Inelastic Momentum Transfer Measurement of a Surface Plasmon Mode

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Surface plasmons are excitations of the electron gas inside a nanoparticle which, due to the spatial confinement of the particle, have a discrete spectrum. They are created by an external time-dependent field, which couples strongly via surface charges to resonant modes of the internal field, whose emanated field can reach much higher peak field strengths than the exciting field. This local field enhancement effect is a major motivation behind plasmonics research since it promises a great boost in the efficiency of photovoltaics and optical sensors. Furthermore, the development of plasmonics-based light transducers anticipate the tight integration of photonics into semiconductor circuitry. To reach such ambitious goals new analytical tools are needed for the characterisation of such transient phenomena at the nanoscale. Here, we present a method for mapping the components of the electrical field inside a single plasmon mode by means of energy-filtered low-angle electron nanodiffraction.

Our basic approach consists of scanning a nanometer size electron beam over a plasmonic particle, where an excited plasmons deflects the electron beam both laterally, creating a shift in a diffraction plane, and longitudinally, causing an energy loss. Accordingly, this inelastic momentum transfer (IMT) corresponds to the induced field integrated over the electron's trajectory. Our novel experimental setup enables measuring both the lateral and longitudinal mean momentum transfer at each scanning point by evaluating the sum and the center of mass (CoM) of the energy filtered electron beam. In conjunction with simulation studies this allows quantitative conclusions about the local field strength of the plasmon mode. Due to the small deflection angles (in the order of microradians) these experiments have to be done in a low-magnification STEM mode, while the low intensity in the energy-filtered beam demands a very stable monochromator and filter setup.

We recordend such energy-filtered diffraction patterns (Fig 1b) of a dipole mode at 1.05 eV (see spectrum in Fig 1c) of an aluminium nanorod (Fig. 1a) with a variation of the sum of the diffraction patterns (Fig 2a) and a shift the CoM (Fig 2c) near the particle. These can be related to spectral components of the induced field which can be simulated from known object geometries. We used the MNPBEM toolbox with an effective medium approach to account for the oxide layer and the silicon nitride substrate and achieved reasonable agreement between the experimental and simulated spectra (Fig. 1c), loss probability (Fig 2 a and b) as well as the induced lateral fields (Fig 2 c and d). We attribute the noticeably lower signal (in the experimental data) below the sample to a shift in the energy selection slit over the course of the experiment (we scanned row-wise from up to down). Another anomalous influence is the vignetting of the non-isoplanatic probe which we cannot correct locally in this approach.Our measurement of the IMT represents the first quantitative measurement of the transient lateral fields of surface plasmons. Theory and simulation indicate that these can be related much more meaningfully to the corresponding local field strengths than the longitudinal, i.e. the EELS, signal.

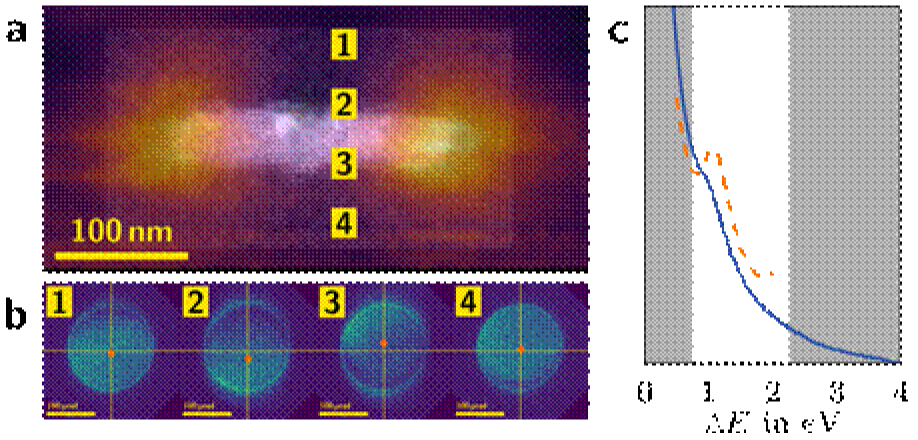

Fig 1: Overview of the experimental results on an aluminium nanorod. (a) HAADF scan (grayscale) overlayed with the local loss probability (color). (b) four energy-filtered (energy range indicated in c) diffraction patterns from positions marked in a. (c) EEL sp spectrum near the left cap with the energy slit indicated as white background and the simulated spectrum as a dashed line.

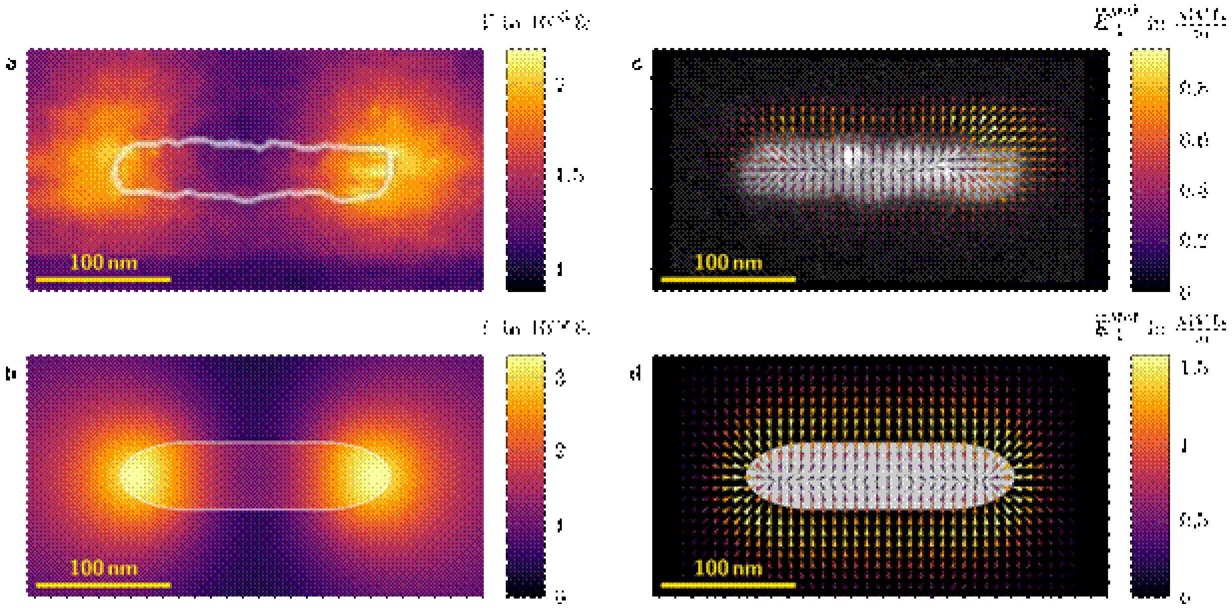

Fig 2: Comparison of loss-probability (a and b) and spectrally resolved lateral fields (c and d) from the experiment (upper row) and the simulation (lower row).