Numerical Characterization of High Harmonic Attosecond Pulses

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A numerical simulation of attosecond harmonic pulse generation in a three-dimensional field-ionizing gas is presented. Calculated harmonic efficiencies quantitatively reproduce experimental findings. This allows a quantitative characterization of attosecond pulse generation revealing information currently not accessible by experiment. The rapid phase variation and spatiotemporal distortions of harmonics are smaller than anticipated, allowing focusing of 30-nm, 750-as pulses to intensities in excess of 10^{13} W/cm^2. Feasibility of such pulses brings novel applications such as extreme ultraviolet nonlinear optics and attosecond pump probe spectroscopy within reach.

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A number of methods have been proposed for attosecond(as)-pulse generation [1]. High harmonic generation (HHG) is currently the experimentally furthest advanced method that can supply single as-pulses [2,3], which is essential for potential applications. While the femtosecond regime presents the natural time scale of electronic motion in the mean field potential of solids and molecules, as-pulses will allow the temporal resolution of correlated electron processes, such as dephasing in metals or core electron processes (e.g., Auger decay) in atoms. Moreover, the high peak intensities associated with short pulse durations are expected to allow the extension of nonlinear optics to the extreme ultraviolet (xuv) regime [4].

One of the main obstacles for the realization of such applications is the lack of experimental tools for the characterization of harmonic as-pulses. Although nonlinear autocorrelation [4] and linear cross-correlation methods [5] for harmonic pulse measurement were proposed and partly realized, no single as-pulses were measured so far. Consequently, not even the peak intensity of as-pulses is experimentally accessible to date. A complete experimental characterization of harmonic pulses that includes spatiotemporal phase and envelope appears to be out of reach for the nearer future. As a result, important problems and questions have remained unanswered. For example, the position of the harmonic as-pulse relative to the laser front changes with the transversal variation of the laser intensity, thus smearing out the as-time structure. Further, harmonic radiation exhibits a rapidly oscillating phase, which may lead to severe spatiotemporal distortions during propagation or focusing, destroying as-resolution. Finally, a central question for as-pump-probe spectroscopy is whether sufficient intensities can be delivered at xuv wavelength.

The absence of experimental methods calls for the development of numerical models capable of supplying detailed, quantitative information on as-pulse generation, which is the purpose of the present paper. A realistic simulation of HHG must account for the interplay between ionization, harmonic generation, absorption, and propagation on a quantitative level. This requires a self-consistent solution of the propagation equations [6], taking properly account of macroscopic (propagation) effects, together with a quantitatively accurate calculation of the microscopic dipole moment of HHG [7]. Although each of these problems was already treated seperately [6,7], a combined solution of both was beyond the reach of current computer capacities so far. To this end, we combine for the first time a self-consistent solution of the macroscopic three-dimensional Maxwell equations with the response of the medium on an atomic level, where the atomic response is drawn from solutions of the time-dependent Schrödinger equation of three-dimensional models for the respective gas species. Our approach unifies and generalizes previous numerical approaches to give the first self-consistent, three-dimensional analysis of HHG. The calculated conversion efficiencies for various noble gases are found to be in good agreement with experiment, demonstrating that our numerical model presents a valid tool for the quantitative characterization of harmonic as-pulse generation.

Going beyond what is accessible by present experimental methods, we find that harmonics are generated in pulses of unexpectedly favorable characteristics. Spatiotemporal effects are smaller than anticipated [8] allowing the focusing of 750-as pulses down to a beam radius of 1.5 μm without significant spatiotemporal distortions, where peak intensities approach 10^{14} W/cm^2. This intensity is sufficient for nonlinear optics and as-pump-probe spectroscopy in the 20–30 nm wave length range.

