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Low-loss EFTEM Imaging of Surface Plasmon Resonances in Ag Nanostructures

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Understanding how light interacts with matter at the nanometer scale is a fundamental issue in optoelectronics, nanophotonics and nanoplasmonics. The optical properties of metallic nanoparticles are entirely dependent on collective excitations of their valence electrons, known as surface plasmon resonances (SPR), under electromagnetic illumination. Measuring these properties locally at the level of the individual nanoobject in combination with spectral information over the entire visible range constitutes a challenging issue for linking of the global response of the nanoparticles and the underlying structure and morphology. The visualization of localized SPRs on the nanometer scale in combination with spectral information over the entire visible range is of prime importance in the field of biosensors, surface-enhanced Raman spectroscopy, and for the design of metamaterials. But also the explanation of abnormal transmission of light through sub-wavelength holes relies on such information.

The high-energy electron beam in a transmission electron microscope (TEM) is an excellent tool for probing the optical properties of metallic nanoparticles in the ultraviolet–near-infrared (UV–NIR) domain with nanometer resolution via SPR mapping by means of electron energy-loss spectroscopy (EELS) in the low-loss range. With the advent of recent instrumental improvements such as electron monochromators and highly dispersive in-column energy filters, energy-filtered TEM (EFTEM) has now become available for the study of the optical response in the UV-NIR regime of materials. This technique was applied to the detection of band gaps [1] as well as to the study of surface plasmons on metal particles, like Ag nanoprisms [2] or holes in an Ag thin film [3]. It offers a spatial resolution in the nanometer range which is well below the resolution of present light-optical techniques.

The experiments were carried out in the 200 kV FEG-TEM Sub-Electronvolt-Sub-Angstrom-Microscope (Zeiss SESAM) equipped with an electrostatic monochromator and the high-dispersion and high-transmissivity in-column MANDOLINE filter [4]. The superior properties of this instrument enable EFTEM imaging in the ultraviolet–near-infrared domain with very high energy resolution and spatial sampling. The EFTEM image series were acquired on a 2k × 2k CCD camera using a 0.2 eV energy selecting slit for both the monochromator and the imaging energy filter. Energy-filtered images were recorded from 0.4 eV to 4 eV with an acquisition time between 20 s and 30 s per image.

In this contribution, EFTEM imaging combining high spatial sampling and high energy resolution of the dielectric response of metallic nanoparticles, like triangular Ag nanoprisms (Fig. 1), and of holes with 180 nm diameter in a 100 nm thick Ag film (Fig. 2), drilled by using a focused ion beam, are presented. We map surface plasmon resonances (SPRs) at optical wavelengths on single triangular silver nanoprisms, where extra multipolar SPRs on these nanoparticles could be detected. The arrangement of the holes was chosen such that well separated holes, holes that are closely spaced, and interpenetrating holes were present. Depending on energy loss, we find a number of resonant features that can be ascribed to either resonances of single holes or coupled resonances of several holes. The coupling effects between adjacent holes lead to very strong field enhancements which occur primarily in the infrared range. It is expected that the understanding of these optical properties can be further improved through the gain in both energy and spatial resolution. These results demonstrate the power of the EFTEM technique for mapping of surface plasmon resonances of complex structures. [5]

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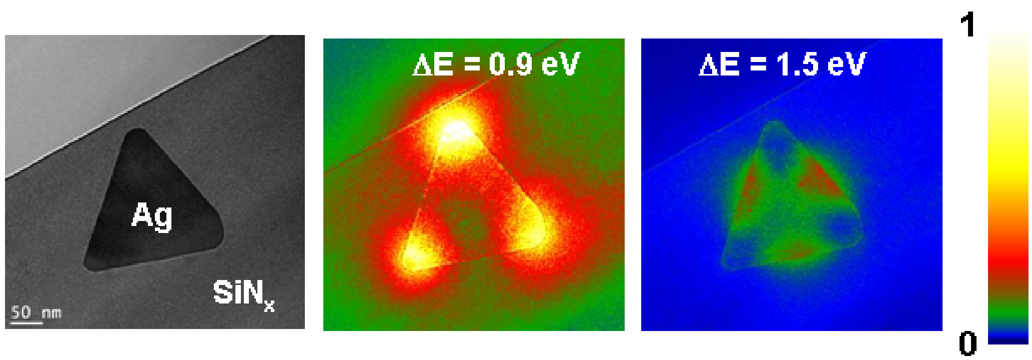

FIG. 1. Collage of three EFTEM images acquired on a triangular silver nanoprism at 0 eV, 0.9 eV and 1.5 eV energy losses. The intensity scale is common to the two images.

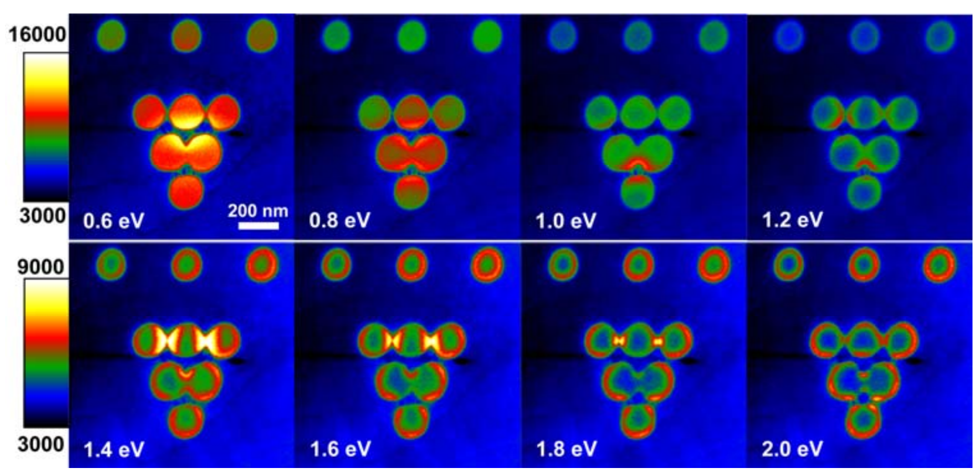

FIG. 2. Series of energy-filtered images of a hole arrangement in a thin Ag foil. A number of localized plasmon resonances can be identified. The images are displayed in false-color (increasing intensity: blue-green-red-yellow).

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