Investigation of electron beams generated by femtosecond surface plasmons

Ph. D. Theses

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Background

There was a significant development in recent years in femtosecond laser technology and its applications. As a results of this, it is possible not only to shape laser pulses almost arbitrarily and to compress them, but also to precisely control the time evolution of the electric field of so-called few-cycle laser pulses (typically having 3-6 fs duration) by the carrier–envelope phase (CEP) [Jones00]. With such phase-stabilized laser pulses becoming ever shorter and having ever higher energies, it was possible to reach intensity regimes where previously unknown processes could be investigated (e.g. production of single attosecond pulses by high harmonic generation [Krausz09] and other extreme nonlinear optical processes). The study of ultrafast atomic, molecular and solid-state processes became within reach with these unique femtosecond and attosecond light sources with unprecedented temporal resolution [Goulielmakis08].

Intense parallel development took place in the field of plasmonics as part of nano-optics research. Surface plasmon polaritons are special electromagnetic waves propagating along metal surfaces. Their field can be localized even to sub-wavelength-sized regions of space, therefore they can be applied in many microscopy methods below the diffraction limit [Stockman07]. In addition, research was also carried out for the development of compact plasmonic opto-electronic devices using surface plasmon waveguides [Outon 08], such as optical chip modifyer devices for e.g. optical computers.

As a synergy of these two fields of research, interesting results came from the investigation of ultrafast photoemission generated from metal targets in different configurations [Apolonoski04], that can be further enhanced by surface plasmons. The investigation of electron beams generated and accelerated in this process is an active field of research [Kupersztych01]. Developing photocathodes based on ultrafast electron sources and controlling the properties of these electron beams could play an important role in the realization of ultrafast structural analysis methods like ultrafast electron diffraction [Siwick03], and in the development of other surface physics methods where high temporal and spatial resolution is available at the same time. Furthermore, ultrafast electron beams can gain importance in new types of X-ray sources and in other fields of research.

In related field, several fundamental research results have been demonstrated by utilizing state-of-the-art femtosecond light sources. For example, the carrier-envelope phase dependence of multiphoton induced photoemission from gold surfaces [Apolonski04] was shown, and attosecond control of electron beams generated from metallic nanotips was demonstrated [Krüger11]. In addition, above threshold ionisation was observed in this configuration [Schenk10], and the extinction of the quiver motion of electrons was demonstrated in the mid-infrared wavelength range [Herink12].

It is known, that the generation of ultrashort electron beams is achievable by surface plasmon polariton induced photoemission [Kupersztych01, Irvine04]. The unique feature of this process is that electron acceleration occurs on the nanoscale (in addition to photoemission) above a certain intensity in the highly evanescent electric field of surface plasmons.

This phenomenon was demonstrated by 30-150 fs long driver laser pulses in the beginning of the millennium, and electron beams were produced with up to 2 keV energy [Irvine04]. However, more control options of the properties of these electron beams have not yet been investigated, and experiments have not been carried out with few cycle carrier-envelope phase stabilized laser pulses, where the laser pulse length is significantly shorter than plasmon lifetime. This would be an interesting option to generate even shorter electron beams and investigate fundamental plasmonic phenomena by an extremely high-bandwidth excitation.

Objectives

I carried out my work at the Research Institute for Solid-State Physics and Optics (currently Wigner Research Centre for Physics) and I joined research efforts concerning the experimental and numerical investigation of surface plasmon enhanced electron acceleration. I set the following objectives:

1. Realization of a simplified model concerning plasmonic electron acceleration, that can describe all relevant aspect of the process, and studying this phenomenon in different parameter regimes.
2. The investigation of control possibilities of ultrafast electron beams that can be generated in this scheme with the help of modelling tool developed. Studying the fundamental scaling laws of the surface plasmon enhanced electron acceleration process by the model applied.
3. Performing experiments concerning this phenomenon with few-cycle carrier-envelope phase stabilized pulses, and investigating electron beams generated in this scheme by

different methods such as surface autocorrelation, electron spectroscopy etc. In addition, evaluation and interpretation of results related to the existence of few-cycle plasmonic wave packets and properties of the plasmonic electron beam.

