

Final Report on OTKA project 48324

We carried out research according to the project proposal concepts. We performed studies of fundamental physical processes of laser-matter interaction both theoretically and experimentally predicted by QED. With these objectives, we planned to continue those research field that we started in previous years, also supported by former OTKA projects, all evaluated as „excellent” in previous final evaluations.

According to the objectives we had to generate intense laser fields to investigate atto- and nanophysical processes on sub-cycle temporal and nanometer spatial scales, all these, however, with very limited financial resources. Therefore, we realized these goals with surface plasmons, the field of which can reach very high field strength values. We managed to generate these field strengths with simple laser oscillators (without having to rely on amplified system) delivering higher pulse energy than standard femtosecond oscillators. The combination of these sources with surface plasmon generation resulted in the generation of plasmonic fields that are two orders of magnitude higher than the plasmon generating laser field itself. Therefore, the $I \cdot \lambda_L$ product (where I is the intensity in the plasmonic field and λ_L is the laser central wavelength) reaches 1 eV, where, as it is well known in intense laser physics, new type of interaction processes appear with non-perturbative character. The investigation of these processes was restricted to groups possessing expensive, amplified laser systems.

To understand the properties of the intense field generated this way (limited to a region extending a couple of times λ_L) and the fundamental processes that take place in this field, we carried out a series of theoretical and experimental studies including the physics of plasmonic fields and the multi-photon and tunneling processes taking place.

Experimental research. – To gain experimental insight into the sub-cycle dynamics of multi-photon-induced photoelectron emission femtosecond time-resolved measurements were carried out on a polycrystalline gold surface with pulses of a Ti:sapphire laser to explain the recently found, unexpectedly low carrier-envelope phase dependence of the photoemission process in this particular case [1]. In the higher-order interferometric autocorrelation distribution additional short side wings appeared suggesting that ultrafast dynamics of hot electrons reduce the carrier-envelope phase dependence of the photoemission electron yield produced by few-cycle laser pulses [1]. Other metals can be investigated with this simple and fast method to pave the way towards the construction of a solid-state-based direct carrier-envelope phase detector.

We also contributed to Ti:sapphire oscillator development efforts to push the limits of the energy of pulses directly coming from a Ti:sapphire oscillator. As a result, in the framework of a collaboration with the Max Planck Institute for Quantum Optics (Garching, Germany) pulse compression of so-called chirped-pulse oscillators was carried out together with pulse characterization [2,3].

As a result of this collaboration one of us (P. Dombi) set up a so-called long-cavity Ti:sapphire oscillator that delivers femtosecond pulses with 250 nJ and 3 MHz repetition rate in Budapest [4,5]. This unique light source delivers ideal driving pulses for several applications, the above mentioned ultrafast electron diffraction being just one of them. These types of high-energy, high repetition rate ultrafast lasers also pose some laser technological challenges (e.g. how to compress their to below 10 fs) which were also investigated. During these tests a new type of backscattering mechanism from optical fibres was found that seems

to be unique to this parameter regime. We published several experimental results related to these experiments in femtosecond laser pulse generation and compression [4,5]. In addition, we have upgraded the pump laser of the existing novel laser oscillator delivering 200 nJ pulses to a commercial solid-state system. A pair of novel transmission gratings was inserted as the pulse compression stage for this oscillator. P. Dombi has spent 10 months at the Vienna University of Technology carrying out the development of an optical parametric chirped pulse amplification laser system. We also did preparations for setting up a simple vacuum chamber combined with an electron spectrometer that will be able to characterize surface-plasmon-enhanced photoelectron emission.

Surface plasmons (SPO) have been in the centre of our interest for many years. Our recent experiments concentrated on their behavior in extremely high laser fields, produced by short pulsed (2ps, 120 fs) titanium-sapphire lasers. The laser pulses excited the SPOs in resonance conditions and the light emitted by them was analyzed, both its angular distribution and spectrum [6,7]. Second harmonic (at 395nm) and around it a broad fluorescent spectrum was found, as seen in the figure. The former one is p-polarized as the exiting light, while the latter one is depolarized. The real origin of this broad spectrum, similarly as the distorted light spot in reflection, occurring at the same laser power density where this light occurs is under further investigation.

