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Conversion efficiency, scaling and global optimization of high harmonic generation

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Abstract: Closed form expressions for the high harmonic generation (HHG) conversion efficiency in the plateau and cut-off region are derived showing agreement with previous observations. Application of these results to optimal HHG-based-XUV-sources is discussed. © 2009 Optical Society of America

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High harmonic generation (HHG) is a promising source for coherent short wavelength radiation in the extreme ultraviolet and soft X-ray (XUV) region with ample application in different areas, such as spectroscopy, metrology, physics, chemistry, material science, and biology. Among the possible applications we can emphasize high resolution imaging, EUV lithography, seeding of X-ray free electron lasers and the possibility of probing the electron dynamics on its natural time scale via attosecond pulses. Much of current research is still focused on determining the optimum drive lasers and media to enable efficient harmonic generation at a given wavelength, especially pushing to the soft-x-ray (keV) region.

Over more than one decade, a considerable effort has been devoted to the theoretical modeling of HHG [1] However due to the computational complexity of the problem, which includes not only the microscopic response of the medium but also macroscopic propagation effects, accurate quantitative simulation of the HHG efficiency can be extremely time-consuming and, considering the numbers of variables involved in the HHG process, a systematic study with several quantities varying simultaneously is prohibitive. Indeed, up to the present date it is not entirely clear how the HHG efficiency scales for different experimental conditions, how far we are from theoretical limits, and how to proceed to construct an HHG source, which operates at the global maximum in efficiency for a particular harmonic or range of harmonics. For example, over the last years, the role of the driving frequency, ω_0 , to HHG scaling has received great attention [2,3]. Dependences of HHG efficiency with ω_0^5 and ω_0^6 are being obtained from numerical simulations using the time dependent Schrödinger equation [2] based on the single-atom response only. Preliminary experimental results [3] are supporting these numerical simulations.

 In this work we present the first, to our knowledge, closed form analytical expressions to the HHG conversion efficiencies both for the plateau region and the cutoff region including both laser and material parameters. The formulas were obtained applying the saddle point method to the dipole acceleration of the improved three step model (ITSM) [4]. Single-active-electron (SAE) approximation and 1D propagation effects were also considered.

The final expression for the efficiency at the cutoff frequency, Ω_{cutoff} , is written as:

$$
\eta = 0.0236 \frac{\sqrt{2I_p} \omega_0^5 |a_{rec}|^2 |g(\Delta k, L)|^2}{E_0^{16/3} \Omega_{cutoff}^2 \sigma^2 (\Omega_{cutoff})} \frac{1 - \beta^{4(N-1)}}{(1 - \beta^4)N} |1 + \beta|^2 \kappa_0 w [E(tb_{cutoff})], \quad (1)
$$

where $g(\Delta k, L) = [e^{i(\Delta k \cdot L)} - e^{-L/(2 \cdot L_{abs})}] / [1 + 2i(\Delta k \cdot L_{abs})]$, Δk is the phase mismatch and L_{abs} is the absorption length. *N* is the number of cycles of the driven pulse and $\beta = |a(\pi/\omega)|^2$ with $|a(t)|^2$ denoting the probability to find the atom on the ground state. I_p is the ionization potential, $w(E)$ is the ionization rate and a_{rec} is the recombination amplitude, as written in Eq. (7) of Ref. [5]. The intra-cycle depletion of the ground state, κ_0 , is given by $|a(tb_{cutoff})|^{2}$, where the respective birth and arrival times are *tb* $_{cutoff} \approx 1.88/\omega_0$ and *ta* $_{cutoff} \approx 5.97/\omega_0$.

