Ann. Phys. (Berlin) **525**, No. 3, A40–A42 (2013) / DOI 10.1002/andp.201300721

annalen der physik

EXPERT OPINION

High-harmonic generation with plasmonics: feasible or unphysical?

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High-harmonic generation (HHG) provides for efficient and coherent XUV generation for a wide range of spectroscopic applications, enabling attosecond spectroscopy, ultrafast photo-emission spectroscopy, or ultrahigh spatial resolution imaging [1]. Conventionally HHG is based on the use of high power amplified femtosecond laser pulses, special phase matching techniques, or intra-cavity field enhancement. As a compelling alternative, resonant plasmonic field enhancement in metallic nanostructures has been proposed to achieve the necessary peak intensity for HHG. Using nJ pulses from a Ti:S oscillator, XUV generation attributed to plasmon-enhanced HHG was reported in 2008 by the group of Seung-Woo Kim from KAIST in Daejeon, Korea, first in Ar [2] and later in Xe [3] using bowtie plasmonic nanostructures, followed in 2011 using plasmonic waveguides [4]. However, many groups failed to observe any appreciable effect in their efforts to reproduce the experiments. Instead, the recent observation of XUV emission via multi-photon fluorescence under very similar experimental conditions [5] called into question the HHG assignment in the original work.

In an effort to clarify the controversy the S.-W. Kim group has provided several new results and experimental details [6] based on which they uphold their original interpretation of the observed signal as HHG. Here, I will provide a critical interpretation of that work in context with the results published by the authors previously. While the new data on the interference of the XUV emission from the funnel waveguides are suggestive of HHG, I will argue that the new spectra obtained from the bowtie structures do not seem to resemble what is expected for HHG emission, and in fact contradict the high quality XUV comb spectra reported in their earlier work. To put the results and reported XUV power into perspective, I will also provide an estimate of the expected plasmon-enhanced HHG intensity from extrapolation from cavity-enhanced HHG experiments. Based on this comparison, due to the field-enhancement for the only nanoscopic volume a significant plasmonic HHG yield seems unlikely.

Figure 1 shows the data reported for Ar [2] and Xe [3] where for

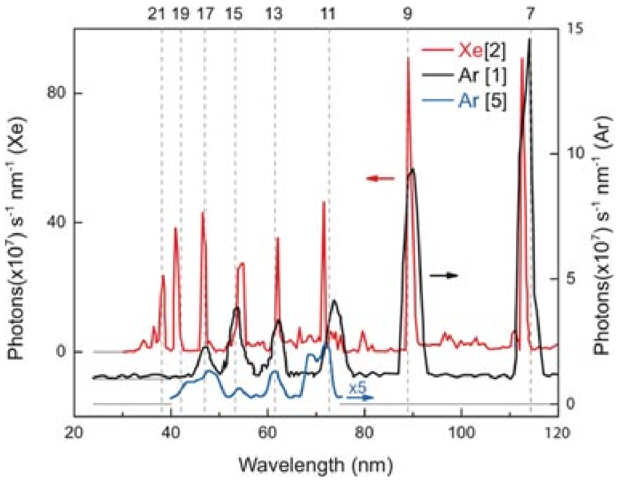

Figure 1 Plasmon-enhanced high-harmonic spectra reported for Ar [2] and Xe [3] with generated intensities of ∼ 0.1..0.2 nW and 2..3 nW for each of the main harmonics, respectively (data digitized from [2] and [3]) in comparison with the new data from [6] (x5, and offset for clarity). Dashed lines indicate the expected spectral position for the different harmonics for the 800 nm pump wavelength.

Table 1 Plasmon-enhanced experimental parameters and reported output P_{exp} from [2–4], in comparison to the expected HHG power P_{theo} extrapolated from cavity-enhanced HHG for similar intra-cavity field-enhancement [9].

	Bowtie	Funnel	Cavity
Length	$h = 50$ nm	$h = 450$ nm	$l_c = 50..150\ \mu$m
Area	1.5×10^3 nm^2	1.4×10^4 nm^2	$A_c = 960\ \mu$m^2
N	150..600	1	1
P_{exp} (Xe)	$\sim 2..3$ nW [1,2]	$\sim 20..30$ nW [3]	$\sim 20..60\ \mu$W [9]
P_{theo} (Xe)	$10^{-6}..10^{-5}$ nW	$10^{-5}..10^{-4}$ nW	

incident intensities of only 10^{11} W/cm^2 onto an array of gold bowtie nanostructures high quality HHG comb spectra were obtained with excellent contrast, plateau, and cut-off at the 17th and 21st harmonics, respectively. The observation has been attributed to the plasmonic field enhancement increasing the local field intensity in the bowtie gap above the established threshold of 10^{13} W/cm^2 for the onset of HHG. The appreciable XUV power reported and the fact that the experiments were performed with many parameters not even optimized has stimulated the imaginations of the wider scientific community. In 2011, using the new design of a hollow tapered metal waveguide, the expanded team reported XUV emission up to 70 eV with 43rd harmonics, albeit with less distinct comb lines [4]. HHG photon yields were found to be 1-2 orders of magnitude higher than in the bowtie experiments.