For the numerical analysis of HHG, Maxwell's equations are solved in the form introduced in Ref. [9] for the laser electric field E_l

$$\partial_\xi E_l(\xi,\tau) - \hat{D}E_l(\xi,\tau') = -\frac{1}{2c}\int_{-\infty}^{\tau}\omega_p^2(\xi,\tau')E_l(\xi,\tau')\,d\tau' - \frac{W_b}{2\varepsilon_0 c}\,\frac{\partial_\tau n_e(\xi,\tau)}{E_l(\xi,\tau)} - \frac{\zeta^{(1)}}{c}\,\partial_\tau[1-n_e(\xi,\tau)]E_l(\xi,\tau)\,, \quad (1)$$

and for the harmonic electric field E_h,

$$(\partial_\xi + \alpha_h)E_h(\xi,\tau) - \hat{D}E_h(\xi,\tau') = \frac{-1}{2\varepsilon_0 c}\,\partial_\tau P_h[E_l(\xi,\tau)] + \text{c.c.} \quad (2)$$

We have introduced a coordinate frame moving at the speed of light in vacuum, $\tau = t - z/c$, $\xi = z$. The electric field is chosen to propagate in z direction and is polarized in the x direction; for the transversal profiles cylindrical symmetry is assumed. Diffraction is determined by the operator $\hat{D} = (c/2)\nabla_\perp^2 \int_{-\infty}^{t} dt'$. Inclusion of diffraction, self-defocusing, and self-phase modulation by the presence of free electrons is crucial for a correct treatment of the highly nonlinear propagation of the fundamental laser pulse. For the parameters of interest here, diffraction of the harmonic pulse is negligible and $\hat{D} = 0$ in Eq. (2).

Equation (1) is solved subject to the initial condition $E_l(0,\tau) = -dA_l/d\tau$, with $A_l(\tau) = E_0/\omega_0 \exp[-(r/a)^2]\,\mathrm{sech}(1.76\tau/\tau_p)\sin(\omega_0\tau)$. Here, E_0, ω_0, τ_p, and a represent the peak electric field strength, laser center frequency, FWHM (full width at half maximum) pulse duration, and beam waist, respectively. In Eq. (2), α_h denotes the xuv absorption coefficient and $P_h = \kappa k n_a \times d_h(E_l)$, the high frequency part of the macroscopic polarization, is determined by the laser electric field E_l resulting from Eq. (1). Here, n_a is the density of atoms, $k = 8.4773 \times 10^{-30}$ C m is the conversion factor between atomic and SI units, and the single atom dipole moment d_h in atomic units is determined by Eqs. (34)–(39) in Ref. [9]. The parameter κ, which corrects for the presence of the Coulomb potential during HHG, is discussed below. Further, the plasma frequency is given by $\omega_p = (e^2 n_e/m\epsilon_0)^{1/2}$, and c, e, m, ε_0, W_b, and $\zeta^{(1)}$, refer to the vacuum light velocity, electron charge, electron mass, permittivity of free space, atomic ionization potential, and linear susceptibility of neutral atoms, respectively. The density of free electrons is determined by

$$n_e(\xi,\tau) = n_a\left(1 - \exp\left[-\int_{-\infty}^{t} d\tau'\, w\{E_l(\xi,\tau')\}\right]\right), \quad (3)$$

where $w(E_l)$ is the optical field ionization rate.

