UNIVERSITY OF PÉCS

Doctoral School of Physics

Nonlinear optics and spectroscopy program

Investigation of plasmonic photoemission and electron acceleration induced by ultrashort laser pulses

PhD dissertation

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1. Introduction and objective

With the development of laser physics, lasers delivering pulsed light appeared and evolved continuously. The application of certain techniques such as chirped pulse amplification [1,2] and the proliferation of broadband laser materials (laser dyes, titanium-sapphire [3]) and dispersion compensating tools [4-6] made it possible to significantly enhance the peak intensity of a laser pulse. It also became within reach to control and measure the carrier-envelope phase of the shortest laser pulses [7].

According to the aforementioned developments and numerous inventions made possible to create shorter and shorter laser pulses. The fields of femtochemistry [8] and attophysics [9] could be developed by exploiting the excellent temporal resolution of ultrashort laser pulses.

Investigation of various light-matter interaction processes became a relevant research field of its own and, for instance, the fundamental knowledge about photoemission from a metal surface has expanded significantly in recent years, too [10-15].

The research on surface plasmons [16] is relevant both from the fundamental research point-of-view and for possible applications, as well. Surface plasmons are charge density oscillations of conduction electrons along a metal surface. The evanescent electromagnetic field inherently belonging to surface plasmons can be significantly higher than the electromagnetic field of the incident light [16] and this enhancement factor strongly influences the properties of photoemission [17-19] and the obtained electron bunch [20-23].

My presented research in this thesis is about the investigation of the basic and until recently unknown properties of plasmonic photoemission. During my research I intended to identify the effect of surface roughness on plasmonically generated photocurrents and the electron acceleration in the plasmonic field. For this research goal, I performed experiments, I set up models and carried out calculations. Furthermore, I aimed at the investigation of the properties of carrier-envelope-phase dependent photoemission from plasmonically resonant and nonresonant nanoparticles. I obtained corresponding conclusions from calculations based on the appropriate physical model. I also intended to understand the relationship between electron acceleration near nanostructured surfaces and THz radiation generated at these surfaces. I performed experiments to reveal the underlying phenomena.

2. Applied methods

For the experimental research of surface plasmon enhanced photoemission and electron acceleration, I applied two lasers: a regenerative amplifier and a long-cavity laser oscillator, both can be found at the Wigner Research Center for Physics, Hungarian Academy of Sciences. The regenerative amplifier delivered laser pulses with 1 kHz repetition rate at 795 nm central wavelength with 4 W output power and the pulse length and energy was 40 (±5) fs and 4 mJ, respectively.

The round-trip cavity length of the long-cavity resonator is 80 m that corresponds to 3.6 MHz repetition rate at mode locked operation. The energy of the laser pulses are 220 nJ and the pulse length is between

95-110 fs after the compressor gratings. In this configuration, it was also necessary to use an electrooptic pulse picker to avoid the overlap of electrons induced by subsequent pulses (i. e. slow electrons from a laser pulse can overlap with fast electrons generated by the next pulse on the detector).

I measured the properties of the electron beam with a time-of-flight spectrometer (TOF) and calibration curves were available from the manufacturer for different voltage settings of the electron optics. The acceptance angle for the electrons can be increased by applying the proper voltage combinations. The voltage applied to the drift tube of the device decreases the time between the photoemission and the detection of electrons which is another method to avoid the overlap of counts from subsequent laser pulses.

To understand the properties of the photoemission and electron acceleration generated by localised surface plasmons, it is essential to construct an appropriate model of these processes and draw conclusions based on these calculations. The essence of the model is that in the quasistatic approximation the Laplace equation can be solved at ellipsoidal nanoparticles to obtain the potential [24]. This makes it possible to give the polarizability of the nanoellipsoids in an analytic form and therefore the determination of the electric field at nanoellipsoids can be performed. To calculate the probability of photoemission from a given point of the ellipsoid, it is necessary to choose the right photoemission model and the corresponding formulae. After this, the surface of the particle should be divided into the appropriate number of regions. Inside a given region the electric field can be considered as homogeneous and therefore the photoemission rate is constant. The trajectories of the photoemitted electrons can then be calculated from the Lorentz equation and certain properties of the electron bunch, such as the electron spectra and the carrier-envelope phase dependent properties, can be deduced.

