Attosecond photoscopy:
How to reveal subcycle dynamics at plasmonic surfaces and nanostructures

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Abstract

State-of-the-art experiments with attosecond time resolution are on the way to resolve sub-cycle electron dynamics from plasmonic interaction of ultrashort driving pulses in surfaces and nanostructures. The approach of attosecond photoscopy utilizes well established techniques from attosecond science and demonstrates a reliable route to extend attosecond technology to an even broader range of surface and nanostructure dynamics. As a proof of concept experiment we demonstrate attostreaking from thin gold films with significant deviation to the gas phase streaking measurements. Furthermore we developed new, non-destructive preparation procedures for Gold nanoparticle (AuNP) samples as basic principle to perform attosecond photoscopy measurements.

In recent years the development of miniaturized optical and opto-electronic circuits became one of the most interesting and challenging topics in industry and science. Integrated opto-electronic circuits allow speeds at Petahertz scale opening the area of research of ultrafast charge transfer dynamics on nanometer scale. Optical phenomena like surface or localized plasmon polaritons (SPP/ LPP) described as light induced collective electronic motions can develop at ultrafast timescales [1]. Resolving and understanding the underlying dynamics is one of the main challenges to taylor new integrated devices with an unprecedented speed.

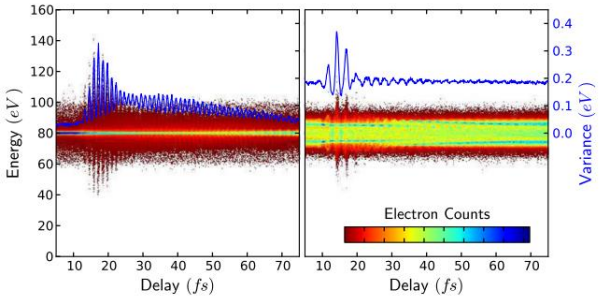

Fig. 1 Photoscopic spectrograms at perpendicular (left) and grazing (right) electron emission. The measurements retrieve plasmonic and NIR fields, respectively. Solid lines are the momentum variances.

Investigating the dynamics of plasmonic excitations in AuNPs and nanostructures we follow the general idea of a newly proposed technique we call attosecond photoscopy [2].

Minor modifications to the attosecond streaking technique may help to overcome the challenging utilization of imaging techniques like PEEM to uncover the details of plasmon propagation and localization with attosecond resolution. In this setup a SPP is launched perpendicular to the incoming light field. As the main difference to the attosecond streaking technique, attosecond photoscopy is a new pump-pump-probe scheme using a few-cycle IR pulse to launch a plasmon (1); the plasmonic field affecting the photoemission (2) probed by an attosecond XUV pulse over time (3) mapping the evolution of the plasmon in time and space (Fig. 1). The plasmonic field can be retrieved from the photoscopic spectrogram recovering the built-up time, lifetime, and damping properties.

As a first proof of principal an attosecond streaking experiment [3] was demonstrated from a polycrystalline gold film hosting some arbitrarily distributed nanoscale discs (~25nm dia, ~10nm height) on a flat surface (0,7nm rms). The analysis of the emitted electron wave packet shows a temporal broadening of 72as in comparison to gas phase streaking measurements at moderate IR intensities (6 GW/cm²).

A preparation strategy for a monolayer of nearly close-packed nanospheres was developed to prepare chemically clean (i.e. no surface contamination, or ligand) samples preserving shape and size. The characterisation procedure with electron microscopy and XPS shows the distribution and size of nanoparticles, and agglomerates as well as the contamination free surface (see Fig. 2).

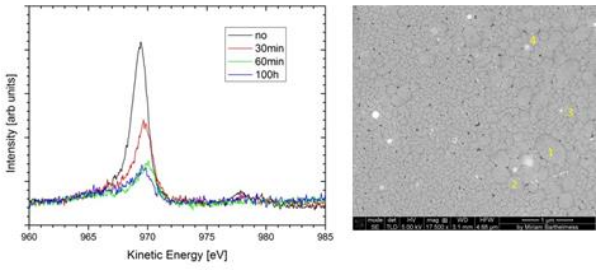

Fig. 2 XPS (left) and SEM (right) of an Au sphere sample showing the surface contamination in the C1s peak and the agglomeration

As a final step attosecond photoscopy measurements will be performed on this new type of samples to verify the theoretical predictions.

References

[1] M. L. Brongersma & V. M. T. Shalaev, *Science* 328, 440 (2010).

[2] M. Lupetti, et al., PRL 113, 113903 (2014).

[3] W. A. Okell et al., Optica 2, 383 (2015).