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SFB 1242

Time-Resolved Plasmoemission Spectroscopy

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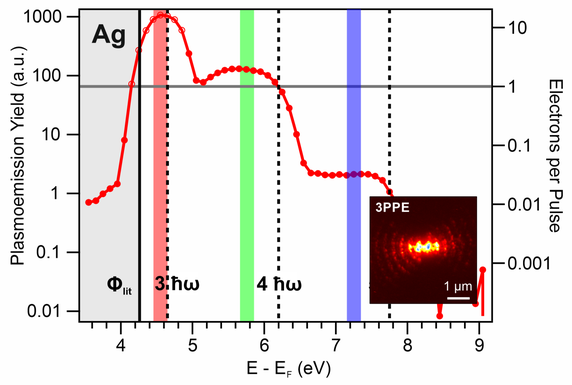
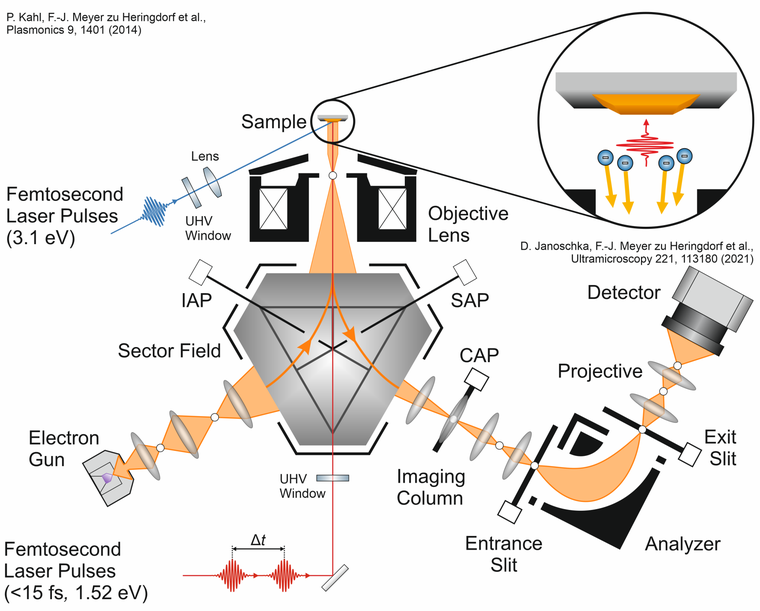
Motivation

- The strong electrical near-field of SPPs in nano-structures can drive nonlinear optical processes at flat metal surfaces.
- Electrons can be liberated from the surface in the absence of light by the absorption of multiple SPP quanta. (above-threshold plasmoemission)
  D. Podbiel, H. Giessen, F.-J. Meyer zu Heringdorf et al., Nano Lett. 17 (11), 6569 (2017)
- Here, we investigate the electron emission process in the presence of strong plasmonic nano-foci on flat gold surfaces.



