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Ultrafast Nanooptics Group

“Lendület” Research Group

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Towards a plasmonic switch by exploring and exploiting hot electron dynamics — In future computing architectures, high-bandwidth optical and optoelectronic circuitry have been envisioned to carry and process information. The optical chip also needs to be compact, and it is a requirement that traditional optical elements cannot fulfil; surface plasmon polaritons however, are well suited since their characteristic size can be tailored through nanofabrication in the tens to hundreds of nanometers regime. Our work [1], published in Nano Letters, presents a simple design for all-optical switching through the generation of non-thermal electron population in gold. Using a novel optical pump/probe scheme (see Figure 1.), we probed ultrafast interband and intraband dynamics with 15 nm interface selectivity, observing a two-component-decay of hot electron populations. Experimental results are in good agreement with a three-temperature model of the metal; thus, we could attribute the fast (~ 100 fs) decay to the thermalization of hot electrons and the slow (picosecond) decay to electron–lattice thermalization. Moreover, we could modulate the transmission of our plasmonic channel with $\sim 40\%$ depth, hinting at the possibility of ultrafast information processing applications with plasmonic signals.

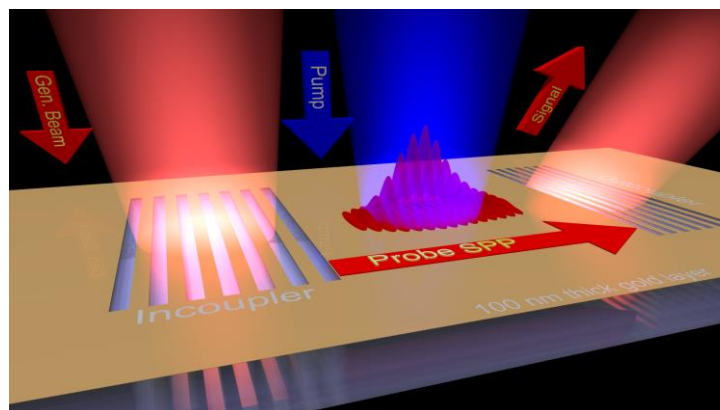


Figure 1. Illustration of the pump–probe concept with the generation and outcoupling of SPPs using grating couplers milled into the 100 nm gold layer. Nonthermal electron distribution is generated in between the grating couplers by a separate, time-delayed pump pulse (shown in blue color).

Ultrafast plasmonics - In strong-field laser-matter interactions, energetic electrons are usually generated through photoemission followed by rescattering, reaching energies up to ten times the ponderomotive potential (U_p) of the laser field. In 2024, in our work [2] published in Physical Review Letters, we demonstrated that combining infrared laser sources with plasmonic nanoemitters, the observation of $\sim 10U_p$ rescattered electrons in the multiphoton-induced is possible (see Figure 2). The advantage of this combination is twofold: on the one

hand, using infrared femtosecond pulses we can take advantage of the quadratic scaling of U_p , thus measuring higher kinetic energy electrons at lower peak intensities. On the other hand, exploiting plasmonic resonance on gold nanorods enhanced the rescattering probability by orders of magnitude. The experimental results aligned closely with a model based on the time-dependent Schrödinger equation, uncovering an unexpected aspect of ultrafast electron dynamics in the multiphoton-induced emission regime. Furthermore in collaboration with group at the Graz University of Technology led by Martin Schultze, we have demonstrated the existence of few-cycle plasmon oscillations at tapered plasmonic waveguide with an optimized grating structure using time-resolved photoelectron microscopy methods [3]

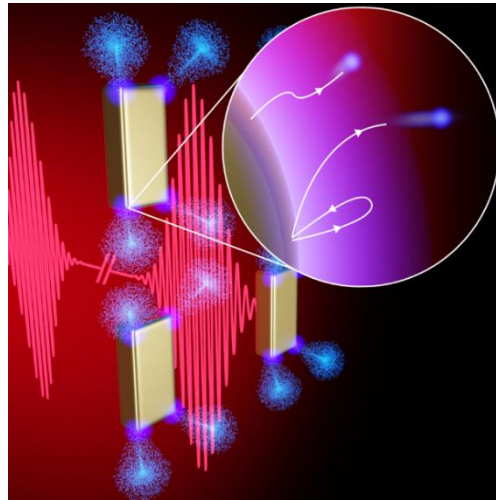


Figure 2. Plasmonic resonance excited by infrared femtosecond pulses. Inset shows the ultrafast electron dynamics in the vicinity of plasmonic fields at the hot-spots.

Nanoplasmonic field engineering - Optical nanoantennas concentrate light into their local fields, thus confining plasmonic resonances with exceptionally high local field intensities on their surface. In our paper [4] published in *Plasmonics* in 2024, we introduced a method for controlling the local-field interferences in the hot spots of nanorods and experimentally demonstrated that the hot-spot field enhancement can be tuned over a wide range. Plasmonic gold nanoparticles were designed with specific phase relations between their orthogonal (longitudinal and transverse) plasmonic eigenmodes. The phase between the components of the external field was adjusted by altering its polarization state of the exciting femtosecond pulses to achieve in-phase excitation of the plasmon modes. To probe the field enhancement properties of the nanorods, we implemented a robust measurement method based on strong-field photoemission using laser pulses with different polarization states. These findings unveiled a new degree of freedom in plasmonic resonance tuning, potentially sparking diverse designs of local-field responses and broadening applications in nanoscale sensing, spectroscopy, and dynamically tunable devices.

References

- [1] <https://pubs.acs.org/doi/10.1021/acs.nanolett.4c01669>
- [2] <https://doi.org/10.1103/PhysRevLett.133.033801>
- [3] <https://doi.org/10.1021/acs.nanolett.3c04991>
- [4] <https://doi.org/10.1007/s11468-024-02212-9>