New scientific results

During the realization of the above mentioned objectives, I achieved and published the following new scientific results:

1. I realized a semi-classical model with a short processing time concerning the plasmonic electron acceleration process based on analytical approximations and a numerical solution of the equation of motion. I could accurately reproduce the results of more complex and more time-consuming models based on the numerical solution of Maxwell's equations. I investigated plasmonic electron acceleration with this self-developed model in different intensity regimes, and for the cases of multiphoton induced and tunneling photoemission. I established that the kinetic energy distribution of the emitted electron beam depends on the tunnelling time and that quasi-monoenergetic spectra are observed for certain tunnelling times [1].

2. I explored control possibilities of the spectra of plasmonic electron beams with the model developed. I found that the maximum kinetic energy of the electrons is tunable by the carrier-envelope phase in the case of few-cycle laser pulses and flat surfaces. Moreover, spectra can be made more monoenergetic by spatial nano-confinement of the emission region [1]. This result is particularly interesting for potential application since such short and monoenergetic electron beams can find a use in many ways, for example in ultrafast electron diffraction enabling high spatial and temporal resolution at the same time.

3. I investigated the fundamental scaling laws of the plasmonic electron acceleration process with the model developed, and I identified non-ponderomotive features of the electron acceleration taking place in the evanescent electromagnetic field for the first time to my knowledge. I pointed out that the maximum kinetic energy does not necessarily scale with the well-known quadratic ponderomotive dependence with the

maximum field strength and the wavelength, and I showed that these anomalous scaling properties depend on the laser pulse length. I identified as the reason of this phenomenon that the acceleration process occurs in a strongly evanescent field with a decay length comparable to the wavelength [2].

4. I performed experiments concerning the plasmonic electron acceleration process with few-cycle carrier-envelope phase stabilized laser pulses. It was possible to achieve electron bunches with keV maximum kinetic energy even with such extremely short pulses and I found no observable phase dependence in the electron-spectra. I identified the existing surface roughness on the samples as the reason for this low phase contrast, based on model calculations, and surface analytical measurements such as scanning tunneling microscopy [3].

5. I developed an evaluation method related to ultrashort surface plasmon characterisation by surface autocorrelation measurements. In order to evaluate higher order autocorrelation functions (exploiting nonlinear plasmonic photoemission), I took into consideration independent measurements concerning the surface roughness of the sample as well as the temporal and spatial intensity profiles of the surface plasmon generating laser pulses. With this evaluation method, we could establish that it is possible to generate few-cycle plasmonic wavepackets by similarly short few-cycle laser pulses [4].

List of publications

Publications related to the thesis

[1] P. Dombi , **P. Rácz**, B. Bódi, *"Surface plasmon enhanced electron acceleration with few-cycle laser pulses,"* Laser Part. Beams **27**, 291–296 (2009).

[2] **P. Rácz** and P. Dombi, *"Non-ponderomtive electron acceleration in ultrashort surface plasmon fields,"* Phys. Rev. A **84**, 063844 (2011).

[3] **P. Rácz**, S. E. Irvine, M. Lenner, A. Mitrofanov, A. Baltuska, A. Y. Elezzabi, and P. Dombi, *"Strong-field plasmonic electron acceleration with few-cycle, phase-stabilized laser pulses,"* Appl. Phys. Lett. **98**, 111116 (2011).

[4] P. Dombi, S. E. Irvine, **P. Rácz**, M. Lenner, N. Kroó, G. Farkas, A. Mitrofanov, A. Baltuska, T. Fuji, F. Krausz and A. Y. Elezzabi, "*Observation of few-cycle, strong-field phenomena in surface plasmon fields,"* Opt. Express **23**, 24206-24212 (2010).