Experimental Setup

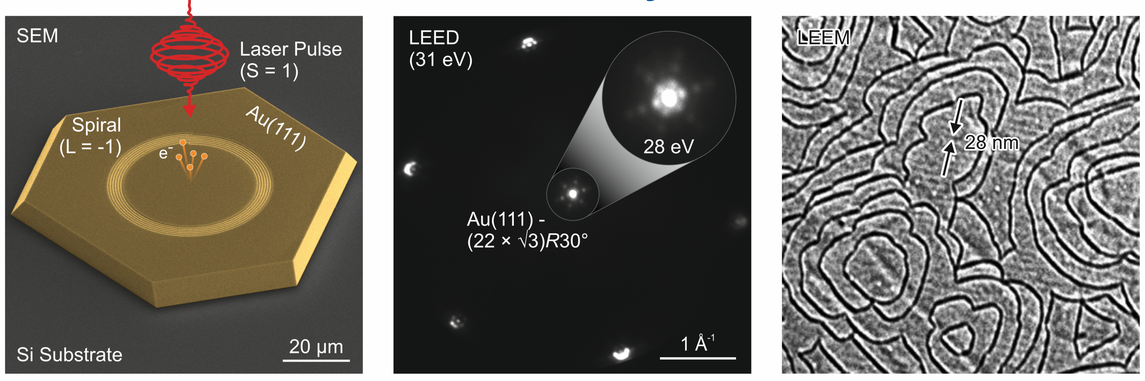
Photoelectron Spectro-Microscopy



- Elmitec SPELEEM for imaging, momentum microscopy, and spectroscopy.
- Multiple appertures provide for filtering of electrons by their real space origin, momentum and energy.
- ~10 nm spatial resolution and <200 meV energy resolution.
- Ti:Sapphire oscillator with ≤15 fs pulse duration and 800 mW average power at 800 nm central wavelength.
- Pump-probe experiments using Mach-Zehnder Interferometer or birefringent delay line.

Nanofocusing of Surface Plasmon Polaritons

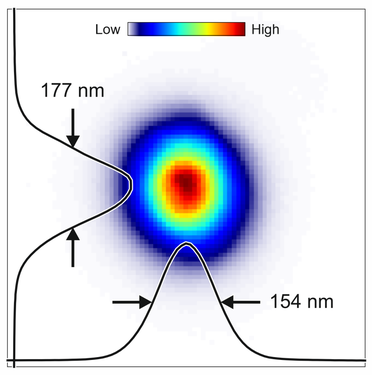
Plasmonic Structures on Atomically Flat Gold Surfaces



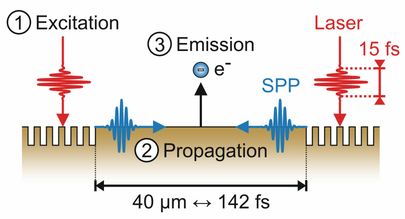
- Single-crystalline gold flakes grown by thermolysis.
  Thermolysis: Radha et. al., Nano Res. 3, 738 (2010)
- Fabrication of plasmonic nanostructures by *ex-situ* focused ion beam milling.
- Herringbone reconstruction of Au(111) surface is visible in *in-situ* microprobe low energy electron diffraction on individual flakes.
- Individual atomic steps and Herringbone reconstruction of atomically-flat flakes are visible in low energy electron microscopy.

Plasmonic Nano-Focusing in Archimedean Spirals

- Illumination of Archimedean spiral with $L = 1$ topological charge by $S = -1$ laser light excites $J = 0$ SPP state.
- Deep sub-wavelength electron emission hotspot due to nonlinearity of emission process.
- Plasmoemission only from the out-of-plane field.

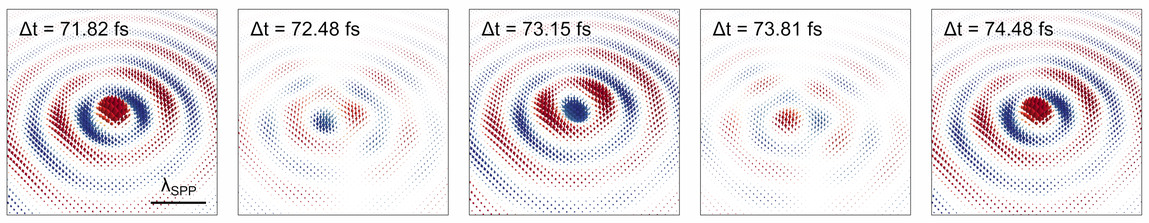


- Spatio-temporal separation of plasmonic and optical contributions to the electron emission yield by structural design.



