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

Ph.D. Thesis Booklet

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Motivation and previous research

Ultrafast nano-optics has witnessed significant advancements in recent years, driven by innovations in femtosecond laser technology, nanofabrication techniques, and the growing demand for precise control of near-fields at the nanoscale. Developments of the recent decade proved that this field has both the potential to uncover fundamental aspects of ultrafast light-matter interactions [1–4] and the possibility for applications in a wide range of fields from micro- and nano-engineering [5], plasmonic sensing [6], to plasmonically enhanced photocatalysis [7], and next-generation on-chip optoelectronic devices [8].

One of the long-established branches of light-matter interactions research is that of laser-induced periodic surface structures (LIPSS). These structures have raised interest due to their unique collective optical and mechanical properties that can be tailored for various applications, including surface functionalization [9], anti-reflective coatings [10–12], and sub-wavelength structures for the semiconductor industry [13,14] and for microfluidic biomedical devices [15,16]. These applications rely on the physical parameters of these structures, such as their size, periodicity, depth, and uniformity. The precise and complete control and these parameters along with uncovering their formation mechanisms, remains a challenge and is a topic for discussions at international conferences.

Another intensely researched area within nano-optics is the properties of plasmonic nanoparticles, as they can enhance numerous nano-optical phenomena. By exploiting plasmon resonance, it is possible to concentrate electromagnetic fields into sub-wavelength spatial domains [17], leading to increased absorption and scattering [18,19], as well as local field enhancement [20]. This plasmonic field enhancement plays a crucial role in various applications, such as surface-enhanced Raman spectroscopy (SERS) [21], single-molecule detection [22], and the development of more sensitive chemical and biological sensors [23]. Integration of plasmonic nanostructures into photonic devices light sensors [24] and photovoltaic cells [25,26] can be realized, and their efficiency and performance can be increased.

Traditional methods aimed at achieving higher plasmonic field enhancement – and realizing resonance with the optical spectrum of the exciting laser – have been based on tuning the geometry of nanoparticles, where resonance is achieved by adjusting the size of nanoparticles parallel to the laser polarization for a given laser wavelength [27–29]. The general limitations of this method (fixed properties of the fabricated nanostructures) and the need for in-situ, dynamically tunable optical circuit elements demand new, innovative solutions that may be suitable for developing future next-generation optoelectronic devices. An excellent non-destructive, ultrafast and robust tool for investigating and measuring (and thus optimizing) the field enhancement appearing at the surface of plasmonic nanoparticles is ultrafast surface photoemission, which has previously been investigated for numerous different nanoparticles [30–34].

In addition, the phenomenon of ultrafast photoionization and photoemission is of significant interest in its own right. The ionization of atoms and molecules by laser light has revolutionized our understanding of ultrafast electron dynamics, contributing to unparalleled temporal resolution of electron motion while laying the foundation for numerous research areas, including high-order harmonic generation [35–37], which ultimately led to attosecond science [38–40] and the 2023 Nobel Prize in Physics.

From the 1970s to the 2000s, the research focus was primarily on photoionization processes. However, over the past decade, interest has shifted to the analogous process in solids, the investigation of ultrafast photoemission from nanostructures (nanotips, nanoparticles, plasmonic dimers, etc.). Using these intriguing nanostructures, numerous fundamental ultrafast photoemission processes have been uncovered, such as ponderomotive and intra-optical cycle electron acceleration [41–43], photo-assisted tunneling [44], and carrier-envelope phase effects [45], as well as enabling in-situ studies of exotic quantum materials [46]. Despite the pioneering results of these studies, there remain many interesting challenges in the field of ultrafast photoemission

from plasmonic nanoparticles, and a more complete understanding of the ultrafast dynamics of photoemitted electrons in nano-optical near-fields could contribute to the development of next-generation ultrafast electron sources [47] and secondary radiation sources.