For the given laser parameters, the wave equations (1) and (2) are practically identical with Maxwell's equations [9]. However, the microscopic model contains approximations which must be corrected for in order to determine absolute numbers of the harmonic efficiency. Our calculation of the single atom dipole moment is based on an extension of the model of Lewenstein *et al.* [10]. The original approach relies on the Keldysh approximation [11], i.e., the effect of the Coulomb potential on the ionized part of the electron wave function is neglected. Although qualitatively this was demonstrated to give excellent results [9], no quantitative predictions of HHG can be made at this level of approximation. HHG consists essentially of three steps, which are optical field ionization, propagation of the free electron in the continuum, and recombination to the ground state of the parent ion [12]. Omission of the Coulomb potential introduces an error in all three steps. In Ref. [13] optical field ionization in d_h was corrected for the presence of the Coulomb potential [9]. Coulomb corrections for the remaining two steps are determined from numerical solutions of the three-dimensional Schrödinger equation in the single active electron approximation [14]. In that model the valence electron moves in the combined fields of the laser pulse and of the atomic core. The core electrons are assumed to remain, unaffected by the laser field, in their ground-state orbitals. The core potential can be determined either by a Hartree Fock-type method [14], or by using a shielding potential [15]. We use the second method which was shown to reproduce experimental findings in great detail [16]. The parameters of the shielding potential are optimized to correctly reproduce the ground state energy and the transition energies to the first excited s- and p state. Solution of the time-independent Schrödinger equation [17] for the optimized potential yields the static ionization rates w for Ar and Ne; see Fig. 1. These ionization rates are used for the calculation of n_e in Eq. (3) and d_h in Eq. (35) of Ref. [9]. Comparison of the dipole moment d_h obtained from our strong field model with a numerical solution of the time-dependent Schrödinger equation for various laser parameters yields a Coulomb correction factor for continuum propagation and recombination. The correction factors for Ar and Ne are found to be $\kappa = 1.4, 2.4$, respectively. The parameter κ varies by only 30% over the relevant range of intensities and harmonic orders and, therefore, was assumed to be constant. We tested our model by comparing the numerical results with the experimental data on the conversion efficiencies in Ar and Ne [9], see Fig. 2. The parameters are $I_0 = 3.5 \times 10^{15}$ W/cm^2/8.8×10^{14} W/cm^2, $\tau_p =$ 7 fs/7 fs, $a = 30$ μm/60 μm, propagation length 3 mm/3 mm, $n_a = 7.9 \times 10^{18}$ cm^{-3} (225 Torr)/1.05×10^{19} cm^{-3} (300 Torr), and $\zeta^{(1)} = 8.3 \times 10^{-4}/2.64 \times 10^{-5}$ [18] for Ar and Ne, respectively. The frequency

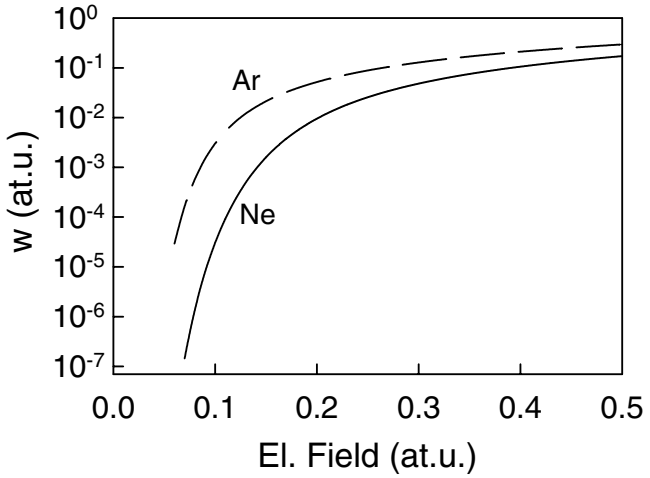

FIG. 1. Ionization rate in atomic units versus electric field strength in atomic units. The full and dashed lines refer to Ne and Ar, respectively.

dependent xuv absorption coefficient α_h is taken from the home page of the Center for X-Ray Optics, Materials Sciences Division, Lawrence Berkeley National Laboratory [19]. Note that here we have specified the laser peak intensity, whereas in Ref. [9] an averaged intensity was given to facilitate a comparison to one-dimensional calculations.

Our calculations are compared with experiments in Fig. 2, which present the first major result of our paper. Despite uncertainties in the experimental parameters the overall agreement is quite satisfactory, showing that our model is capable of characterizing HHG in terms of absolute numbers. The agreement between experimental and numerical data is accurate to within a factor 2–3 for a broad range of harmonic orders. The deviation for low harmonic orders in Argon depends on the absorption data in the region below $N = 27$, where absorption increases drastically. A reduction of the absorption coefficient by a factor of 2 gives agreement with experiment within a factor of 3. The discrepancy in the cutoff region of Ne arises from uncertainties in the experimental parameters. Either a reduction of the refractive index by a factor of 2 or a reduction of the gas pressure by 30% would result in an increase of the signal in the cutoff region by a factor of 4. The lower harmonics are not affected by these uncertainties. Except for the absorption coefficient, the most important theoretical input in our calculations is the ionization rates. Allowing for an uncertainty of a factor of 2 in these rates affects the conversion efficiencies in Fig. 2 by less than a factor of 3.