However, in all experiments very limited details were provided, and few experimental parameters were varied systematically. One critical overarching question the authors did not discuss in any of their work is the mechanism and general feasibility of HHG under the given experimental conditions. Concerns regarding the only nanoscopic field-enhanced sample volume compared to conventional far-field experiments were already raised by Sivis *et al.* [5]. A straight forward estimate of the expected XUV photon yield is possible extrapolating from cavity-enhanced HHG using frequency combs, as these experiments are also based on ~100 MHz repetition rate laser oscillators with low pulse energies [7,8]. In that case, cavity enhancements by a factor of a few hundred provide intra-cavity intensities in the range of typically 2–5 × 10^{13} W/cm^2 [8] and up to 9 × 10^{13} W/cm^2 [9], i.e., comparable or just above what is assumed for plasmonic field-enhanced intensities. Table 1 shows the expected (details of calculations can be found as online supplement) HHG yield P_{theo} in a cavity-enhanced experiment if the gas volume were reduced to the aggregate volume of plasmonic field-enhanced regions assuming a similar target gas density.

How can these exceedingly low expected HHG yields be reconciled with the orders of magnitude higher reported experimental power levels P_{exp} for bowties and funnel waveguide? The damage threshold would preclude the field intensity in the bowtie gap or funnel being significantly higher. The new disclosure [6] that the photon yield stated as measured in [2,4] was in actuality corrected for the efficiency of the detection system is already taken into account in the estimate. Including the revision for the number of bowties illuminated (600 instead of 150) [6], would only increase the field-enhanced area from 1.5×10^3 to 6×10^3 nm^2. A significantly higher gas density in the plasmonic HHG experiments ($\sim 10^2 - 10^3$ times would be needed) seems unlikely given the similar nozzle sizes and backing pressures used. The plasma could recombine faster in the presence of the metallic nanostructures, and the shorter pulse duration for the same peak power does increase HHG efficiency, but not by orders of magnitude. Even the remote possibility of a coherent enhancement via a phased array antenna effect [10], with emission power scaling $\propto N^2$ instead of $\propto N$ could not explain the results. It would add a factor of 100 in power, not only still far off the signal level shown in Fig. 1, but also no such effects have been observed in corresponding linear or low-order nonlinear studies of plasmonic arrays.

The experiments of Sivis *et al.* performed under similar conditions showed that multi-photon or high-field fluorescence can dominate the spectra in the 120 to 30 nm spectral range (corresponding to the expected H7 to H23 range) [5]. These processes are in fact interesting in themselves and their plasmon-enhanced generation deserves further investigation. They are favorable in nano-scopic field enhanced volumes due to only linear interaction length scaling, their resonant enhancement, and they are effective below the ionization threshold. This is in contrast to HHG which can be seen as the coherent non-resonant analogue of the strong interaction of a light field with atoms favored at power levels above the ionization threshold [13].

However, in my opinion multi-photon fluorescence cannot explain the origin of the nearly perfect comb spectra observed in [2, 3] and shown in Fig. 1. While multi-photon fluorescence exhibits peaks from different resonances, these peaks are not equally spaced and are unrelated to the 800 nm pump wavelength [5, 8]. Such multi-photon fluorescence in fact has been observed in early cavity-enhanced comb spectra [8], but due to its distinct non-periodic multispectral signature, it cannot be confused with HHG even with a miscalibrated spectrometer. In comparison with the reported spectra for Ar and Xe from [2, 3], the new data presented for Ar [6], now with improved bowtie illumination and XUV detection (blue, Fig. 1), and after background subtraction, no longer resemble in spectral characteristics and power the earlier results (black). Unfortunately the authors do not comment much on that fact in [6].

Having said that, the new results on XUV emission from the funnel waveguides are interesting with some signature of HHG conceivable, superimposed on fluorescence emission. The diffraction experiment shown is going in the right direction of resolving the question of the physical origin of the observed XUV emission. However, phase noise and pulse fluctuations would destroy the interference, making the interpretation of the observed interference features as evidence for HHG emission difficult. Note also that the multi-photon fluorescence via its long-lived resonances and possibly involving Stark-shifted Rydberg levels as was noted in the context of cavity-enhanced experiments [8], allows for directional and partial phase coherent emission.

Plasmon enhanced HHG is not fundamentally impossible. Also, as suggested theoretically [11, 12] the near-field gradients can influence the ponderomotive motion and allow for new selection rules that can possibly enhance the HHG process. However, as with many plasmonic effects, nano-scale local field-enhancement does not necessarily translate into high macroscopic ensemble efficiency. In summary, given the difficulties with the claim of plasmon-enhanced HHG by Kim *et al.,* in the light of the limited data, the conflicting spectra, the in part inconsistent presentation, and apparent problems with reproducibility, we shall all be reminded of the statement credited to the famous skeptic Marcello Truzzi, and formulated by Carl Sagan as “Extraordinary claims require extraordinary evidence”. I believe that this extraordinary evidence has yet to be found.

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Supporting information for this article is available under http://dx.doi.org/10.1002/andp.201300721.

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