FINAL REPORT ON THE RESULTS OF PROJECT NKFI PD 109472

Investigation of ultrafast processes in nanolocalized electromagnetic fields

1. Experiments on electron emission and acceleration from nanoparticles

There was a significant development in the research of electromagnetic fields localized on nanometer scale in recent years [1-3]. Intense parallel development occurred in the investigation of ultrafast light-matter interaction processes on the nanoscale [4-6]. 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 [7,8].

As a continuation of this research direction, in this project nonlinear photoemission was investigated in different configurations [9-11]. *First, the plasmonic photoemission was induced from plasmonic films with femtosecond, mid-infrared pulses at 3.1 mm wavelength* [9]. With this type of mid-IR light sources it is possible to exploit favourable scalings of the Keldysh gamma parameter and the ponderomotive energy, because these quantities scale inversely proportional and quadratically with wavelength, respectively.

During this experiment, carried out with a setup that I built in Budapest, the photocurrent and photoelectron spectra were measured by the combination of a retarding grid analyser and an electron multiplier. Based on incident laser intensity dependence of the total photocurrent, the transition between multi-photon induced and tunneling emission was demonstrated at an unprecedentedly low intensity < 1 GW/cm^2 (Figure 1.)

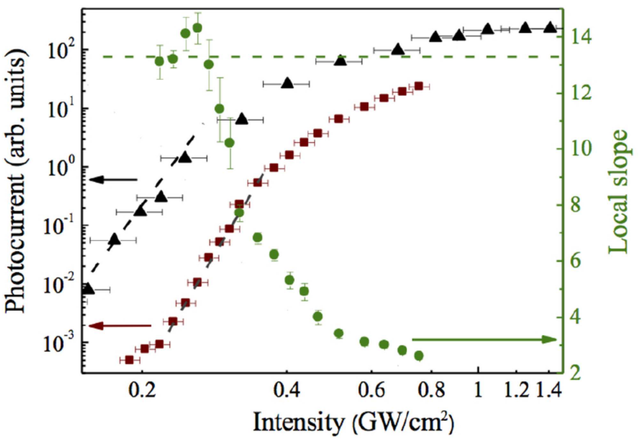

Figure 1. Laser intensity dependence of the total plasmonic photocurrent for two independent scans. The slopes of the fits to the initial sections (dashed lines) are 12.3±1.8 and 13.1±0.6, respectively. The local slope of the second curve is also plotted (green circles) to illustrate the gradual transition between multi-photon-induced and tunneling photoemission at very low laser intensities.

Spectrally resolved measurements of the strong-field photoelectron bunch were also performed with a retarding field analyser. According to these measurements, it was possible to generate electron bunches with 50 eV maximum kinetic energy during the nanoscale acceleration process in the field of surface plasmons even at two orders of magnitude lower incident laser intensity than in the previous measurements at 800 nm wavelength. Thereby, strong-field nanophysics can be accessed at extremely low intensities by exploiting nanoscale plasmonic field confinement, enhancement and ponderomotive wavelength scaling at the same time. Results agree well with quantum mechanical modelling. This scheme demonstrates an alternative paradigm and regime in strong-field physics.

In a second work [10] *the influence of surface roughness was investigated therefore the ultrafast plasmonic electron emission was demonstrated from Ag nanolayers having three different roughness with near infrared femtosecond pulses at 800 nm wavelength.* The energy distributions of the electrons were obtained by a time of flight spectrometer and it was found that the surface roughness deeply influences the properties of the electron spectra. Based on these spectrally resolved measurements, it can be concluded that the highest electron energies are measured from the sample having the highest RMS roughness. Also numerical investigations were performed to better understand the observed phenomena. During the numerical simualtion the surface roughness can be considered as nanoparticles on a metal surface, and the surface roughness was approximated by nanoellipsoids. These numerical calculations support the observations because in the case of highest roughness propagating surface plasmons coupled to localized plasmons on surface grains more resonantly.

Finally and most importantly, I experimentally demonstrated and validated a novel measurement technique of the maximal plasmonic field enhancement with ultrafast photoemission and acceleration process in any metallic, nanostructured environment at a surface [11].