The good agreement between experiment and theory opens the possibility to perform a quantitative analysis of harmonic as-pulses, giving duration, peak intensity, temporal and spatial shape, and phase of harmonic pulses. We choose the following parameters: 2 mm propagation distance in Ar, $n_a = 1.75 \times 10^{19}$ cm^{-3} (500 Torr), $\tau_p =$ 5 fs, $a = 150$ μm, $I_0 = 1.6 \times 10^{14}$ W/cm^2, $\lambda_0 = 0.8$ μm. A few-cycle laser pulse generates a smooth harmonic spectrum close to the cutoff, which corresponds to a single as-pulse [3,20]. The laser intensity is chosen in such a way that the cutoff lies around $N = 30$, where HHG is most efficient; see Fig. 3. The steep drop of the low frequency part of the spectrum is due to the abrupt rise of the absorption coefficient below $N = 27$. Apart from small fluctuations [21] the spectrum is smooth across the pulse cross section and ranges over approximately 15 harmonics. The Fourier transform of the spectrum for $N > 27$ corresponds to a 200-as pulse. As the best available xuv mirrors have a bandwidth covering only about 3 harmonic orders [22], the full bandwidth in Fig. 3 cannot be utilized for focusing. The finite spectral width of the mirror is taken into account by filtering the spectrum with a flat top profile extending from $N = 27$ to $N = 30$. We assume here ideal focusing by a quadratic mirror with focal length $f = 18$ mm. Limitations imposed by aberration and mirror dispersion on as-pulse generation will be subject to further investigations. The resulting pulse is depicted in Fig. 4. The pulse duration is 750 as, the beam radius is 1.5 μm, and the peak intensity is $I_x = 5 \times 10^{13}$ W/cm^2.

Figure 4 presents the second major result of our paper. Pulse distortions due to spatiotemporal effects or due to the rapid variation of the harmonic phase are considerably smaller than anticipated, resulting in a high quality as-pulse. This can be attributed to the fact that the as-harmonic spectrum is close to the cutoff, where only a small range of laser intensities contributes to HHG. As a result, spatial and temporal distortions arising from the strong dependence of HHG on the laser intensity are

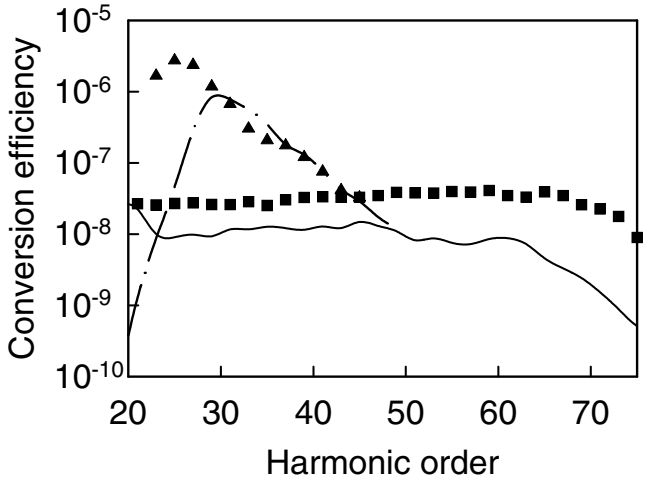

FIG. 2. Comparison of experimentally measured conversion efficiencies in Ar and Ne [9] with numerical results. The parameters are given in the text. The full line (numerics) and squares (experiment) represent Ne data, the dash-dotted line (numerics) and triangles (experiment) refer to Ar.

