $|P\rangle = (2\pi\hbar)^{-3/2}\exp(i\mathbf{P}\mathbf{r}/\hbar)$. Note that $\mathbf{P}$ is the canonical momentum, which is related to the classical momentum by $\mathbf{p} = \mathbf{P} - (e/c)[\mathbf{A}(t) + (\mathbf{r}\nabla)\mathbf{A}(t)]$. The ground state of the Schrödinger equation (1) in the velocity gauge $|\tilde{0}\rangle$ differs from the ground state obtained in the length gauge $|0\rangle$. The two ground states are related by the gauge transformation $|\tilde{0}\rangle = |0\rangle\exp[i\chi(\mathbf{r}, t)]$, where $\chi = e/(\hbar c)[xA(t) + (1/2)xz\partial_z A(t)]$. In order to be consistent with the expansion of A in Eq. (1), the vector potential in χ and in the definition of $\mathbf{p}$ was also expanded to first order in $\mathbf{r}$. For the fully $\mathbf{r}$-dependent expressions see, for example, Ref. [13]. The correctness of our perturbative approach was checked by transforming Eqs. (1) and (2) to the length gauge. In the limit of the dipole approximation the ansatz (2) becomes equivalent to the ansatz used by Lewenstein *et al.* in the length gauge [11]. Applying the gauge transform to the first order expansion of Eq. (1) gives the Schrödinger equation in the length gauge, where the laser electron interaction is determined by the potential $H_i = H_{ed} + H_{eq} + H_{md}$. Here $H_{ed} = -exE(t)$, $H_{eq} = -(xz/2)\partial_z E(t)$, and $H_{md} = i\hbar e/(2mc)B(t)(z\partial_x - x\partial_z)$ are the electric dipole, electric quadrupole, and magnetic dipole moment, respectively.

Integration of Eq. (1) by using the ansatz (2) yields

$$b(\mathbf{V}, t) = -\frac{i}{\hbar}\int_{-\infty}^{t} dt' \langle \mathbf{V} - \mathbf{T}(t')\,|H_i(t')|\tilde{0}\rangle \times \exp\left[-\frac{i}{\hbar}S(\mathbf{V}, t, t')\right], \quad (3)$$

where the quasiclassical action $S(\mathbf{V}, t, t')$ is given by

$$S(\mathbf{V}, t, t') = \int_{t'}^{t} dt'' \times \left\{\frac{1}{2m}\left[\mathbf{V} - \mathbf{T}(t'') - \frac{e}{c}\mathbf{A}(t'')\right]^2 + W_b\right\}. \quad (4)$$

For the integration of Eq. (1) we have used the variable transformation $\mathbf{V} = \mathbf{P} + \mathbf{T}(\mathbf{t})$ with $T_x = 0$, $T_y = 0$, and $T_z = -e/(mc)\int dt\,[P_x - (e/c)A(t)]\partial_z A(t)$. We assumed that the vector potential may be written as $A(z, t) = A(t - z/c)$; i.e., vacuum propagation dominates and propagation effects such as ionization induced phase changes are small. Then the time integral in T_z can be solved resulting in $T_z = e/(mc^2)[P_x A(t) - (e/2c)A^2(t)]$. Finally, note that the multipole contributions $\langle \mathbf{V} - \mathbf{T}(t')\,|H_i(t')|\tilde{0}\rangle$ in Eq. (3) arise from the gauge transformed ground state $|\tilde{0}\rangle$.

The multipole corrections to HHG in the exponent of Eq. (3) dominate over those in the prefactor. Therefore contributions from the transition matrix elements H_{md} and H_{eq} are neglected in the prefactor to give $\langle \mathbf{V} - \mathbf{T}(t')\,|H_{ed}(t')|\tilde{0}\rangle$. To the same order of approximation the emission of HHG in the direction of the laser field is determined by the second time derivative of the dipole moment $\langle \Phi(\mathbf{r}, t)\,|\ddot{\mathbf{r}}|\Phi(\mathbf{r}, t)\rangle$, and higher order contributions arising from the magnetic dipole and electric quadrupole terms can be neglected [14]. The electric dipole moment including the multipole corrections in the exponent is given by

$$x_m(t) = \int d^3\mathbf{V}\,\langle\tilde{0}|x|\mathbf{V} - \mathbf{T}(t)\rangle b(\mathbf{V}, t) + \text{c.c.} \quad (5)$$