In accordance with Eq.(1), the efficiency at the cutoff region scales with a factor of ω_0^5 . A cubic dependence with ω_0 is due to quantum diffusion. An additional factor of ω_0 comes from the fact that we are considering the conversion efficiency into a single harmonic, and the bandwidth it occupies is $2\omega_0$. The fifth ω_0 comes from the energy carried by a cycle of the driving laser field which scales with its duration $2\pi/\omega_0$ for a given electric field amplitude. On the other hand, the plateau formula, too length to be shown here, has an additional term related to the derivative of the action, $\partial_t^2 S$, which is related to the chirp of the attosecond pulses by the short and long trajectories. This leads to additional energy spreading over the harmonics reduction the power in each by another factor of ω_0 , Therefore, in general, the scaling of HHG efficiency with the driving frequency is ω_0^5 at the cutoff and ω_0^6 in the plateau region for fixed harmonic wavelength in agreement with previous numerical simulations [2]. However, for

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Efficiency

 $1.0E-7$

 $5.0E-7$ $1.0E-6$

 $2.0E-6$ $3.5E-6$

 $5.0E-6$ $7.0E-6$

 $1.0E-5$

the complete wavelength scaling others contributions have to be taken into account. For example, keeping constant the geometrical conditions and the number of cycles, any change on the driving wavelength implies a change in the field strength, E_0 , in order to keep perfect phase matching conditions. The medium characteristics, recombination amplitude and absorption cross section, also play an important role if cutoff extension is the target. To illustrate the significance of that statement, absorption limited HHG from neon is considered and the optimum drive wavelength for maximum conversion efficiency in the cutoff region is determined using Eq. (1).The result is displayed in Fig. 1(a) as a function of drive wavelength and cutoff energy. A global maximum for Ne efficiency is clearly observed for $\lambda_0 = 1.2 \mu m$ corresponding to $\Omega_{cutoff} = 451 eV$. Although, the maximum conversion efficiency shifts for different λ_0 , the peak efficiency does not exhibit any strong dependence with the driver wavelength, in contradiction to the λ_0 ^{-5..-6} scaling. . The reason for this unexpected behavior is that in the range from 30 to 800 eV for Ne, the recombination amplitude, *arec* increases and the absorption cross section, σ decreases. In particular, the absorption cross section decreases more than two orders of magnitude for that range.

(a)

 Ω_{cutoff} (eV)

Fig.1. Neon HHG efficiency at the cutoff region, using Eq. (1), as a function of the driven wavelength, λ_0 , and the cutoff energy, Ω_{cutoff} , $|g(\Delta k, L)|^2 = 1$ and a 5-cycle-driverpulse were assumed.

In Fig. 2, the same problem is considered but more constrains are imposed. In Fig. 2(a) the efficiency at cutoff for a 5 mm long gas cell at 1 bar of pressure is shown assuming perfect phase matching. In this case, the maximum value for HHG efficiency is reached for $\lambda_0 = 0.6 \mu m$ and $\Omega_{\text{cutoff}} = 148 \text{eV}$. In Fig. 2(b), the plasma and neutral atom phase mismatching are included to the problem. In this final case, the maximum value was reached at $\lambda_0 = 0.8 \mu m$ and $\Omega_{cutoff} = 107 eV$. This behavior is also reproduced using a Gaussian pulse and computing numerically the full spectrum via the ITSM.

Fig. 2. Neon HHG efficiency at the cutoff region as a function of the driven wavelength, λ_0 , and the cutoff energy, Ω_{cutoff} . (a) Considered a 5-cycle-driver-pulse, $L = 5$ mm and pressure 1 bar. (b) Same as (a) but also considering the plasma and neutral atom phase mismatching.

 In conclusion, we report the first derivation of closed analytical expressions for the high harmonic generation (HHG) conversion efficiency, both for the plateau region and the cutoff region. Specifically they show the origin of the power scaling with drive wavelength analytically. Moreover, the presented formulas include both laser and material parameters and eliminate most of the computational complexity related to HHG simulations. Based on the derived expressions, the computed HHG conversion efficiencies, without any parameter fitting, are in good agreement with experimental results obtained from different groups under different experimental conditions [6]. These formulas therefore simplify the HHG optimization problem considerably and enable a detailed scaling analysis of the HHG efficiency.

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