FINAL REPORT ON THE RESULTS OF PROJECT NKFI PD 125249

Ultrafast dynamics in nanooptics

There was significant progression in the research field of nanooptics and its applications in recent years [1-4]. Intense parallel development occurred in the investigation of ultrafast light-matter interaction processes on the nanoscale [5-6]. Along these lines, we and other research groups started to carry out the examination of ultrafast electron emission and electron acceleration in the nanolocalized and highly enhanced electromagnetic fields of propagating and localized surface plasmon polaritons (SPPs) or nanotips [7-10].

As a continuation of this research direction, in this project ultrafast photoemission near-field probing technique was developed [11] to investigate the fundamental question of plasmon–plasmon coupling and its effect on large field enhancement factors. By measuring and analyzing plasmon field enhancement values at different nanostructured surfaces based on the spectrally resolved electron emission measurements, it was possible to separate the contributions from propagating and localized plasmons (Figure 1). When resonance conditions are met, a significant field enhancement factor can be attributed to the generation of localized plasmons on surface nanostructures, acting as dipole sources resonantly driven by the propagating plasmon field. The coupling of propagating surface plasmon waves and localized plasmon oscillations in nanostructures is an essential phenomenon determining electromagnetic field enhancement on the nanoscale.

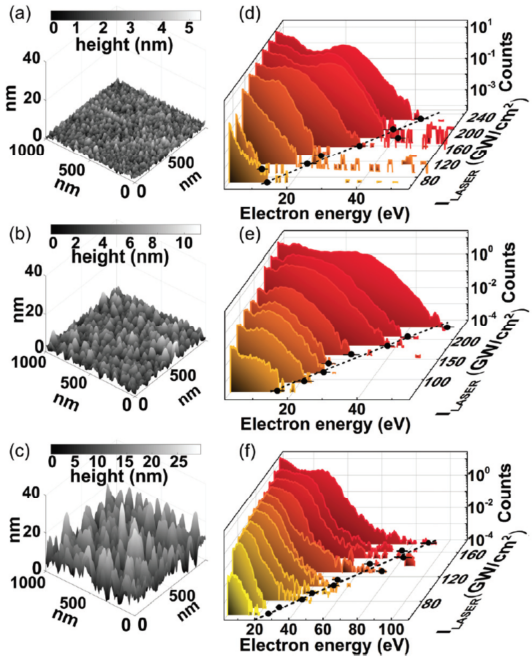

Figure 1. Surface morphology and the corresponding electron spectra. (a–c) Atomic force microscope scans of plasmonic surfaces with different rms roughness of 0.8 nm, 1.6 nm and 4.5 nm. (d–f) Plasmonic photoelectron spectra from the corresponding three surfaces by generating plasmons as a function of laser intemsity. The black symbols correspond to electron spectral cutoffs.

Furthermore measurements were performed by a time of flight spectrometer investigating the surface plasmon-induced photoemission on the ordered sample [12]. During the sample

preparation 100 nm dielectric spheres were deposited as an ordered structure by Langmuir-Blodget method on a rectangular prism and it was covered by a 45-nm gold film for plasmon coupling. Oscillations with a certain period could be identified based on the measurements in the time of flight spectra, and furthermore narrower peaks were detected with shorter time periodicity. (Figure 2) The possible interpretation of these peaks is the detection of high plasmon filed induced electron pairs, consistent with previous results [13-14]

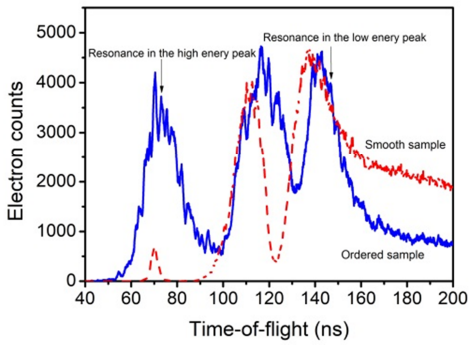

Figure 2. Typical time-of-flight spectra for smooth (dashed red line) and ordered (solid blue line) samples at 65GW/cm^{-2}.