Research goals

The aim of my PhD research is to investigate ultrafast nano-optical interactions using near-infrared laser pulses by employing conductive thin films, various surface nanostructures, and femtosecond laser sources. During these studies, I have conducted investigations across several related areas, including femtosecond laser-induced periodic surface structures, tuning plasmonic field enhancement of nanoparticles, and ultrafast photoemission from nanoparticles:

- LIPSS on indium-tin-oxide (ITO) thin films: Using near-infrared femtosecond pulses on ITO thin films, my goal was to generate periodic surface structures and study their physical properties. Accordingly, using femtosecond pulses with varying wavelengths and peak intensities, I have conducted the creation of different surface morphologies and investigated the fundamental processes responsible for their formation.

- Polarization control of plasmonic field enhancement: My goal was to experimentally validate a method for the control of local field enhancement on plasmonic nanoparticles by exploiting the interference of plasmonic modes. By demonstrating the effectiveness of the method with nanoparticles of various sizes, new opportunities arise for the implementation of diverse, dynamically tunable devices, such as ultrafast nano-optical switches.

- Ultrafast photoemission in the multi-photon-induced emission regime: Using resonant plasmonic nanoparticle arrays combined with femtosecond near-infrared pulses, my goal was to study the rescattering processes of photoemitted electrons in the low-intensity, multi-photon-induced emission regime. By using pulses centered at 3 different infrared wavelengths and a time-of-flight spectrometer, the kinetic energy spectra of electrons can be measured with sufficiently high resolution. This enables detailed investigation of ultrafast photoemission processes, including ponderomotive electron acceleration occurring in the vicinity of nanoparticle surfaces.

New scientific results, thesis items

I. I generated laser-induced periodic surface structures (LIPSS) on indium-tin-oxide thin films with femtosecond, near-infrared laser pulses. With central wavelengths between 1.6 and 2.4 µm, I observed LIPSS morphologies with periodicities close to $\lambda/10$ scaling. By inducing inhomogeneous local material ablation, I identified different morphologies generated on the thin-film surface and determined the corresponding threshold intensity values for their formation [T1].

II. I experimentally demonstrated a novel approach for the in-situ manipulation of field enhancement on plasmonic nanoparticles. Using three different resonant nanoparticles and three different incident laser polarizations, I measured the maximum near-field enhancement values for each configuration via a technique based on ultrafast photoemission. I showed that by solely altering the polarization of the incident laser pulses, phase-dependent mixing of the plasmon modes can be realized. This can result in significantly increased near-fields, which can lead up to 50% higher field enhancements compared to the traditional geometry-based resonance tuning [T2].

III. I experimentally demonstrated the transition between multi-photon-induced and strong-field emission regimes using near-infrared femtosecond pulses and resonant plasmonic nanoparticles. Employing this method, I proved that in case of ultrafast photoemission processes – contrary of previous observations – rescattering of emitted electrons is present in the multi-photon-induced emission regime and is not limited to the strong-field regime [T3].

The results of my research in the field of ultrafast nanooptics contribute to the comprehensive understanding of ultrafast interactions occurring on the nanoscale across numerous related research areas, ranging from laser surface processing to controlling plasmonic field enhancement at different surfaces, and extending to the fundamental investigation of ultrafast interaction phenomena.

Publications corresponding to thesis items

T1 B. Bánhegyi, L. Péter, P. Dombi, and Zs. Pápa, "Femtosecond LIPSS on indium-tin-oxide thin films at IR wavelengths", Appl. Opt. **61**, 386–391 (2022). https://doi.org/10.1364/AO.444653

T2 B. Bánhegyi, L. Tóth, P. Dombi, J. Budai, V. Hanus, P. Rácz, and Zs. Pápa, "Controlling Plasmonic Field Enhancement via the Interference of Orthogonal Plasmonic Modes", Plasmonics, online publikált cikk (2024). https://doi.org/10.1007/s11468-024-02212-9

T3 B. Bánhegyi, G. Zs. Kiss, Zs. Pápa, P. Sándor, L. Tóth, L. Péter, P. Rácz and P. Dombi, „Nanoplasmonic photoelectron rescattering in the multi-photon-induced emission regime", Phys. Rev. Lett. **133**, 033801 (2024). https://doi.org/10.1103/PhysRevLett.133.033801

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