Our calculation is limited to the long wavelength regime $\gamma = \omega(2mW_b)^{1/2}/(eE_0) \ll 1$, where γ denotes the Keldysh parameter [15]. In this limit tunneling is the dominant ionization mechanism, and the integrals in Eq. (5) can be evaluated by a stationary phase integration over P [11], and saddle point integration with respect to t, where a singularity at the saddle point must be taken into account. The equation determining the real part of the saddle point t_0 is given by $P_x(t, t_0) - (e/c)A(t_0) = 0$, where $P_x(t, t_0) = e/[c(t - t_0)]\int_{t_0}^{t} A(t')\,dt'$. Here t_0 can be interpreted as the time of birth of the electron trajectory which returns at time t to the parent ion and generates high harmonic radiation. Calculation of the integrals yields the final result

$$x_m(t) = \sum_{t_0} \eta \exp\left[-\frac{(P_{zd}^2 + 2mW_b)^{3/2}}{3m\hbar e|E(t_0)|}\right] \times \exp\left[-\frac{i}{\hbar}(S_d + S_m)\right] + \text{c.c.}, \quad (6)$$

where the real exponential function arises from the ionization probability at time t_0 and the imaginary exponential function determines the phase picked up by the electron along its trajectory between birth and recombination time. The sum extends over all birth times t_0, but in practice, for given t, only a few birth times t_0 make a significant contribution. The phase is determined by the classical action $S_d(t, t_0) = \int_{t_0}^{t} dt'\,(1/2m)[P_x(t, t') - (e/c)A(t')]^2 + W_b$ calculated in the electric dipole approximation and by the multipole contribution to the classical action, $S_m(t, t_0) = (1/2m) \int_{t_0}^{t} dt' \{P_z(t, t') - e/(mc^2)[P_x(t, t') - e/(2c)A^2(t')]\}^2$. Further, the momentum $P_{zd} = -P_z(t, t_0) + 1/(2mc)P_x^2(t, t_0)$ and $P_z(t, t_0) = e^2/(mc^3)\{A^2(t_0) - 1/[2(t - t_0)] \int_{t_0}^{t} A^2(t')\, dt'\}$. Finally, as we have neglected multipole effects in the preexponential factor, the constant η in Eq. (6) refers to the electric dipole preexponential expressions for HHG, as, for example, given in Ref. [2].

The quantum mechanical result (6) has an intuitive classical interpretation that corroborates the validity of our analysis; see Fig. 1. This becomes evident from a perturbative solution of the classical equation of motion $m\ddot{\mathbf{r}} = e\mathbf{E} + (e/c)\mathbf{v} \times \mathbf{B}$. For $v/c \ll 1$ terms proportional to