In addition, time-resolved autocorrelation experiments based on nonlinear electron emission were performed from plasmonic nanoparticles. Because multi-photon-induced emission takes place from gold and silver at the utilized 800 nm wavelength therefore this highly nonlinear process can be applied for constructing a nonlinear interferometric autocolerrator probing the local field with the help of photoelectrons. The characteristic emission mechanism is the three or four photon induced emission because of the work functions of the gold and the silver are around 5 eV and the photon energy is around 1.5 eV at 800 nm wavelength.

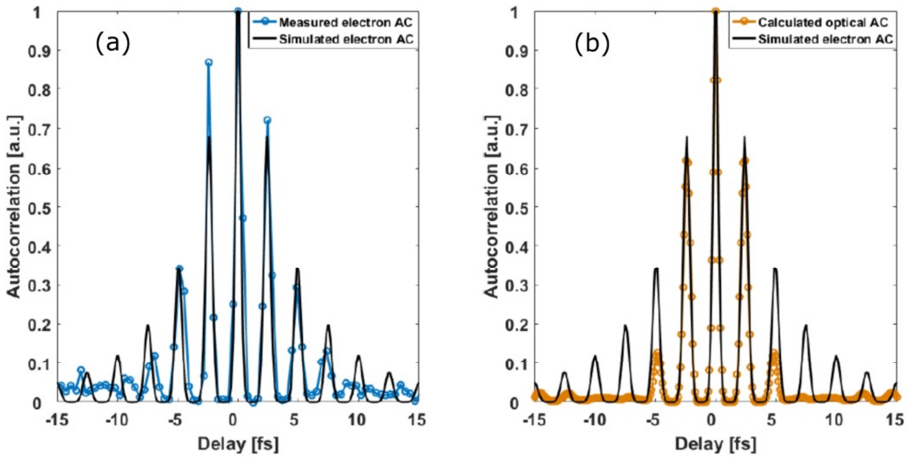

Figure 3. (a) Simulated and measured fourth order interferometric photoelectron autocorrelation traces. The signal is defined by the photoelectron current as a function of the delay. (b) Calculated optical autocorrelation of the exciting laser pulse and simulated electron autocorrelation showing that the plasmon autocorrelation broadens because of spectral filtering.

During this experiment the total plasmonic photocurrent can be measured as a function of the delay between the two arms of an ultrabroadband interferometer. Because the electron emission is determined by the plasmonic near field therefore the time evolution of the plasmon filed can be determined from this type of measurements. Based on the early measurements with ultrashort few optical cycle laser pulses (5,5 fs), short few cycle localized plasmon field can be generated. But the characteristic time evolution of plasmon near field is longer than pulse length of the generating optical filed because of spectral filtering caused by plasmon couplings (Figure 3)

Based on further measurement on rectangular nanoparticles, different plasmon pulse lengths could be demonstrated at different polarization excitations in accordance with different resonance frequencies depend on the characteristic lengths of the nanoparticles in different directions of the polarization. In addition, the autocorrelation measurements are also performed in energy resolution. The autocorrelations for a given electron energy range have become measurable (Figure 4). In this way, for example it is possible to study the time evolution of nanooptic field regions with the maximum field-enhancement, since the electrons with the maximum energy are emitted from these spatial regions, and consequently we may be able to make space and time resolved measurements. Further experiments are still ongoing and the first results have not yet been published in referred journal, but have already been presented at conferences [15-17].