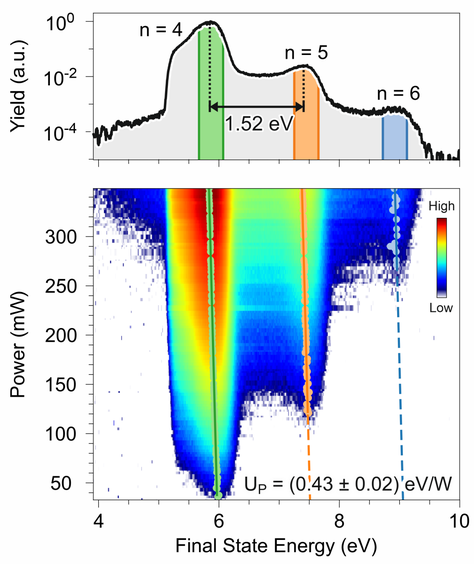
Vector-Polarimetry of Plasmonic Nanofocus

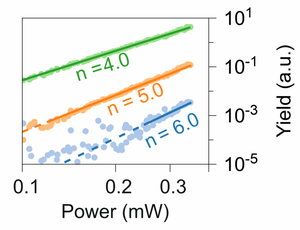
- $J = 0$ SPP vortex state corresponds to a Bessel-type SPP mode with an exclusively out-of-plane field maximum at the center.
- SPP field amplitude performs one full oscillation within an optical cycle without changes of the local field polarization.



Nonlinear Plasmoemission Spectroscopy

Ponderomotive Shifts in Multi-Plasmon-Plasmoemission

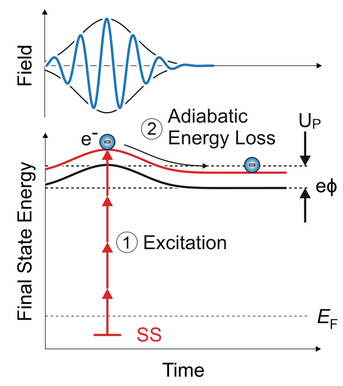




- Keldysh Parameter: $\gamma \sim 4.4$
- Adiabaticity: $\delta \sim 4700$
- Max. SPP Field: $E \sim 4.3$ V/nm
- Max. SPP intensity: $I \sim 2.4 \times 10^{12}$ W/cm^2

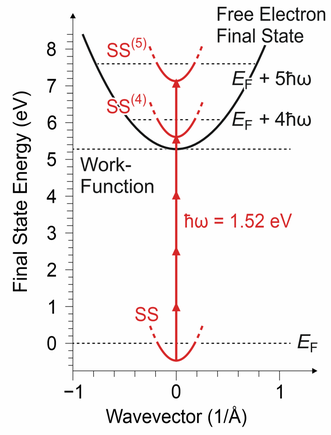
P. Dreher, F.-J. Meyer zu Heringdorf et al., Nanophotonics 11, 3687 (2022)

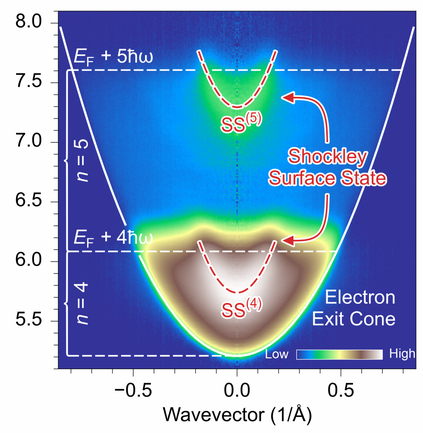
- Energetic shift given by ponderomotive interaction in the focus.



Angle-Resolved Plasmoemission Spectroscopy (ARPLES)

- Angular distribution of the emitted electrons is limited by the free-electron exit cone.
- Above-threshold emission: electron emission maxima can be found at multiples of the SPPs' energy.
- Dispersion of the Au(111) Shockley surface state (SS) is clearly visible in the angle-resolved spectrum.





