Ultrafast Nano-optical Interactions of Femtosecond Near-Infrared Laser Pulses with Solids

PhD thesis summary

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In the first part of my thesis, I give a short introduction into the basics of nonlinear optics, then present the fundamentals of surface and localized plasmon polaritons. I also summarize the most important works previously carried out in the fields involved in my research.

In the main part of my thesis, I present my research results. In the first subsection, I investigated laser-induced periodic surface structures (LIPSS) formation on conductive indium-tin-oxide (ITO) thin films, which I generated by femtosecond near infrared (NIR) laser pulse illumination at three distinct wavelengths (1,6 µm, 2,0 µm and 2,4 µm). I present the experimental setup that I designed, with which I observed high spatial frequency LIPSS (HSFL), as well as low spatial frequency LIPSS (LSFL). The experimentally produced HSFL structures appeared with a period of $\lambda/10$ relative to the incident laser wavelength. The results of finite-difference time-domain (FDTD) simulations indicated that the initial surface roughness of the ITO thin film and the consequent localization of the laser field lead to the formation of the observed periodic structures [T1].

In the second subsection, I present a novel experimental method for maximizing the field enhancement of plasmonic nanoparticles based on the polarization tuning of the incident laser pulses. The basic idea of this work is the control of the plasmonic field enhancement by exploiting the interference of the plasmon modes excited on the nanoparticle. I describe the measurement setup designed for the experimental validation of the method realizing the interference control of the plasmonic modes by tuning the polarization of the incident laser pulses. Using three different nanorod arrays and three different laser polarizations, I measured the local plasmonic field enhancements using ultrafast photoemission. The results clearly proved that this method is suitable for the in-situ, dynamic control of plasmonic field enhancement, thus enabling the dynamic resonance tuning of plasmonic nanostructures [T2].

In the third subsection, I present new phenomena of photoemission induced by ultrashort laser pulses in the vicinity of plasmonic nanostructures. Our preliminary expectation was that the contribution of the rescattered electrons to the measured electron spectra is significant even at higher Keldis parameter values above 3 in the multi-photon-induced emission regime. To support the theoretical findings, I performed ultrafast photoemission experiments on a sample containing plasmonic nanorods. Evaluation of electron spectra detected by the time-of-flight spectrometer revealed scaling of cutoff electron energies with $10U_p$ (where U_p denotes the ponderomotive energy – the cycle-averaged energy of an electron oscillating in an electromagnetic field) even at lower intensities, indicating the presence of electron rescattering processes in the multiphoton emission regime, even at relatively high Keldysh parameter values between 5 and 6 [T3].