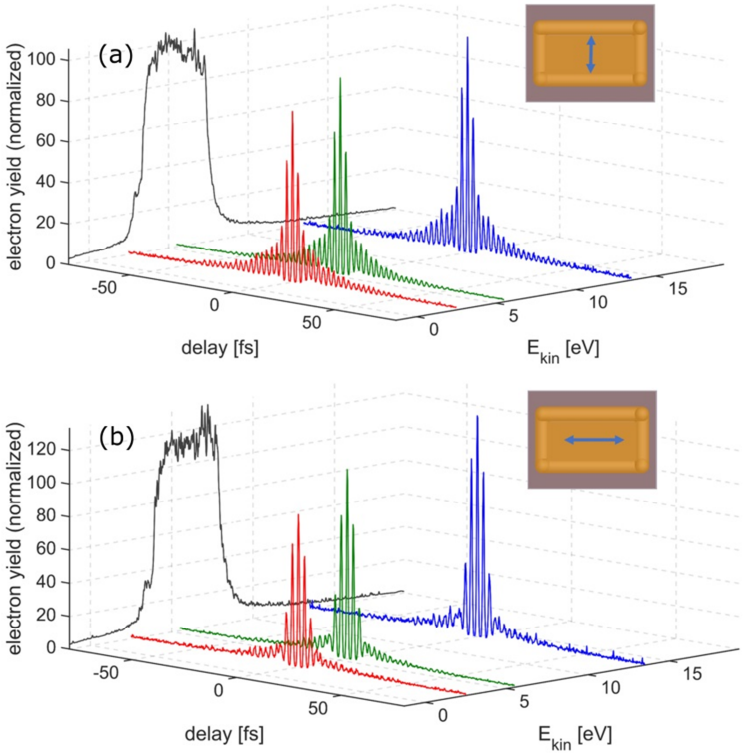

Figure 4. Electron autocorrelation traces evaluated at different electron energies using laser polarization (a) perpendicular to long edge of the nanorod and (b) parallel to long edge of the nanorod.

I was also involved in a research project connected to the field of attosecond science. High harmonic generation (HHG) was studied on noble gas targets with different cluster sizes [18]. (Clusters are groups of atoms that usually exist only for short periods of time and under special conditions, such as supersonic jets or flows). High harmonic spectra were measured for different backing pressures and gases (Ar, Xe) as a function of driver pulse ellipticity. Different cluster sizes can be produced by different backing pressures at different noble gases, and the cluster sizes can be measured by light scattering [19].
Since the ellipticity-dependent HHG decay is essentially the same for the different gas-pressure pairs and therefore for different cluster size, we can conclude that the recombination process is dominated by atom-to-itself recollisions irrespective of cluster size and material.
This conclusion was supported by classical electron trajectory simulations in elliptic electromagnetic fields. Based on this simulation the ratio of return events with respect to all events (that is, returns and events when the electron misses the cluster) was calculated. This quantity was called "return fraction" (Figure 5). Based on the FWHM of these return fraction curves provide a rough first estimate on the threshold ellipticity one can expect and since this type of increase of threshold ellipticity which could be originate from cluster to itself recombination process was not observed experimentally, we could rule out this mechanism and therefore the atom-to-itself recombination is dominated.

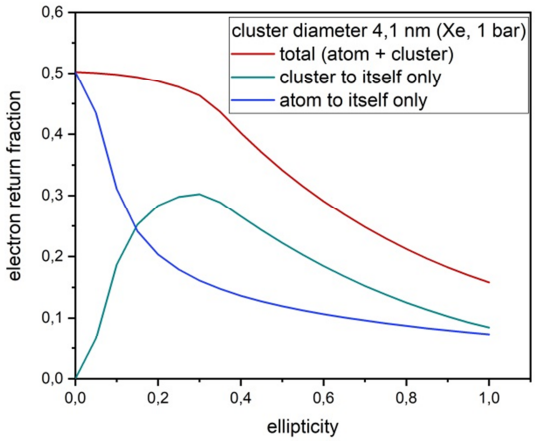

Figure 5. Calculated the ratio of return events (return fraction) as a function of ellipticity.. Here, only atom-to-itself and cluster-to-itself events are distinguished.

The realization of those simulations was my major contribution in this publication. For this work I was able to use my previous experience and my own model of plasmonic electron acceleration and electron trajectory computation [20]. Of course, I have adapted this model and simulation to the given problem [18].

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References:

[1] M. I. Stockman, M. F. Kling, U. Kleineberg, F. Krausz, "*Attosecond nanoplasmonic-field microscope,"*, Nat. Photonics **1**, 539 (2007).

[2] H. A. Atwater; A. Polman, *" Plasmonics for improved photovoltaic devices"*, Nat. Mater, **9**, 205 (2010)

[3] C. Haffner et al. "*Low-loss plasmon-assisted electro-optic modulator*", Nature, **556**, 483 (2018)

[4] A. R. Rastinehad, N. J. Halas et al."*Gold nanoshell-localized photothermal ablation of prostate tumors in a clinical pilot device study*", PNAS, **116**, 18590 (2019)

[5] Falk, A. L. Koppens, F. H. L. Yu, C. L.; Kang, K. de Leon Snapp, N. Akimov, A. V. Jo, M.-H. Lukin, M. D. Park, H. "*Near-field electrical detection of optical plasmons and single-plasmon sources*" Nat. Phys. **5**, 475 (2009).

