Plasmonic nanofocusing for broadband near-field spectroscopy

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The coherent coupling between dipole momenta via optical near fields determines the optical properties and function of many nanostructures [1]. These interactions extend only over a few nanometers with sensitive dependence on relative dipole orientation, spectral detuning and dephasing. This makes it challenging to analyze or control these interactions experimentally.

This colloquium discusses recent progress in near-field spectroscopy using plasmonic nanofocusing [2] as a tool to detect coherent dipole couplings with 5-nm spatial resolution [3]. The apex of a conical gold nanotaper serves as a small dipolar antenna probe, which is excited solely by the evanescent fields of surface plasmon polariton (SPP) wavepacktes nanofocused along the taper. We couple this antenna to plasmon resonances in a single gold nanorod and resolve the mode couplings, resonance energy shifts and Purcell effects of the system as a function of dipole distance and relative orientation. Our measurements reveal how these effects arise from different vectorial components of the interacting optical near-fields [3].

As an extension of this method, we show that the taper apex can not only excite the sample but also collect light by converting optical near-fields back into SPP waves propagating up the taper shaft [4]. Incident and back-propagating SPPs give rise to inherently phase stable interferograms from which we deduce amplitude and phase of the local optical near fields around individual nanoparticles [4]. These measurements directly provide broadband maps of the projected local density of optical states (LDOS) around the sample, making plasmonic nanofocusing spectroscopy a promising tool for the study and control of the optical properties of coupled hybrid nanosystems.

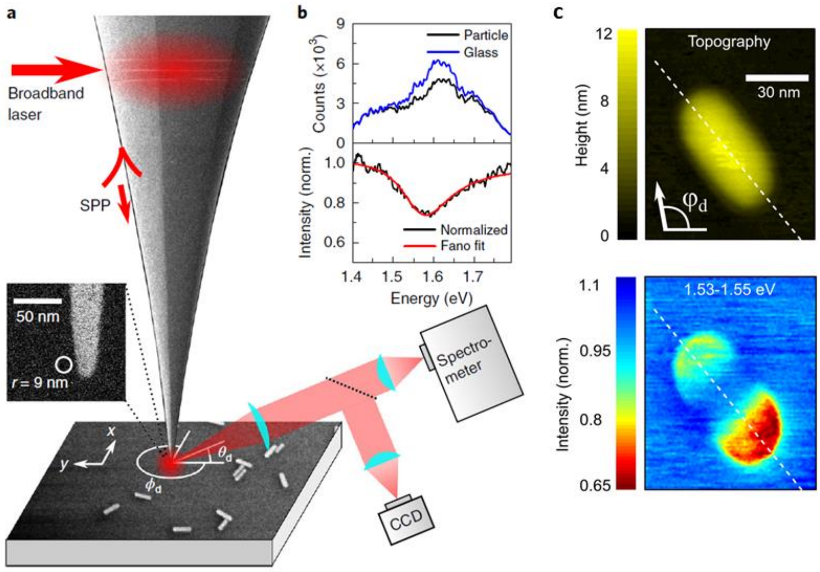

***Figure 1. Plasmonic nanofocusing spectroscopy of gold nanorods**. **a**, Surface plasmon polariton (SPP) waves are nanofocused to the apex of a gold taper serving as a local dipole antenna. **b**, (top) Spectra recorded with the nanofocusing source placed on the glass substrate (blue) and at the apex of a nanorod (black). (bottom) Normalized Spectrum (black) with a fit to a Fano lineshape (red). **c**, (top) Topography scan of a gold nanorod. (bottom) Spectra were recorded at each pixel during the scan. The false color image shows the normalized scattering intensity close to the nanorod resonance (1.54 eV).*

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[2] Stockman, M.I., PRL **93**, 137404 (2004).
[3] M. Esmann et al., Nature Nanotechnology **14**, 698 (2019).
[4] M. Esmann et al., Nanophotonics (**in print** 2019).