## OTKA 73728 Zarojelentes.

Az anyag attoszekundumos dinamikája ultraintenzív lézerterekben optikai cikluson belülii időbeli és hullámhosszon belüli, nanométeres térbeli tartományokban.

## OTKA 73728 Final report.

Attosecond dynamics of matter in ultra-high laser fields with sub-cycle temporal and sub-wavelength, nanometer-scale spatial resolution.

Short summary. We analysed entanglement in the interaction of electrons with a strong quantized radiation field, which leads to the generation of number-phase minimum uncertainty states. On the basis of these results we introduced the concept of "attosecond shot-noise" and shown how the Hanbury Brown and Twiss (HBT) type correlations can be used for probing extreme light signals. We developed the Wigner function description of the kinematics of electrons in twodimensional geometry. We studied in details the role of carrier-envelope phase difference effects in strong field laser- matter interactions at surfaces (thin metal or plasma layers and graphene). We predicted the new phenomena of "relativistic clipping" appearing in the scattering of ultrasort pulses on graphene, which may lead to rectangular optical pulses. We have observed experimentally and described theoretically that in surface plasmon enhanced evanescent electric fields, strong-field optics effects exist (high-harmonic generation, nonlinear electron emission and acceleration). These are characteristic for surface plasmon physics, they otherwise occur only in fields, higher than those produced by our exciting laser. We demonstrated that the electrons leave the surface in 2-3 fs-long bunches, following the intensity envelope of the surface plasmons. We measured and theoretically interpreted for the first time HBT type correlations in decaying surface plasmon light, and found the transition from antibunching to bunching.

Results. Continuing our work on the above-threshold electron de Broglie waves, generated by an intense laser pulse at a metal surface [E1], we have analysed the dynamics and statistics of attosecond electron pulses. Accordingly, above the metal surface, there exist "collapse bands", where the electron current is erratic or noise-like, and there exist "revival layers", where the electron current consists of ultrashort pulses of attosecond duration [26]. In the course of our research, we have put an emphasize on the study of the statistical properties of extreme light signals, both in the extremely low and very high intensity regime. In part we have used the new quantum coherence theory, which have been developed by us recently [E2, 9]. We have worked out a unified treatment of classic and recent Hanbury Brown and Twiss type counting experiments for both bosons and fermions. The new formalism relies on the Boole algebra of counting events and the associated classical probability space. In the framework of this description several two-point correlation experiments have been successfully described without the use of standard second quantization, even in the single-particle regime [9, 21, 22, 30, 34]. The exact analytic solutions of the energy eigenvalue equation of the system consisting of a free electron and one mode of the quantized radiation field are used for studying the physical meaning of a class of number-phase minimum uncertainty states. It is proved that the most fundamental interaction in quantum electrodynamics – namely the interaction of a free electron with a mode of the quantized radiation field – leads quite naturally to the generation of the mentioned minimum uncertainty states [3]. In the electron's coordinate representation the physical meaning of the expansion coefficients of these entangled states are the joint probability amplitudes of simultaneous detection of an electron and of a definite number of photons. The

photon occupation probabilities in these states preserve their functional form as time elapses, but they depend on the location in space-time of the detected electron. An analysis of the entanglement entropies derived from the photon number distribution has also been given [4, 12]. The study of the time evolution of entanglement between the initially separated electron wave packet and the radiation mode led to the conclusion that in general there are non-vanishing entropy remnants in the subsystems after the interaction. On the basis of the model discussed, the calculated values of the entropy remnants crucially depend on the character of the assumed switching –on and –off of the interaction, and explicitly reflect back the irreversible character of the high-intensity Compton process [4, 39]. We have also studied the question, whether to what extent attosecond pulses can be considered as simply broad-band classical signals? If they are not classical, then by what means could one sample them to study their spatio-temporal structure? Instead of first-order interference, we proposed to carry out intensity-intensity correlation measurements on split nano-pixels, which are not sensitive against phase distortions. By changing the tilt angle of the impinging attosecond pulses with respect to the detector array, the modal structure of the pulses could be monitored by measuring the current-current correlations, and this would yield a delayed coincidence curve. We have modelled the attosecond pulses by many-mode quantum-mechanical phase eigenstates, and introduced the concept of attosecond shot noise. According to our theory, the contrast of the coincidence curve is expected 4/3 relative to the Poissonian shot noise level [25, 31, 32, 35, 36].