During my research I also investigated the role of plasmonic photoemission in THz generation. For these experiments, it was necessary to obtain and compare the laser intensity dependent THz signal and photocurrent, as well. For the same samples, I performed intensity dependent photoemission measurements by the time-of-flight spectrometer. Measurement of the THz signal and its dependence from the applied intensity was performed on the same samples by an external collaborating research group. The implementation of the experiment was performed with electro-optical sampling method applying a ZnTe crystal, widely used in THz technology [25,26]. By modifying the delay between the THz signal and the laser pulse the temporal profile of the THz signal can be reconstructed and with Fourier transformation the spectral profile can also be obtained.

3. Theses

By applying the aforementioned methods throughout my experiments and model calculations, I obtained the following new results.

1. In my experiments, I detected electrons with high kinetic energy that is several tens of times higher than the energy of the exciting photons. The extension of the high-energy end of the spectra is attributed to the local ponderomotive potential modulated on nm scale along the rough surface. Therefore, the ponderomotive potential has a different effect on electrons photoemitted from different locations of the surface. The relatively high cutoff value typical for the spectra can be attributed to electrons photoemitted from relatively few hot-spots of the surface. Furthermore, I also demonstrated experimentally that the relationship between the extent of surface roughness and the most energetic electrons is neither trivial nor monotonous. This problem therefore requires further numerical investigation. [F1]

2. With model calculations, I have shown that plasmonically enhanced electron acceleration from a rough surface is determined by the coupling of the surface plasmon polariton and localised surface plasmon modes. In the physical model that I created, the effect of the surface roughness can be approximated with nanoellipsoids on the surface having the appropriate size distribution. With the help of the calculations based on the model, it was possible to prove that the plasmon resonance of such surface grains determine the maximal electron kinetic energies in the measured spectra. [F1]

3. I proved with further model calculations that the left-right asymmetry of the photoemission and acceleration from isolated plasmonic nanoparticles can be used to determine the carrier-envelope phase of few-cycle laser pulses, provided that the resonance eigenfrequency of the nanoparticle is far from the carrier frequency of the applied laser pulse. In case of resonant nanoparticles, the effect of the carrier-envelope phase is washed out because of the limited bandwidth of the plasmon resonance. [F2]

4. I delivered an experimental proof that THz radiation generated on nanostructured samples with ultrashort laser pulses is determined by plasmonically photoemitted and accelerated electrons above a certain laser peak intensity. I found that the critical intesity was around 15 GW/cm^2. Above that intensity the photocurrent scaling is similar to the intensity scaling of the THz radiation, whereas below that intensity significant scaling differences can be observed, which can be attributed to another THz generation mechanism. In this way, I described different mechanisms of visible-THz conversion on nanostructured samples. [F3]

4. Publications:

Publications related to the thesis:

[F1] **I. Márton**, V. Ayadi, P. Rácz, T. Stefaniuk, P. Wróbel, P. Földi, P. Dombi, Plasmonics **11**, 811 (2016).

[F2] P. Földi, **I. Márton**, N. Német, V. Ayadi, and P. Dombi, Applied Physics Letters **106**, 013111 (2015).

[F3] D. K. Polyushkin, **I. Márton**, P. Rácz, P. Dombi, E. Hendry, W. L. Barnes, Physical Review B **89**, 125426 (2014).

Conferernce abstract related to the thesis

[K1] P. Földi, **I. Márton**, N. Német, P. Dombi: Fém nanorészecskék fotoemissziójának kontrollja rövid, plazmonikusan erősített lézerimpulzusokkal, Kvantumelektronika 2014, Budapest, Magyarország

Publications not related to this thesis:

[F4] P. Rácz, Zs. Pápa, **I Márton**, J. Budai, P. Wróbel, T. Stefaniuk, C. Prietl, J. R. Krenn, P. Dombi, Nano Letters **17**, 1181 (2017).

[F5] M. Aladi, J. Bakos, I.F. Barna, A. Czitrovszky, G. Djotyan, P. Dombi, D. Dzsotjan, I. Földes, G. Hamar, P. Ignácz, M. Kedves, A. Kerekes, P. Lévai, **I. Márton**, A. Nagy, D. Oszetzky, M. Pocsai, P. Rácz, B. Ráczkevi, J. Szigeti, Zs. Sörlei, R. Szipöcs, D. Varga, K. Varga-Umbrich, S. Varró, L. Vámos, Gy. Vesztergombi, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment **740**, 203 (2014).

[F6] M. Aladi, **I. Márton**, P. Rácz, P. Dombi, I.B. Földes, High Power Laser Science and Engineering **2**, e32 (2014).

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