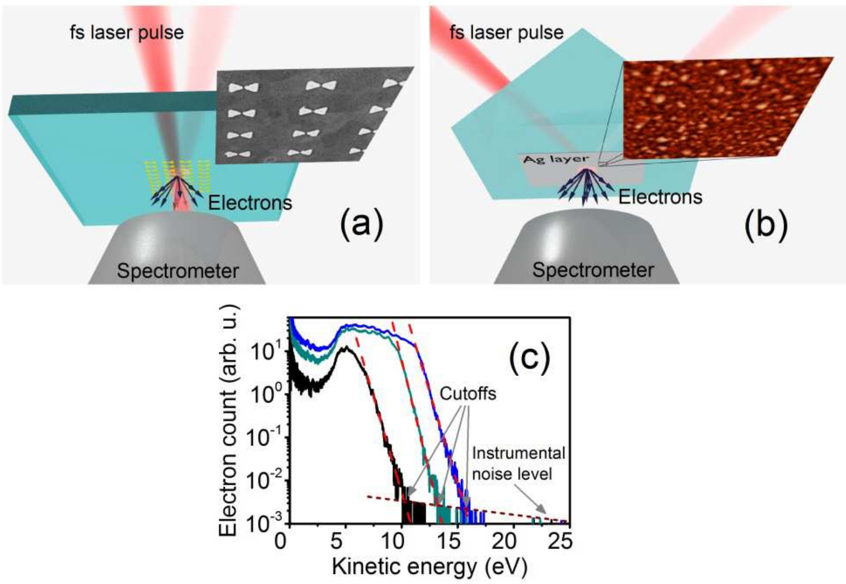

Figure 2. Experimental schemes for measuring photoemission spectra and plasmonic field enhancement induced (a) by localized plasmon fields at gold nanoparticle arrays and (b) by propagating SPP in Kretschmann-coupling configuration on thick silver layers. Photoelectrons emitted from the nanoparticles and from metal layers can directly enter a time-of-flight electron spectrometer. (c) Typical plasmonic photoelectron spectra. Intersection of the red dashed line fitted to the decaying sections of the spectra and the

line fitted to the baseline (instrumental noise floor) define the maximum electron kinetic energy.

This method is based on the nanooptical, plasmonic near-fields induce photoemission from metal surfaces and nanoparticles. The photoemitted and accelerated electrons that acquire the highest kinetic energy in the nanolocalized field are the rescattered electrons from surface, and there is a well-defined, simple relationship between the local field strength and the maximum electron kinetic energy, based on the ponderomotive acceleration mechanism of electrons. I demonstrated that by measuring the highest electron kinetic energy for a given plasmonic structure, the maximum field enhancement can be extracted from the data. The electron spectroscopy measurements were carried out by time of flight spectrometer technique. (Figure 2). Directly with this method measured field enhancement values for various samples was in good agreement with the numerical solution of Maxwell equation based on finite-difference time-domain (FDTD) simulations. Figure 3 shows a field enhancement measurement as example on the sample contains bowtie shape nanoparticles. Results of these experiments and calculations were published in Nano Letters (impact factor of 12.7).

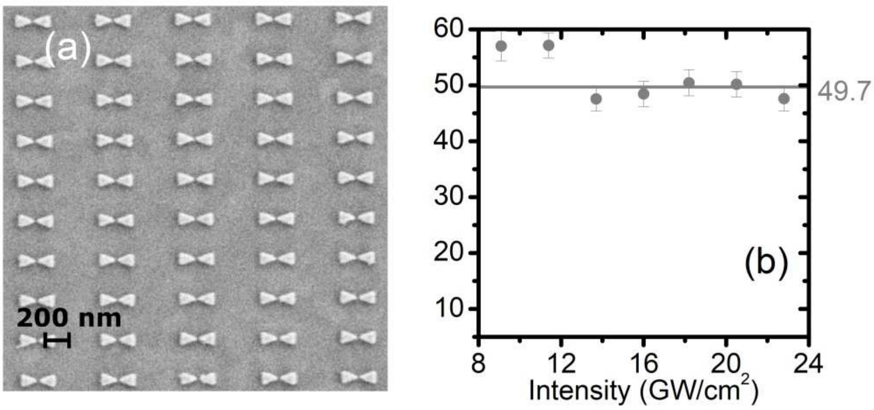

Figure 3. (a) SEM image of sample contains plasmonic bowtie shape nanoparticles and (b) maximum plasmonic field enhancement values extracted from the electron spectral cutoffs as a function of laser intensity. The horizontal line shows the simulated field enhancement value