Theoretical research. – We elaborated a new approach to generation of ultrashort (attosecond duration) pulses based on the Sommerfeld precursor phenomena [8]. The numerical method for calculation of spectra of the first (Sommerfeld) precursor in a single-resonance Lorentz media based on representation of the initial signal as a sum of a number of semi-infinite pulses is developed for an arbitrarily shaped amplitude-modulated signal. Obtained spectra of the precursor generated by the signal created by “plasma mirror” technique show the potential possibility of production of electromagnetic pulses in attosecond regime.

On the basis of an exactly solvable model of classical electrodynamics we have determined the high-harmonic spectrum of a few-cycle Ti: sapphire laser radiation scattered on a plasma layer [9]. The spectra obtained qualitatively differ from each other depending on the carrier-envelope phase difference of the incoming pulse. We have generalized the calculation to relativistic intensities, and, in addition, we have included the effect of a homogeneous external magnetic field. The spectrum of the magneto-Raman scattering can in principle be used to measure the mentioned absolute phase.

We also proposed a novel way of tailoring electron pulses with the help of ultrashort, carrier-envelope phase controlled laser pulses and surface plasmons [10]. The new scheme allows for controlling photoelectron emission from metals surfaces spatially, spectrally and temporally thus providing a unique tool for time-resolved pump-probe-type studies with an electron beam. These developments can have significant impact on the currently developed ultrafast electron diffraction method which holds promise of uniting atomic resolution in space and attosecond resolution in time in material science. We are also investigating new ways of generating attosecond pulses with surface harmonics which has the advantage that much less energetic femtosecond pulses can be converted to attosecond oscillation than before.

We have analysed the reflection and transmission of a few-cycle femtosecond Ti:Sa laser pulse impinging on a metal nano-layer [11]. It has been shown that in general a non-oscillatory frozen-in wake-field appears following the main pulse with an exponential decay

and with a definite sign of the electric field. The scattering of a laser pulse impinging on a thin plasma layer has also been analysed at relativistic intensities [9]. The nonlinearities originating from the relativistic kinematics of the electrons lead to the appearance of higher-order harmonics in the scattered spectra. In this system we have found significant carrier-envelope phase difference effects. In the frame of our investigations concerning the physics of surface plasmon polaritons, we have worked out a new model which confirms the existence of enhanced electromagnetic fields bound to a thin metal layer evaporated on a glass substrate. Our latest result in attophysics has been to show that the above-threshold electron de Broglie waves, generated by an intense laser pulse at a metal surface, are interfering to yield attosecond electron pulses [12]. Owing to the inherent kinematic dispersion, the propagation of attosecond de Broglie waves in vacuum is very different from that of attosecond light pulses, which propagate without changing shape. The clean attosecond structure of the current at the immediate vicinity of the metal surface is largely degraded due to the propagation, but it partially recovers at certain distances from the surface. Accordingly, above the metal surface, there exist “collaps bands”, where the electron current is erratic or noise-like, and there exist “revival layers”, where the electron current consist of ultrashort pulses of attosecond duration. The attosecond structure of the electron photocurrent can, perhaps be used for monitoring ultrafast relaxation processes in single atoms or in condensed matter.

We have worked out the theory of high-density black-body radiation on the basis of the new concept of binary photons which satisfy the exclusion principle, thus they behave like fermions [13]. We have derived explicit expressions for the Wigner function of wave functions in arbitrary high D dimensions which depend on the hyperradius – that is, of s waves [14,15]. Due to the constraint on the dependence of the s wave on the coordinates, s waves describe entangled quantum systems. Since an isotropic Bose-Einstein condensate is described by an s wave, the Wigner function provides a deeper insight into the physics of these states and, in particular, correlations between position and momentum spanning quantum phase space. In the context of quantum optics, we have analysed the correlations of detection events in two photodetectors placed at the opposite sides of a beam splitter in the frame of classical probability theory. It has been assumed that there is always only one photon present in the measuring apparatus during one elementary experiment. It is explicitly shown in several examples that the bunching and anti-bunching of the counts in serieses of elementary single-photon experiments is governed by the statistical properties of grouping the sequences of the elementary measurements.

In case of the interaction of short and even longer laser pulses with material, the high temperature production in the target electron cloud can be significant. This high intensity radiation can be decomposed to fermionic components as shown in Refs [13, 16, 17]. The behaviour of these fermionic components is strongly separated and shows extremely short (attosecond) structures. The detailed study of these phenomena is a future task.

We have also dealt with the famous Ádám-Jánossy-Varga experiment in quantum optics [18]. Here we have calculated most conclusions with classical probability theory, without the quantization of the electromagnetic field, in accordance with the experimental results.

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