Au(111): Coherent SPP-Electron Coupling

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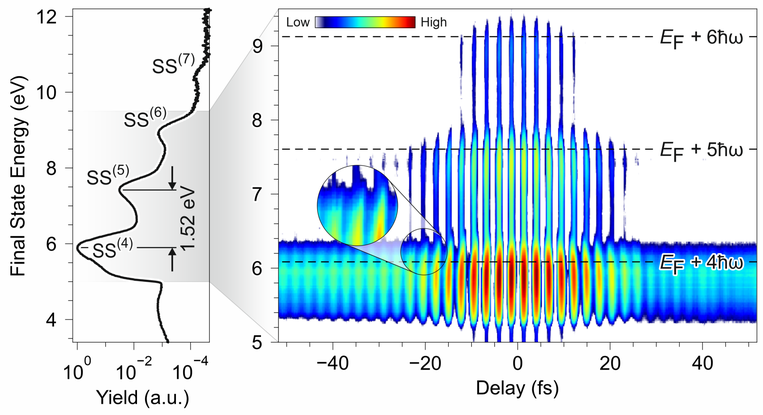
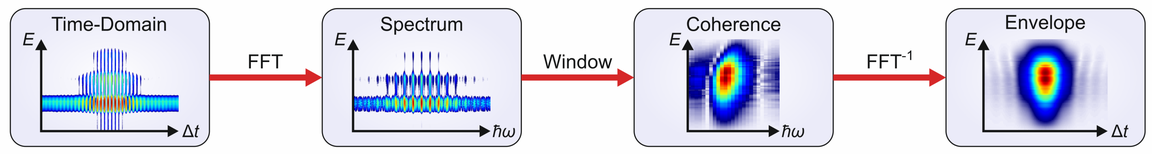
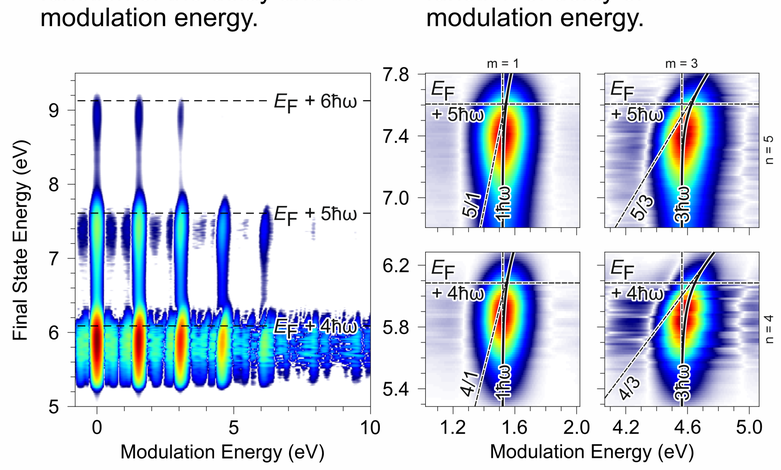


- Coherent two-dimensional photoelectron spectroscopy gives access to coherences in the electron emission process.
  M. Reutzel et al., Phys. Rev. X 9, 011044 (2019)
- Coherent emission: fringes tilt as a function of the final state energy and the pump-probe delay.
- Incoherent emission: no tilts observable.



- Correlations between the final state energy, emission order, and the modulation energy appear as coherence peaks in the Fourier transform of the interferogram.
- The slope of coherent emission peaks is given by n / m for the emission order n and the modulation harmonic m.
- Peak tilts have the expected slope for final state energies above the SS band minimum.
- The coupling of the SPPs to the SS during nonlinear plasmoemission is coherent.

- Coherent emission: fringes and peaks tilt as a function of the delay and the modulation energy.
- Incoherent emission: no tilts observable as a function of delay or modulation energy.



Conclusions

- Above-threshold-plasmoemission up to order $n = 7$ is observed from a single plasmonic nano-focus.
- The ponderomotive energy was used to measure the peak intensity in the SPP focus.
- The Au(111) Shockley surface state is visible in the above-threshold and angle-resolved plasmoemission spectra, demonstrating the direct coupling of SPPs to electronic states in the bandstructure of a solid.
- Time-resolved plasmoemission spectroscopy shows that the coupling of SPPs to the electron system is a coherent process.
- Our results enable a photoemission spectroscopic approach to sudying the interaction of strong nano-focused SPPs with the electroninc system of a solid.