

Electron Energy Loss Spectroscopy on Lithographically Defined Photonic Metamaterials

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Abstract

We investigate the spatial and spectral distribution of plasmonic modes on different metamaterial building blocks via STEM-EELS. Our samples have been fabricated utilizing electron-beam lithography on TEM compatible substrates. Experimental data for the four lowest-order plasmonic modes of split-ring resonators and complementary split-ring resonators are presented.

1. Introduction

Localized plasmonic modes in metallic nanostructures are key ingredients for many photonic metamaterials. The corresponding electromagnetic near-field distributions and resonance frequencies can be controlled by a proper choice of the sample geometry and the constituent materials. In return, a thorough understanding of the optical properties of a given metallic nanostructure requires the knowledge of the electromagnetic near-field distribution for a range of photon energies. For this purpose, several efficient numerical calculation schemes have been developed which provide data for idealized structures. However, in order to test these calculations and in order to study the influence of structural imperfections, experiments on the spatial and energetic distribution of plasmonic modes are required.

The combination of scanning transmission electron microscopy (STEM) with a monochromatic electron beam and low loss electron energy loss spectroscopy (EELS) allows imaging of the plasmonic modes on metallic nanostructures [1-3] with a spatial resolution of a few nanometres. Here, we present experimental EELS maps of the four lowest-order plasmonic modes of split-ring resonators (SRRs) and complementary split-ring resonators (CSRRs).

2. Sample Fabrication and Electron Microscopy

A reliable and reproducible fabrication method is mandatory for systematic studies on photonic metamaterials. In this work, we employ electron beam lithography for this purpose. A 30 nm thin silicon nitride (Si_3N_4) membrane fixed on a silicon frame serves as an electron transparent substrate which was covered with 5 nm of indium tin oxide (ITO) providing the required electronic conductivity for electron beam lithography. The structures were defined by a computer controlled 30 kV electron beam in a 200 nm thick film of polymethyl methacrylate (PMMA). After developing the exposed PMMA film with a 1:3 mixture of methyl isobutyl ketone and isopropyl alcohol, we deposited a 2 nm thin layer of chromium followed by a 35 nm thin film of gold via electron-beam evaporation. In the final lift-off step the remaining PMMA was removed in warm acetone. The inset of Fig. 1 (a) depicts a high angular annular dark field (HAADF) image of a SRR fabricated by the procedure described above.

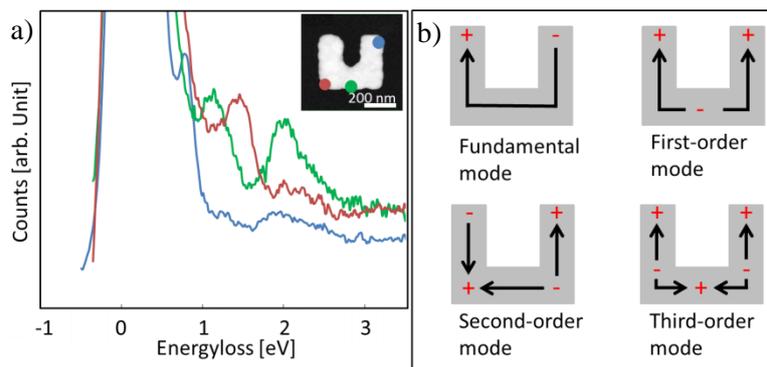


Fig. 1: (a) Three typical EEL spectra acquired at different locations on the SRR depicted in the inset. The beam locations are marked in the same colour as the respective spectra. (b) Scheme of the four lowest-order modes of a SRR. The plus and minus signs indicate antinodes of the charge density oscillations.

The STEM-EELS experiments were performed with a Zeiss Libra200 MC (CRISP) [4] operated at 200 kV. A monochromator enhances the energy resolution of the electron beam from typically 0.7 eV to < 0.2 eV. The instrument is equipped with a Cs-corrector for the illumination system. Spectra were recorded using the 90° in-column energy filter and a 2k x 2k SSCCD camera (Gatan, Ultrascan 1000). In STEM-mode, the system was operated with a beam convergence semiangle of 25 mrad and a collection semiangle of 7 mrad. The dispersion of the spectrometer was set to 0.016 eV/channel and the acquisition time for each spectrum was 2 s. The spatial resolution for our settings in the STEM-mode was in the order of 1-2 nm, which is much smaller than the pixel step width of 10 nm used in the EELS maps.

3. EELS on SRRs- and CSRRs

Fig.1 (a) depicts three typical EEL spectra of a SRR. The corresponding electron beam locations are color coded in the HAADF image of the SRR (see inset). Each spectrum has been normalized to the maximum value of its zero-loss peak (ZLP). Additionally, the maximum of the ZLP has been shifted to 0 eV. The energetic resolution (FWHM) in our experiments is 0.2 eV on the gold structures and 0.16 eV on the Si₃N₄-membranes. All three spectra exhibit pronounced resonances in the energy range from 0.8 eV to 2.5 eV which can be clearly separated from the ZLP without further data processing. These resonances correspond to different plasmonic modes of the SRR.