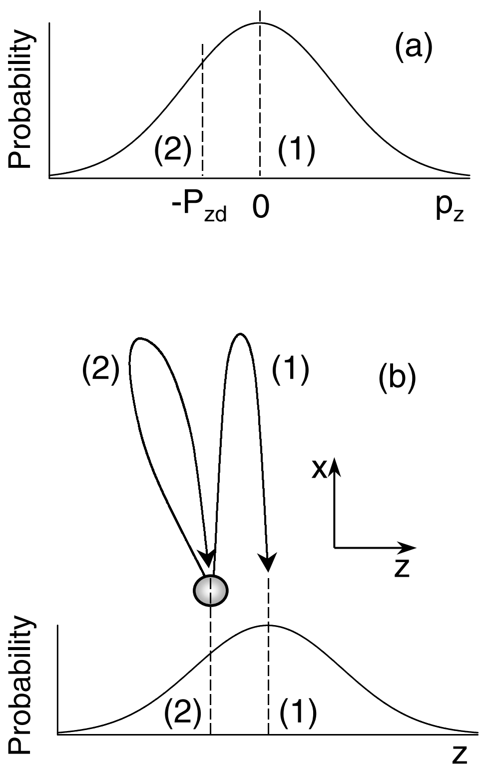

FIG. 1. Schematic diagram of the influence of the Lorentz force on HHG. The laser field is polarized in the x direction and propagates in the z direction. (a) The momentum distribution of the electron wave packet at its time of birth. The electron trajectories corresponding to the initial momenta (1) and (2) in (a) are plotted in (b). The electron trajectory (1) is born with zero velocity in the z direction. During oscillation in the laser polarization direction the Lorentz force pushes the electron in the z direction. As a result it misses the parent ion during its return and HHG cannot take place. In contrast to that electron trajectory (2) is born with an initial momentum $p_z(t_0) = -P_{zd}$, which exactly compensates the effect of the Lorentz force so that the electron directly returns to the nucleus and HHG takes place. The ionization probability of (2) is lower than the peak ionization probability for $p_z(t_0) = 0$, which results in a reduction of HHG due to the Lorentz force. The lower graph in (b) shows the relation between classical and quantum mechanics. The wave function of the returning electron is plotted versus z. The peak of the wave function which is shifted in z corresponds to the electron trajectory (1). The part of the wave function that overlaps with the nucleus corresponds to trajectory (2).

B may be neglected in the equation along the x direction. Integration subject to the initial condition, that the electron is born with zero velocity at time t_0, determines the momentum of the electron in the x direction as $m\dot{x}(t) = p_x(t) = -(e/c)[A(t) - A(t_0)]$. To obtain the momentum and motion in the z direction, p_x is inserted into the equation of motion for $\ddot{z}$. Integrating twice we obtain $m[z(t) - z(t_0)] = 1/(2mc) \int_{t_0}^{t} p_x^2(t')\, dt' + p_z(t_0)(t - t_0) = [P_{zd} + p_z(t_0)](t - t_0)$, where $p_z(t_0)$ refers to the electron birth momentum in the z direction. Efficient HHG requires recollision of the electron with the parent ion, i.e., $z(t) = z(t_0)$, so that the electron initial momentum is determined as $p_z(t_0) = -P_{zd}$ with $P_{zd} \geq 0$. The probability for starting the electron with nonzero initial momentum $-P_{zd}$ is given by quantum mechanical tunneling theory [16] as $\exp[-(P_{zd}^2 + 2mW_b)^{3/2}/(3m\hbar e|E(t_0)|)]$. This is in exact agreement with the full quantum mechanical result, Eq. (6).

The dependence of the real exponential factor in Eq. (6) on P_{zd} leads to a decrease of the high harmonic yield. The magnitude of the decrease close to the harmonic cutoff, where the quiver energy and therewith the multipole effects are largest, can be estimated using the relation $P_{zd}^2(t) \approx U_p^2/c^2$, which was verified by direct numerical evaluation. Here $U_p = (eE_0)^2/4m\omega^2$ is the ponderomotive potential, and E_0 denotes the peak electric field. Inserting the inequality in Eq. (6), expanding the term $(U_p^2/c^2 + 2mW_b)^{3/2}$, and substituting $|E(t_0)|$ by E_0 for harmonics close to the cutoff, the ratio of multipole to electric dipole moment close to the cutoff is found to be

$$\left|\frac{x_d}{x_m}\right|^2 \approx \exp\left[\frac{\alpha_0 U_p}{4a_0 mc^2}\right], \tag{7}$$

where $\alpha_0 = eE_0/(m\omega^2)$ is the electron excursion amplitude in the direction of the laser polarization and $a_0 = \hbar/\sqrt{2mW_b}$ is the Bohr radius. The influence of multipole effects on HHG scales with the ratio of the electron ponderomotive energy to its rest energy.

