Exploring the inelastic interaction between phase-shaped electron beams and plasmonic resonances

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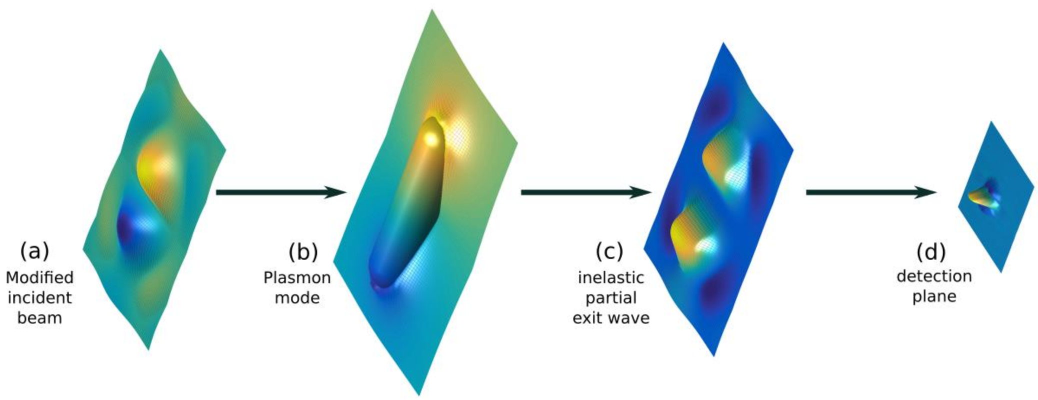
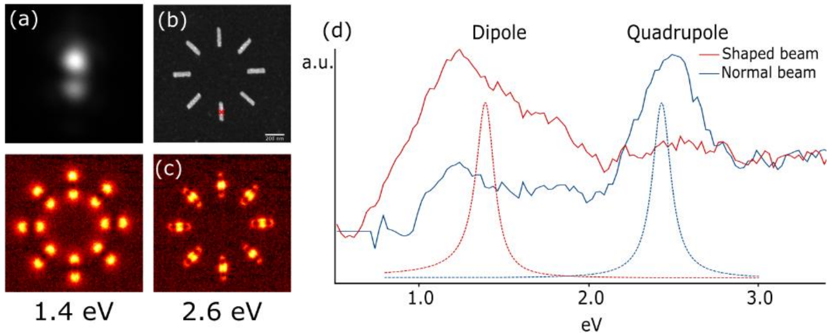
Electronic spectroscopies are important in the study of localised surface plasmon resonances (SPRs) of metallic nanostructures, allowing to detect and image the strong spatial variations in the electrical field of the induced resonances of a single nanoparticle. These techniques do however present some drawback when compared to their optical counterparts. While optical spectroscopies can make use of polarisation to directionally probe the response of a nanoparticle, an electronic beam can't discriminate between energy-degenerate eigenmodes and is also blind to optical activity and dichroism. New developments (e.g. PINEM) have reduced this gap, without however eliminating the root of the problem: electron spectroscopies probe the intensity of the plasmon's field, but are blind to its sign and direction.



Here we show how the electrical potential of a SPR inelastically couples to the in-plane phase modulation of the electron beam. We exploit this in a radically new approach based on the idea of controlling, through beam shaping methods [1], the wave function of the electronic probe. By fitting the probe to the plasmonic modes under investigation, it becomes possible to measure previously inaccessible properties. We theoretically describe this coupling, then design an experimental setup allowing to selectively detect localised plasmonic excitations that possess the same symmetry as the electron probe [2]. Figure 1 illustrates this schematically. A modified incident beam (fig1a) is created, possessing the same symmetry as the plasmonic mode of interest (fig1b). The signature of the combined symmetries is contained in the inelastically scattered wave (fig1c), and causes the presence of an on-axis intensity peak whenever the two symmetries match (fig1d). Post-selecting this feature with a spectrometer entrance aperture we can selectively detect only those excitations fulfilling this symmetry requirement. Finally, as shown in figure 2, we experimentally test this by selectively detecting the dipolar mode of a set of nanorods (fig2b) with a two-lobed probe (fig2a), which we successfully generate in a TEM by applying state of the art phase manipulation techniques. The recorded spectra (fig2d) clearly display the desired selectivity. This demonstrates the viability of this new approach and opens the way to a new generation of plasmon-oriented TEM experiments, which will progress in parallel with the development of phase manipulation methods.



[1] G. Guzzinati et al., Ultramicroscopy 151, 85 (2015).
[2] G. Guzzinati et al., Nature Communications, Accepted / Arxiv:1608.07449
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