Classically, the energy loss experienced by an electron can be interpreted as the work done by the electron against the electric field of the excited modes. We expect a strong EELS signal for a given plasmon energy and electron beam position if the corresponding plasmonic mode has a large electric field component E_z along the trajectory (z-axis) of the electron. For planar metallic nanostructures like our SRR or CSRR, these positions correspond to the antinodes of the plasmonic charge density oscillation which usually provide a large E_z -component.

In a first approximation, a SRR can be considered as a straight antenna whose ends have been folded upwards. In this picture, the different SRR modes simply result from bending of the corresponding well-known antenna modes which are standing waves of the current density oscillation (see Fig. 1 (b)). For the fundamental mode, we find current density nodes at the ends and a current density antinode in the middle of the SRR. The corresponding charge density oscillation has antinodes at the two ends. The charge density oscillation of the first-order mode has an additional antinode in the middle of the SRR. Note that this mode cannot be excited in a conventional far-field optical experiment. From analogous considerations we can also deduce the locations of the charge density antinodes of the higher-order SRR modes.

Fig. 2 (b) – (e) depict experimental EELS maps of the four lowest-order plasmonic resonances of the SRR shown in Fig. 2 (a). As expected, for the fundamental resonance at 0.8 eV we find strong EELS

signals at the two ends of the SRR (see Fig. 2 (b)). The first-order mode at 1.1 eV exhibits a strong EELS maximum in the middle of the lower edge of the bottom wire as well as faint maxima at the ends of the SRR (see Fig. 2 (c)). The EELS signals at the lower corners stem from the tails of the second-order mode spectrally centred at 1.3 eV (compare with Fig. 2 (d)). Finally, the third-order mode at 2.0 eV features three EELS maxima distributed along the wire. Note, that we do not find the anticipated EELS maxima at the ends of the SRR for the second-order and third-order mode. We are currently working on exact numerical calculations to clarify this point.

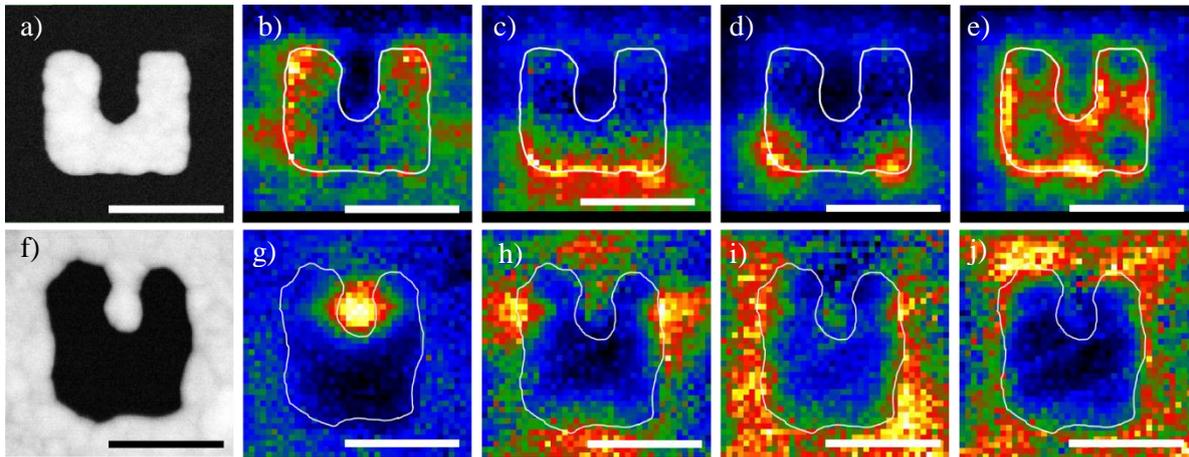


Fig. 2: (a) HAADF image of an SRR. (b) – (e) Corresponding EELS maps of the four lowest-order plasmonic modes spectrally centred at 0.8, 1.1, 1.3 and 2.0 eV, respectively. (f) HAADF image and (g) – (j) EELS maps of the first four plasmonic modes of a CSRR. The EELS maps were recorded at energy losses of 0.8, 1.1, 1.4 and 1.9 eV, respectively. The energy range shown in the EELS maps was set to 0.016 eV. The length of the scale bars is 200 nm. The white curves indicate the boundary of the SRR and the CSRR, respectively.

Babinet's principle relates the mode profiles of a SRR to the mode profiles of the corresponding CSRR by interchanging the E -field and B -field distributions [5]. For example, the fundamental mode of an SRR has a strong B_z -component in the centre of the SRR which results from the oscillating current along the entire ring. Therefore, the fundamental CSRR mode exhibits a strong E_z -component in the middle of the CSRR which is connected with a strong EELS signal (see Fig. 2 (g)). The EELS maps of the higher CSRR modes in Fig. 2 (h) – (j) can be explained accordingly.

4. Conclusions

STEM-EELS is a powerful method to map the plasmonic mode distributions of lithographically defined photonic metamaterials like SRRs or CSRRs.

References

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