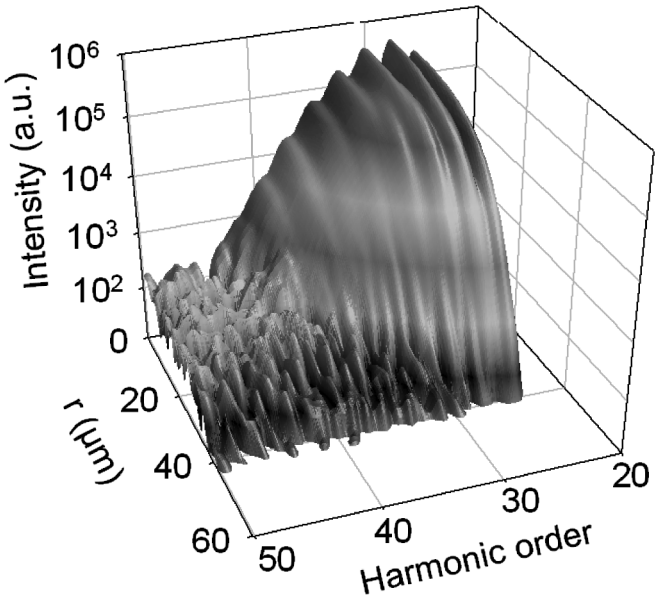

FIG. 3. Harmonic spectrum generated in Ar gas; for parameters see the text. The radial coordinate r is perpendicular to the propagation direction.

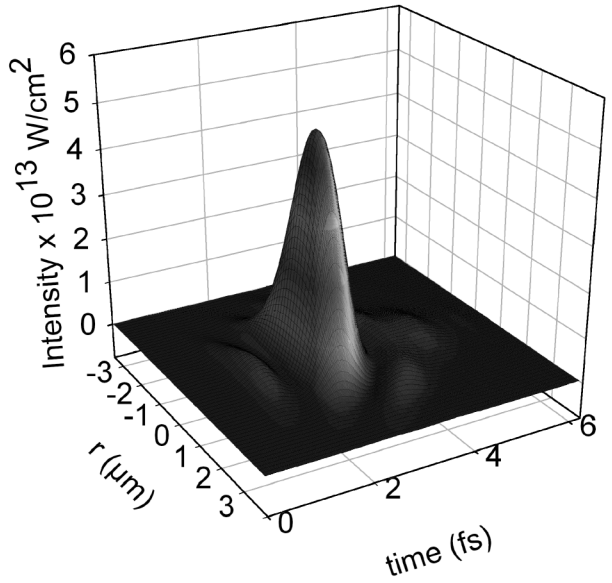

FIG. 4. Fourier transform of the spectrum between $N = 27$ and $N = 30$ in Fig. 3 after focusing. Ideal focusing by a quadratic mirror with focal length $f = 18$ mm has been assumed.

minimized. Calculations for higher laser peak intensities show that away from the cutoff more than one as-pulse is generated and that the temporal and the spatial quality of the as-pulse decreases with decreasing harmonic order.

As a result of the high pulse quality unexpectedly high xuv peak intensities are obtained. Our calculations are based on idealized assumptions for filtering and focusing, neglecting aberration, reflection losses and other effects, thus, giving an upper value for the achievable peak intensity. Note, however, that the pulse energy chosen in our calculation is 1 order below the maximum pulse energy realizable in few-cycle Ti:S laser systems [9]. Hence, by putting a factor of 10 more energy into the laser pulse and by increasing the pulse area such that the peak intensity remains unchanged, our calculation predicts a maximum peak intensity of 5×10^{14} W/cm^2. Therefore, even in the presence of losses, realization of intensities between 10^{13} and 10^{14} W/cm^2 in the 20-30 nm range is realistic. According to theoretical predictions [23], such intensities are sufficient to realize first xuv nonlinear optics experiments in the wavelength range above 20 nm.

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