2. Numerical study of nanoplasmonic electron acceleration

The influence of electron rescattering and image charge was investigated by my own model on nanoplasmonic photoemission and electron acceleration processes induced by femosecond laser pulses [12]. In this model the temporal evolution and spatial distribution of the SPP field was described by an analytic formula. By solving the equations-of-motion for a large number of test electrons in SPP field with different initial positions and photoemission instants, one can model electron spectra measured in corresponding experiments. Electron rescattering can be switched on or off in the model meaning that those electron trajectories that reach the surface again either undergo a fully elastic bounce at a certain instant or are considered reabsorbed, respectively. For describing the influence of image charge during solving the equations of motion, force derived from the image charge was taken into account.

Our simulations confirm that electron rescattering determines essentially the final maximum kinetic energies of electrons, being important for nanoplasmonic field probing [11]. On the other hand, no significant effect was shown from the image charge in a broad range in intensity and wavelength which are relevant for experiments. These results also support the validity of own developed field enhancement measurement method [11].

3. Investigation of nonlinear effects in nanolocalized electromagnetic fields

Surfaces plasmon based magnetic anomalies were study in high intensity laser field [13]. In this work, continuing the former STM and electron emission investigations, concerning electron pairing in strong laser fields, I performed further time-of-flight electron emission studies , changing the angle of polarization of the incident light, exciting surface plasmon oscillations. It has been found, that those parts of the electron spectrum which have been attributed to electron pairing have a significantly different angular dependence around 80 GW/cm^2 where the pairing effect has been found with respect to outside this region (e.g., 120 GW/cm^2). These results have been interpreted as the appearance of ideal or partly ideal diamagnetism, on the one hand, and as anomaly in the magneto-optical effect (rotation) on the other hand, in the same laser intensity region where the pairing effect has been found.

Furthermore the plasmon dispersion was investigated in a high-intensity regime [14]. It has been found experimentally that the surface plasmon dispersion has an oscillatory character as a function of the exciting laser intensity in intense femtosecond laser fields (Figure 4). The oscillatory character of dispersion was detected by the very small change of plasmon resonance angle.

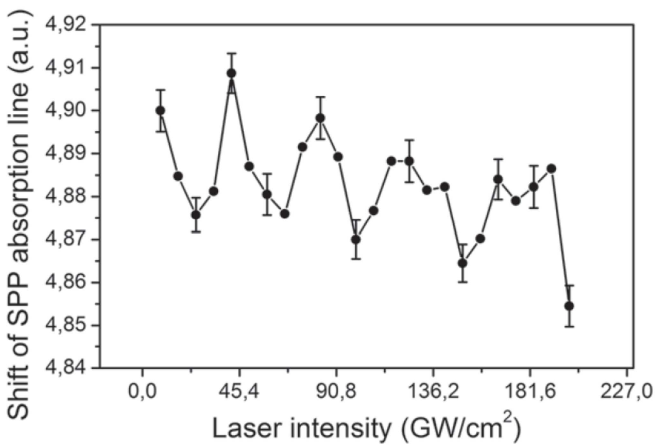

Figure 4. Laser intensity dependence of the angular minimum in the reflected beam.

This phenomena has been interpreted as the result of the dynamic screening of electrons by the strong laser field. A simple model is described in addition to the experimental results, being in good agreement with these findings. The results imply an electron effective mass of around 10 percent less than the free electron mass. The effective mass decreases with increasing laser intensity.

In addition, we published two review articles during the project connecting this research direction [15,16]. In the first review article [15] the former results were summarized concerning electron pairing in strong laser fields. Surface plasmons have been excited by intense femtosecond laser pulses on a gold film at room temperature and their near field has been analyzed by the intensity dependent response of an STM and by studying the spectra of multiplasmon emitted electrons as a function of laser intensity and polarization angle.

In the second review article [16] the theory and basic properties of surface plasmon polaritons were summarized. Plasmons have several unique properties, including their strong-field enhancing effect in near field. This means, among other things, that nonlinear phenomena may be studied at much lower laser intensities. Experiments on both photon and electron emission from a gold film were also presented.

Furthermore during the project 5 conference contributions [17-21] were achieved and I started to supervise one M. Sc. students.

4 References

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