We carried out further experimental investigations of femtosecond phenomena in surface plasmon polariton (SPO) fields[15-19]. We found that the scattered light from the surface where SPOs propagate has a significantly different angular distribution when the phenomenon is induced by intense femtosecond laser pulses. The SPO-emitted light intensity shows a nonlinear dependence on that of the inducing laser light. Nonlinear frequency conversion phenomena mediated by SPOs on the surface, such as second harmonic generation and continuum spectrum generation were also observed. In a collaboration with the Vienna University of Technology we investigated SPO enhanced electron acceleration induced by few-cycle laser pulses. Using state-of-the-art lasers in Vienna, we could demonstrate that the electrons leave the surface in 2-3 fs-long bunches. The length of the electron bunch is determined solely by the intensity envelope of the SPO generating laser pulse.

Concerning above-threshold nonlinear photoelectric effect, in accord with our theoretical calculations, the broad energy spectra of surface *plasmon mediated electron emission* was also found experimentally. The results of these investigations support the physical picture offered by our concept of plasmon-induced oscillating near field of a double layer at the metal-vacuum interface. The electrons have been described by dressed quantum states (generalized Volkov states), which contain the interaction with the plasmon field and the induced dipole interaction non-perturbatively. Similarly to the electron spectra, large deviations from the perturbative behaviour have also been found in the intensity dependence of the emitted fundamental and the *second harmonic signals*, even at moderate incoming laser intensities [5, 6, 8, 20, 27].

In studying the quantum dynamics of the surface electrons, we have shown that the wave function for a free particle in two dimensions and in a state with definite values of the energy and angular momentum shows some unusual effects (e.g. the appearance of quantum anti-centrifugal force). Based on the *Wigner function*, we have identified the origin of these subleties as interference in two-dimensional space where Huygens' principle breaks down [7].

In the nonlinear surface-plasmon-mediated electron emission, a platoless energy distribution was found, when the pulse duration of the exciting laser was comparable with the life-time of the

surface plasmon oscillation. The appearance of the large-energy electrons cannot be explained with standard non-perturbative theories, even if one takes into account the field enhancement due to the surface plasmons. The experimental results for lower laser intensities are reproduced quite well in our theoretical calculations, both in the low-enegy and in the high-energy part of the spectra. These results firmly support the applicability of our concept of plasmon-induced oscillating near field of a double layer at the metal-vacuum interface [10]. We have studied the statistical properties of light emitted by suface plasmon oscillations, generated in a gold layer in the Kretschmann geometry, and a detailed analysis of the coherence properties of the SPOs has been carried out. By developing a new formalism for describing the quantum statistical properties of the SPOs as single-photon emitters, the Fano factor and the covariance of counting events, in delayed or spatially separated detectors have been calculated on the basis of the derived analytic formulas. The transition from antibunching to bunching has also been predicted and we have received a reasonable agreement with our experimental results [11, 29, 30, 37]. In the context of our theoretical work on extreme light sources, we have investigated in details the characteristics of the synchrotron radiation emitted by ultrarelativistic electrons in the interior of a coaxial cylindrical mirror. This is an unconventional geometrical arrangement, where the superradiance may be realized in two dimensions, in the Röntgen regime [13, 40], thanks to the found whispering-gallery modes of very high azimuthal angular momenta. This analysis supported our original physical picture according to which, if the electron meets with its own radiation field emitted earlier, and reflected back by the mirror, then there is a constructive interference between this retarded self-field and the actually emitted radiation field. In the context of extreme electromagnetic signals and the photon localization problem, we have made a connection of our research with the famous needle radiation concept [24, 28, 33, 36], and used this study [32] in describing the spatio-temporal localization [39] due to high angular momenta. We have derived new results concerning the reflection and transmission of few-cycle laser pulses on the thin layer of electrons and holes of graphene, which follow ultrarelativistic kinematics. Graphene, the two dimensional one- or few-atom layer of carbon atoms, has many unusual and unique electronic properties, which are well described by massless charged Dirac particles. We expected that the electromagnetic response of such a material would be very sensitive to the finest temporal details of ultashort laser pulses, even at moderate intensities. On the basis of numerical solutions, we illustrated the considerable distortions of the radiation during the scattering, and the effect of the carrier-envelope phase difference. A 'saturation' effect has been found, which causes a sort of 'relativistic clipping', resulting in generation of rectangular optical signals, which follow the carrier-enevelope phase difference [14].