[6] M. Ono, M. Notomi et al. "*Ultrafast and energy-efficient all-optical switching with graphene-loaded deep-subwavelength plasmonic waveguides*", Nat. Photonics **14**, 37 (2020)

[7] S. Thomas, M. Krüger, M. Förster, M. Schenk, and P. Hommelhoff, " *Probing of Optical Near-Fields by Electron Rescattering on the 1 nm Scale*", *Nano Lett.*, **13** , 4790 (2013)

[8] B. Piglosiewicz, C. Lienau, et al. "*Carrier-envelope phase effects on the strong-field photoemission of electrons from metallic nanostructures*", Nat. Photonics, **8**, 37 (2014)

[9] P. Dombi, A. Hörl, **P. Rácz**, I. Márton, A. Trügler, J. R. Krenn, U. Hohenester, "*Ultrafast strong-field photoemission from plasmonic nanoparticles*", Nano Lett., **13**, 674 (2013)

[10] **P. Rácz***, Z. Pápa*, I. Márton, J. Budai, P. Wrobel, T. Stefaniuk, C. Prietl, J. R. Krenn and P. Dombi *"Measurement of nanoplasmonic field enhancement with ultrafast photoemission,"* Nano Lett, **17**, 1181 (2017)

[11] J. Budai, Z. Pápa, I. Márton, P. Wróbel, T. Stefaniuk, Z. Márton, **P. Rácz**, P. Dombi, "*Plasmon-plasmon coupling probed by ultrafast, strong-field photoemission with <7 Angström sensitivity*," Nanoscale **10**, 16261 (2018).

[12] **P. Rácz**, N. Kroó, *" Surface-Plasmon-Assisted Cooper Pair Formation in Ordered Gold Films at Room Temperature*" Phys. Wave Phenom., **27** 192 (2019)

[13] N. Kroó, **P. Rácz**, S. Varró, *"Surface plasmon assisted electron pair formation in strong electromagnetic field"*, *EPL,* **105** 67003 (2014)

[14] N. Kroó, **P. Rácz**, S. Varró, *" Surface-plasmon-assisted magnetic anomalies on room temperature gold films in high-intensity laser fields"*, *EPL,* **110** 67008 (2015)

[15] B. Lovász, P. Sándor, Zs. Pápa, B. Éles, B. Bánhegyi, **P. Rácz**, C. Prietl, J. R. Krenn, P. Dombi, "*Few-femtosecond plasmon transients probed with nm-scale sensitivity*" The 9th International Conference on Surface Plasmon Photonics (SPP9), Denmark, Copenhagen, 2019

[16] **P. Rácz**, Zs. Pápa, P. Sándor, B. Lovász, B. Bánhegyi, P. Dombi, ”*Nanooptikai közelterek meghatározása szubnanométeres érzékenységgel*” Magyar Fizikus Vándorgyűlés 2019, Sopron

[17] B. Lovász, P. Sándor, Zs. Pápa, B. Éles, B. Bánhegyi, P. Rácz, C. Prietl, J. R. Krenn, P. Dombi, ”*Few-femtosecond plasmon transients with nm-scale sensitivity*” CLEO Europe 2019 Germany, Munich, 2019

[18] B. Bódi, M. Aladi és **P. Rácz**, I. B. Földes és P. Dombi, ”*High harmonic generation on noble gas clusters*” Opt. Express, **27** 26721 (2019)

[19] M. Aladi, R. Bolla, D.E. Cardenas, L. Veiszb; I.B. Földes ” *Cluster size distributions in gas jets for different nozzle geometries* ” JINST, **12** C06020 (2017)

[20] **P. Rácz**, V. Ayadi, P. Dombi, "*On the role of rescattering and mirror charge in in ultrafast nanooptical field probing with electrons*," J. Opt. **20**, 015501 (2018)