Other publications

[5] P. Dombi and **P. Rácz**, "*Carrier-envelope phase-controlled laser-surface interactions,"* Proc. SPIE **6892,** 1J (2008).

[6] P. Dombi and **P. Rácz**, *"Ultrafast monoenergetic electron source by optical waveform control of surface plasmons,"* Opt. Express **16**, 2887 (2008).

[7] P. Dombi, **P. Rácz**, M. Lenner, V. Pervak, F. Krausz *"Dispersion management of femtosecond laser oscillators with highly dispersive mirrors,"* Opt. Express **17**, 20598 (2009).

[8] M. Lenner, **P. Rácz**, P. Dombi, G. Farkas and N. Kroó, *"Field enhancement and rectification of surface plasmons detected by scanning tunneling microscopy,"* Phys. Rev. B **83**, 205428 (2011).

References

[Apolonski04] A. Apolonski, P. Dombi, G. G. Paulus, M. Kakehata, R. Holzwarth, T. Udem, C. Lemell, K. Torizuka, J. Burgdörfer, T. W. Hänsch, and F. Krausz, *"Observation of light-phase sensitive photoemission from a metal,"* Phys. Rev. Lett. **92,** 073902 (2004).

[Goulielmakis08] E. Goulielmakis, M. Schultze, M. Hofstetter, V. Yakovlev, J. Gagnon, M. Uiberacker, A. L. Aquila, E. M. Gullikson, D. T. Attwood, R. Kienberger, F. Krausz, U. Kleineberg, *"Single-cycle nonlinear optics,"* Science **320**,1614 (2008).

[Herink12] G. Herink, D. R. Solli, M. Gulde, C. Ropers, *"Field-driven photoemission from nanostructures quenches the quiver motion,"* Nature **483**, 190 (2012)

[Irvine04] S. E. Irvine, A. Dechant, A. Y. Elezzabi, "Generation of 0.4-keV Femtosecond Electron Pulses using Impulsively Excited Surface Plasmons," Phys. Rev. Lett. **93**, 184801 (2004).

[Jones00] D. J. Jones, S. A. Diddams, J. K. Ranka, A. Stentz, R. S. Windeler, J. L. Hall,and S. T. Cundiff, *"Carrier-Envelope Phase Control of Femtosecond Mode-Locked Lasers and Direct Optical",* Science **288***,* 635 (2000)

[Krausz09] F. Krausz, and M. Yu. Ivanov, *"Attosecond Physics,",* Rev. Mod. Phys. **81**, 163 (2009).

[Krüger11] M. Krüger, M. Schenk, P. Hommelhoff, "*Attosecond control* of *electrons emitted from* a *nanoscale metal tip,"* Nature **475**, 78 (2011).

[Kupersztych01] J. Kupersztych, P. Monchicourt, M. Raynaud, *"Ponderomotive Acceleration of Photoelectrons in Surface-Plasmon-Assisted Multiphoton Photoelectric Emission,"* Phys. Rev. Lett. **86**, 5180 (2001).

[Oulton08] R. F. Oulton, V. J. Sorger, D. A. Genov, D. F. P. Pile, X. Zhang,"*A hybrid plasmonic waveguide* for *subwavelength confinement and long-range propagation ,"* Nat. Photonics **2**, 496 (2008), and references therein.

[Schenk10] M. Schenk, M. Krüger, P. Hommelhoff, *"Strong-Field Above-Threshold Photoemission from Sharp Metal Tips,"* Phys. Rev. Lett. **105**, 257601 (2010)

[Siwick03] B. J. Siwick, J. R. Dwyer, R. E. Jordan, and R. J. D. Miller, *"An atomic-level view of melting using femtosecond electron diffraction,"* Science **302**, 1382-1385 (2003).

[Stockman07] M. I. Stockman, M. F. Kling, U. Kleineberg, F. Krausz, *"Attosecond nanoplasmonic-field microscope,",* Nat. Photonics **1**, 539 (2007) and references therein.