## Publications.

[E1] Varró S and Farkas Gy: Attosecond electron pulses from interference of above-threshold de Broglie waves. *Laser and Particle Beams*, Vol. **26**, No.1, 9-19 (2008). (Earlier work.)

[E2] Varró S: Correlations in single-photon experiments. Fortschritte der Physik, 56, 91-102 (2008). (Earlier work.)

[3] Varró S: Entangled photon-electron states and the number-phase minimum uncertainty states of the photon field. *New Journal of Physics*, **10**, 053028 (35 pages) (2008)

[4] Varró S : Entangled states and entropy remnants of a photon-electron system. Physica Scripta **T140** (2010) 014038 (8pp)

- [5] Kroó N, Farkas Gy, Dombi P, Varró S: Nonlinear processes induced by the enhanced, evanescent field of surface plasmons excited by femtosecond laser pulses.

  OPTICS EXPRESS 21656, **16**, No.26, (6 pages) (2008)
- [6] Kroó N, Varró S, Farkas Gy, Dombi P, Oszetzky D, Nagy A, Czitrovszky A: Nonlinear plasmonics. *Journal of Modern Optics*, **55**, (19-20), pp. 3203 3210. (2008)
- [7] Schleich W P, Dahl J P and Varró S: Wigner function for a free particle in two dimensions. *Optics Communication*, **283**, 786-789 (2010)
- [8] Varró S, Kroó N, Farkas Gy and Dombi P: Spontaneous emission of radiation by metallic electrons in the presence of electromagnetic fields of surface plasmon oscillations. *Journal of Modern Optics*, **57**, No. 1. 10 January, 80-90. (2010)
- [9] Varró S; The role of self-coherence in correlations of bosons and fermions in linear counting experiments. Notes on the wave-particle duality;

Fortschritte der Physik – Progr. Phys.; **59**, No. 3 – 4, 296 – 324 (2011)

- [10] Varró S, Kroó N; Nonlinear photoelectron emission from metal surfaces induced by short laser pulses. The effects of field enhancement by surface plasmons; *Applied Physics B: Lasers and Optics* (Germany); **105**, 509-515 (2011)
- [11] Varró S, Kroó N, Oszetzky D, Nagy A, Czitrovszky A; Hanbury Brown Twiss type correlations with surface plasmon light; *Journal of Modern Optics*; **58**, 2049-2057 (2011)
- [12] Varró S; Intensity effects and absolute phase effects in nonlinear laser-matter interactions; In *Laser Pulse Phenomena and Applications* (Ed. Duarte F J); Chapter 12, pp 243-266 (2011) (Rijeka, InTech, 2010) ISBN: 978-953-307-405-4.
- [13] Varró S; Interference phenomena and whispering-gallery modes of synchrotron radiation in a cylindrical wave-guide;

In *Free Electron Lasers* (Ed. Varró S); in press, Chapter 7, pp 175-186 (2012) (Rijeka, InTech, 2012) ISBN: 978-953-51-0279-3.