As a numerical example, based on Eq. (6) we have calculated HHG in He^+ ions ($W_b = 54.4$ eV) with the following laser parameters: $\lambda_0 = 0.8\ \mu$m, full width at half maximum pulse duration $\tau_p = 5$ fs, peak intensity $I_0 = 5 \times 10^{16}$ W/cm^2, and a sech pulse shape. Figure 2 shows the (Fourier transformed) second derivative of the dipole expectation value calculated in dipole approximation ($|\ddot{x}_d|^2$, dotted line) and calculated including multipole effects in lowest order ($|\ddot{x}_m|^2$, solid line). Comparison of the two spectra shows that frequency dependent changes of the harmonic yield around the cutoff, which are mainly due to the multipole phase S_m, remain small with only a slight shift of the cutoff frequency towards higher harmonics. The effect of the Lorentz force increases with electron velocity and therewith is stronger for higher harmonic orders. As a result, the ratio between the electric dipole and the multipole harmonic spectrum increases for higher harmonic orders. The maximum ratio in Fig. 2 is $\approx$2, revealing that multipole effects are insignificant in

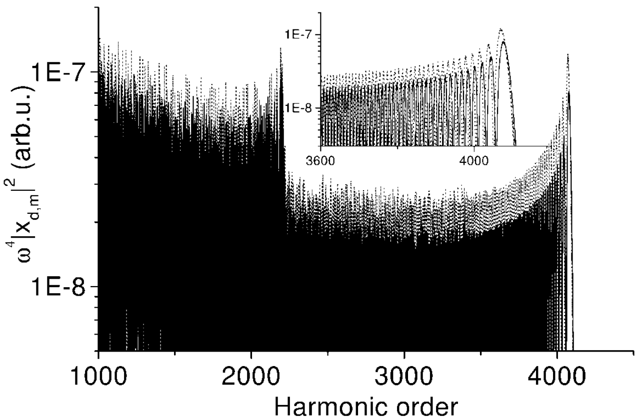

FIG. 2. HHG in He^+ ions ($W_b = 54.4$ eV) with the following laser parameters: $\lambda_0 = 0.8$ μm, full width at half maximum pulse duration $\tau_p = 5$ fs, peak intensity $I_0 = 5 \times 10^{16}$ W/cm^2, and a sech pulse shape. The dotted and the full lines denote the Fourier transform of the second derivative of the electric dipole ($|\ddot{x}_d|^2$) and of the multipole ($|\ddot{x}_m|^2$) moment, respectively. The inset shows a part of the harmonic spectrum close to the cutoff between $N = 3600$ and 4200.

this parameter range. Inserting the parameters of Fig. 2 in Eq. (7) we find $|x_d/x_m|^2 \approx 3$ in fair agreement with the numerical result. Equation (7) reveals that $|x_d/x_m|^2 \approx 20$ for $I_0 = 10^{17}$ W/cm^2; i.e., multipole limitations of HHG may become severe in an intensity range, where relativistic effects must be taken into account. Finally, the plateau in Fig. 2 extends at nearly constant intensity to a harmonic order $N = 4000$ which corresponds to an x-ray photon energy of 6 keV. As a result, HHG holds in principle the potential for the realization of a revolutionary pulsed x-ray source that supplies ultrashort (sub-10 fs) pulses in the several keV region. To determine whether this single atom response can be exploited for efficient harmonic generation, effects of propagation in the medium must be included. First studies at somewhat lower intensities indicated the possibility of phase matching for above keV harmonic radiation [8]. Extension of these results to the present intensities will be the subject of future investigations.

In conclusion, the dominant multipole effect in high harmonic generation is the radiation pressure. This mechanism is not limited to harmonic generation, but will equally affect any process in atomic physics that depends on the recollision of the electron with the parent ion [10], such as nonsequential laser ionization and above threshold ionization. The analytic theory presented here gives a convenient and intuitive description of multipole effects in strong field atomic physics without compromising with respect to the physical model, most importantly by keeping all three spatial dimensions. This is particularly important, since complete numerical calculations in the intensity regime where multipole and relativistic effects become dominant are extremely demanding and still unchallenged in three dimensions. Finally, our method not only can quantify the dominant multipole effects, but also holds the potential to account for relativistic corrections based on the Volkov solutions of the Dirac equation [17].

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