Diving into the 3D plasmonic near field: high-energy electron-light-matter interactions

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High-energy electrons in a cathodoluminescence scanning electron microscope perform 3D plasmon near-field tomography and holography, demonstrate a cylindrical plasmonic metalens and climb quantum ladders in extreme plasmonic near fields.

Control over the nanoscale distribution of light fields in the optical spectral range is of great importance in many technological fields, including photovoltaics, photocatalysis, and molecular sensing. A unique way to study optical near fields at very high spatial resolution is by cathodoluminescence (CL) spectroscopy in which a high-energy (1-30 keV) electron polarizes a plasmonic material and the emitted optical radiation is analysed [1]. The spatial distribution of the CL signal measured in a scanning electron microscope (SEM) provides a two-dimensional (x,y) map of the plasmonic density of states at nanoscale spatial resolution.

3D plasmon tomography

In this work, we use CL spectroscopy to dive into the third dimension, to reconstruct the near field in the z-direction. We perform CL spectroscopy at multiple electron energies and take advantage of the fact that the electron-plasmon coupling strength is proportional to the Fourier transform of the near field at a spatial frequency determined by the electron velocity. We image near field of ultrasmall (20-100 nm) gold colloids, both in aloof geometry and under normal incidence, and introduce a recoil model that takes into account the electron energy loss inside the plasmonic particle.

Plasmon holography

Next, we study angle-resolved CL emission from plasmonic Au nanocubes and nanoholes in a Au film that are excited by electron-beam generated surface plasmon polaritons. We develop a holography geometry in which transition radiation generated by the electron beam serves as a reference. From the angle- and spectrally-resolved interference patterns we directly derive the phase fronts of the plasmonic scattering fields, and perform a full decomposition into the multipolar electric and magnetic radiation modes. We find that the strong UV spectral band that is often assigned to interband or intraband transitions is governed by coherent processes, where the light emission has a clear phase relation with the electron excitation.

Scraping along plasmon metasurfaces

Then, with collaborators from Technion, we excite plasmonic near fields along a Au metasurface using an electron beam that grazes along the surface. This creates an array of radiating plasmonic multipoles with well-defined phase evolution, determined by electron velocity and metasurface geometry, and creates radiation in the far field (Smith-Purcell radiation). By judicious design of the spatial profile of the metasurface geometry we tailor the angular and spectral distribution, creating a cylindrical metalens for plasmon radiation for wavelengths from the UV to the near-infrared. The electron beam serves as a direct probe of the phase evolution across the plasmonic metasurface.

Electrons climbing plasmonic quantum ladders

Finally, we will present work with our collaborators at the Univ. Göttingen, ICFO and Univ. Taragona to explore the Photon-Induced Near-field Electron Microscopy (PINEM) effect in ultrasmall plasmonic near fields. We create Au nanostars with ultrasharp tips (radius 3 nm) and create intense nanoscale plasmonic hot spots using 3 ps pulsed laser irradiation (1.55 eV, 1 GW/cm^2). This results in efficient electron-plasmon energy exchange, creating side bands in the electron energy spectrum spaced by the photon/plasmon energy, reflecting a plasmon-induced quantum superposition state of the electron. We reconstruct the plasmonic near-field distribution from the spatially-resolved side band spectra and correlate them to the CL maps. From the data we derive a full 3D reconstruction of the plasmonic near field.

References

[1] Electron-beam spectroscopy for nanophotonics, A. Polman, M. Kociak, and F.J. Garcia de Abajo, Nature Mater. **18,** 1158 (2019)