- [14] Varró S; Graphene-based carrier-envelope phase difference meter. Contributed talk. Book of Abstracts LEI-2011, p. 43., The 2<sup>nd</sup> International Conference on Light at Extreme Intensities. (November 14-18, 2011 Szeged, Hungary). To appear in the AIP Conf. Proc. of LEI-2011 (2012).
- [15] P. Dombi, P. Rácz "Ultrafast electron emission from metals: the role of surface plasmon ", invited paper number 7600-39, SPIE Photonics West, San Francisco, USA, (2010).
- [16] P. Dombi, P. Rácz, M. Lenner, N. Kroó, G. Farkas, A. Mitrofanov, A. Baltuska, T. Fuji, F. Krausz, S. E. Irvine, A. Elezzabi, "Ultragyors fényforrásokkal keltett felületi plazmonok", Fizikus Vándorgyűlés (in Hungarian), Pécs, Hungary (2010).
- [17] P. Rácz, S. E. Irvine, M. Lenner, N. Kroó, G. Farkas, T. Fuji, F. Krausz, A. Y. Elezzabi and P. Dombi, "Few-cycle surface plasmon enhanced electron acceleration", 31st European Conference on Laser Interaction with Matter (ECLIM), Budapest, Hungary (2010).
- [18] K. Varjú, J. A. Fülöp, P. Dombi, Gy. Farkas and J. Hebling, "Attosecond Pulse Generation in Noble Gases in the Presence of Extreme High Intensity THz Pulses", Conference on Lasers and Electro-optics (CLEO), paper: JThE120 (2010).
- [19] E. Balogh, J.A. Fülöp, J. Hebling, P. Dombi, G. Farkas, K. Varjú, "Attosecond Pulse Generation in Noble Gases in the Presence of Extreme High Intensity THz Pulses", 31st European Conference on Laser Interaction with Matter (ECLIM), Budapest, Hungary (2010).

- [20] Kroó N; Multiplasmon processes in enhanced laser fields; Invited talk. Book of Abstracts PQE-2010, p. 152., The 40<sup>th</sup> Winter Colloquium on the Physics of Quantum Electronics. (January 3-7, 2010 Snowbird, Utah, USA)
- [21] Varró S; Correlations in single-quantum experiments. Notes on the wave-particle duality; Invited talk. Book of Abstracts PQE-2010, p. 252., The 40<sup>th</sup> Winter Colloquium on the Physics of Quantum Electronics. (January 3-7, 2010 Snowbird, Utah, USA)
- [22] Varró S; Neutron antibunching in a recent experiment; Poster. Book of Abstracts PQE-2010, p. 253., The 40<sup>th</sup> Winter Colloquium on the Physics of Quantum Electronics. (January 3-7, 2010 Snowbird, Utah, USA)
- [23] Varró S; Classical phenomenology of photon anticorrelations in linear counting experiments; Contributed talk. Book of Abstracts CEWQO-2010, p. 63., 17<sup>st</sup> Central European Workshop on Quantum Optics. (June 6-11, 2010 St Andrews, United Kingdom)
- [24] Varró S; Selényi Pál és a kvantumelektrodinamika. Az interferencia szerepe egyfotonos korrelációs kísérletekben (Pál Selényi and the quantumelectrodynamics. The role of interference in single-photon correlation experiments); Invited talk. Book of Abstracts, p. 75. Fizikus Vándorgyűlés (Hungarian Physicists Meeting, August 24-27, 2010 Pécs, Hungary) (in Hungarian)
- [25] Varró S; Attosecond shot noise; Contributed talk. Book of Abstracts ECLIM XXXI, p. 46., 31<sup>st</sup> European Conference on Laser Interaction with Matter. (September 6-10, 2010 Budapest, Hungary)
- [26] Varró S; Temporal Talbot effect in propagation of attosecond electron waves. Poster. Book of Abstracts ECLIM XXXI, p. 83., 31<sup>st</sup> European Conference on Laser Interaction with Matter. (6-10 September, 2010 Budapest, Hungary)
- [27] Varró S, Kroó N; Nonlinear plasmonics with short laser pulses. Invited talk. Book of Abstracts ALT'10, p. 94., 18<sup>th</sup> International Conference on Advanced Laser Technologies. (September 11-16, 2010 Egmond aan Zee, the Netherlands)
- [28] Varró S; On the question of the existence of needle radiation. The 100 years old experiment on wide-angle interference by Pál Selényi and the modern concept of photons; Invited talk. Book of Abstracts PQE-2011, p. 252., The 41<sup>st</sup> Winter Colloquium on the Physics of Quantum Electronics. (January 2-6, 2011 Snowbird, Utah, USA)
- [29] Kroó N, Varró, Czitrovszky A, Nagy A, Oszetzky D; Hanbury Brown and Twiss type correlations with surface plasmons; Invited talk. Book of Abstracts PQE-2011, p. 152., The 41<sup>st</sup> Winter Colloquium on the Physics of Quantum Electronics. (January 2-6, 2011 Snowbird, Utah, USA)
- [30] Varró S, Kroó N; Theoretical aspects of Hanbury Brown and Twiss type correlations mediated by surface plasmon oscillations; Poster. Book of Abstracts PQE-2011, p. 253., The 41<sup>st</sup> Winter Colloquium on the Physics of Quantum Electronics. (January 2-6, 2011 Snowbird, Utah, USA)
- [31] Varró S; Attosecond shot noise and electron interference; Poster. Book of Abstracts PQE-2011, p. 253., The 41<sup>st</sup> Winter Colloquium on the Physics of Quantum Electronics. (January 2-6, 2011 Snowbird, Utah, USA)
- [32] Varró S; Spatio-temporal extension of extreme electromagnetic signals; Invited talk. Book of Abstracts CSCP-2011, p. 17., The 9<sup>th</sup> Workshop on Complex Systems of Charged Particles and their Interaction with Electromagnetic Radiation. (April 13-15, 2011 Moscow, Russia)
- [33] Varró S; Needle radiation, wide-angle interference and quantum nano-optics. The 100 years old experiment on wide-angle interference by Pál Selényi, a Hungarian scholar at the cradle of the modern concept of photons; Invited talk. Conference Guide EuroNanoForum 2011, p. 11., EuroNanoForum 2011. (May 30 June 1, 2011 Budapest, Hungary)
- [34] Varró S; Neutron antibunching as a single-particle self-coherence effect; Contributed talk. Book of Abstracts ECNS-2011, p. 253., 5<sup>th</sup> European Conference on Neutron Scattering (July 17-22, 2011 Prague, Czech Republic)

