

Varró S, OTKA 73728 Final Report (2012).

OTKA 73728 Zarojelentes.

Az anyag attoszekundumos dinamikája ultraintenzív lézerterekben optikai cikluson belüli 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 two-dimensional 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 ultrashort 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 subtleties 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-energy 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 surface 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 ultrashort 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-envelope phase difference [14].

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