Supporting Information: High-harmonic generation with plasmonics: feasible or unphysical?

Markus. B. Raschke

HHG power estimate. The estimates of the expected plasmon-enhanced HHG power shown in Table 1 are based on the following considerations. Intracavity field intensities range from $2-5$ [1] up to 9×10^{13} W/cm^2 [2], i.e., comparable to what is assumed for plasmonic field-enhanced intensities. Under most recent optimized cavity conditions, e.g., for 4×10^{13} W/cm^2, just before the saturation regime, HHG intensities of $\sim 20-60$ μW (generated) from 9th to 19th harmonics for Xe, and about an order of magnitude less for Kr were obtained [2]. The interaction volume is given by the laser focus with an area of $A_c = 960$ μm^2, and extends phase matched over the coherence length of $l_c \sim 50-150$ μm. In both cases the gas density in the laser interaction volume is not well known, but can be assumed to be comparable with the noble gas exposure performed in a similar fashion in both the plasmonic and cavity-enhanced experiments.

We can then ask how much HHG one would obtain in a cavity-enhanced experiment if the gas volume were reduced to the aggregate volume of plasmonic field enhanced regions as shown schematically in Fig. S1. For an enhanced intensity of 4×10^{13} W/cm^2 the expected HHG power $P_{\mathrm{theo,pl}}$ generated by the ensemble of N plasmonic nanostructures, of height h and field-enhanced area A_{pl} can be extrapolated from the measured conventional cavity-enhanced HHG power output $\mathrm{P}_{\mathrm{exp,cavity}}$ as

$$P_{\mathrm{theo,pl}} = P_{\mathrm{exp,cavity}} \left(\frac{h_{\mathrm{pl}}}{l_c}\right)^2 \frac{N A_{\mathrm{pl}}}{A_c} \quad (1)$$

with quadratic dependence in interaction length due to phase matching for the relevant length scales. Judging from the SEM images in [3,4], $h = 50$ nm. The field enhanced area seems to be 50 nm by 30 nm in extent, i.e. 1.5×10^3 nm^2. From the focus size and bow-tie surface density N is ~ 150 [3] and ~ 600 in the new experiment [5]. This results in maximal $P_{\mathrm{theo,pl}} = (2.3 \times 10^{-10}$ to $2.1 \times 10^{-9}) P_{\mathrm{exp,cavity}}$ with the range defined by the most optimistic or most pessimistic choice of values from the range of values above. This translates into power levels of $P_{\mathrm{theo,pl}} \sim 10^{-5}$ nW at the very best for Xe, and another order of magnitude weaker for Kr, and yet another order of magnitude less for Ar. A similar analysis for the funnel waveguide geometry with a larger volume with $h = 450$ nm and 1.4×10^4 nm^2 cross sectional area, yet being a single structure, yields at most $P_{\mathrm{theo,pl}} \sim 10^{-4}$ nW for Xe.

Photoinduced damage. With the lack of success of reproducing the results by other groups over the years, photoinduced damage of the plasmonic devices has been discussed as a bottleneck. However, in particular for few fs transient excitations, metal nanostructures can withstand peak intensities in the $\sim 10^{13}$ W/cm^2 range if coupled to a bulk or substrate with high thermal conductivity. E.g., for the case of second-harmonic generation (SHG) from the apex of a sharp gold metal tip, which can be viewed as a half bow-tie, the local field enhancement of ~ 10 for $\lesssim 10$ nm apex radii translates incident peak intensities of $0.5-1.2 \times 10^{11}$ W/cm^2 of 10 fs pulses into apex localized intensities of $(0.5-1.2) \times 10^{13}$ W/cm^2, without at least short term degradation [6]. However, with ionization potential of Au of 9.2 eV and thus lower than even Xe, bond breaking and Au ion desorption leading to sample degradation is expected to proceed rapidly at intensities in excess of a few times the above value.

Acknowledgement. I would like to acknowledge in particular Rudi Bratschitsch, Ralph Ernstorfer, Peter Hommelhoff, Henry Kapteyn, Mathias Kling, Margaret Murnane, Claus Ropers, Thomas Schibli, Christian Spielmann, Wolf Widdra, and Jun Ye for many stimulating discussions on this subject. I thank Ben Pollard and Vasily Kravtsov for help with the analysis and the figures.

Markus. B. Raschke
Department of Physics, Department of Chemistry, and JILA
University of Colorado, Boulder, CO 80309, USA
E-mail: markus.raschke@colorado.edu

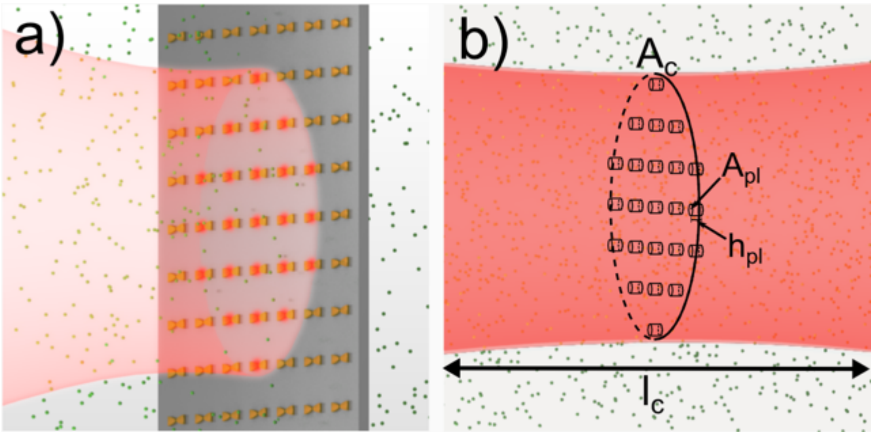

Figure 1 Schematic of the general plasmonic sample geometry proposed for high harmonic generation (a) compared to cavity enhanced HHG (b). Assuming the same cavity enhanced intensity and weighted averaged peak pulse intensity in the bow-tie gap regions of $\sim 4 \times 10^{13}$ W/cm^2, and assuming similar noble gas densities one can relate the two experiments directly to each other. From scaling the cavity enhanced HHG power in the micron sized laser focus by the bow-tie enhanced nanoscopic interaction volume the plasmonic HHG yield is expected to be exceedingly weak (see text for details). This is in contrast to resonant multi-photon fluorescence, which as a incoherent process scales linearly as opposed to quadraticly with interaction length.

References

[1] C. Gohle, T. Udem, M. Herrmann, J. Rauschenberger, R. Holzwarth, H. A. Schuessler, F. Krausz, and T. W. Hansch, Nature **94**, 193201 (2005)

[2] A. Cingöz, D. Yost, T. K. Allison, A. Ruehl, M. E. Fernmann, I. Hartl, and J. Ye, Nature **482**, 68 (2012)

[3] S. Kim, J. Jin, Y. J. Kim, I.-Y. Park, Y. Kim, and S. W. Kim, Nature **453**, 757 (2008)

[4] S. Kim, I.-Y. Park, J. Choi, and S.-W. Kim, in "Progress in Ultrafast Intense Laser Science VI," (ed. K. Yamanouchi, Springer-Verlag, Berlin Heidelberg, 2010), High harmonic generation by plasmonic enhancement of femtosecond pulse laser, pp. 129–144

[5] I.-Y. Park, J. Choi, D.-H. Lee, S. Han, S. Kim, and S.-W. Kim, Ann. Phys. (Berlin) **525**, 205–214 (2013)

[6] C. C. Neacsu, G. A. Reider, and M. B. Raschke, Phys. Rev. B **71**, 201402 (May 2005)