- [35] Varró S; Extrém elektromágneses sugárzások kiterjedése térben és időben (Spatio-temporal extension of extreme electromagnetic signals); Invited talk. Sokszínű Optika (8<sup>th</sup> School on Quantum Electronics, August 24-26, 2011 Szeged, Hungary) (in Hungarian)
- [36] Varró S; Needle radiation and wide-angle interference. Invited talk. Book of Abstracts ALT'11, p. 49., The 18<sup>th</sup> International Conference on Advanced Laser Technologies. (September 3-8, 2011 Golden Sands, Bulgaria)
- [37] Varró S, Kroó, Oszetzky D, Nagy A, Czitrovszky A; Intensity-intensity correlations of surface plasmon light. Poster. Book of Abstracts ALT'11, pp. 160-161., The 19<sup>th</sup> International Conference on Advanced Laser Technologies. (September 3-8, 2011 Golden Sands, Bulgaria)
- [39] Varró S; Multiphoton Kramers–Heisenberg formula for treating high harmonic generation on bound electrons. Poster. Book of Abstracts LEI-2011, p.108., The 2<sup>nd</sup> International Conference on Light at Extreme Intensities. (November 14-18, 2011 Szeged, Hungary)
- [40] Varró S; Constructive self-interference of synchrotron radiation along a cylindrical mirror. Poster. Book of Abstracts LEI-2011, p.122., The 2<sup>nd</sup> International Conference on Light at Extreme Intensities. (November 14-18, 2011 Szeged, Hungary)
- [41] Varró S; Graphene-based carrier-envelope phase difference meter. Contributed talk. Book of Abstracts LEI-2011, p. 43., The 2<sup>nd</sup> International Conference on Light at Extreme Intensities. (November 14-18, 2011 Szeged, Hungary). To appear in the *AIP Conf. Proc. of LEI-2011* (2012).