Probing Magnetic and Toroidal Dipole Moments in Upright Split Ring Resonators using Correlative Electron Energy Loss Spectroscopy and Cathodoluminescence Spectroscopy

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Surface plasmon resonances (SPR) are collective oscillations of conduction electrons confined on the surface of nanostructures. Their ability to concentrate electromagnetic energy on the nanoscale and create artificial dipole moments has made SPR valuable in the engineering of optical metamaterials. The split ring resonator (SRR), in particular, has gained much interest because of its ability to support resonances which create a magnetic dipole moment [1]. When the SRR is fabricated upright on the substrate, as shown in Figure 1a, the magnetic dipole moment is parallel to the substrate and can be easily coupled to incident light or to neighbouring SRRs. Coupling between neighbouring SRRs allows the structure to support more exotic dipole moments, such as the toroidal dipole. The toroidal dipole moment is an electromagnetic dipole moment created from circulating magnetic fields: with an array of four coupled upright SRRs, a toroidal dipole moment can be created from the coupling of the magnetic dipole SPR mode in the four SRRs [2] (Figure 1h).

We present a correlative study of the SPR modes in both the single SRR and the coupled array of four SRRs using both electron energy loss spectroscopy (EELS) and cathodoluminescence (CL) spectroscopy. EELS maps were acquired with an energy resolution of 55-60 meV on an FEI Titan STEM (Figure 1b,i); CL data was acquired on an FEI XL-30 FEG-SEM (Figure 1d). The use of both techniques reveals the full spectrum of SPR modes in the single and coupled SRRs. We take advantage of the wide spectral range of EELS to probe the magnetic dipole mode in the single SRR (Figure 1c) and the hybridization of this mode in the coupled SRRs into two modes: a mode with a net magnetic dipole moment (Figure 1j) and a higher energy mode with a toroidal dipole moment (Figure 1k).

We will discuss the use of CL data in the visible spectrum to identify higher order modes in both the single and coupled SRRs, showing the hybridization of dipole modes on the coupled rims of the hollow SRR pillars. In the single SRR, we identify longitudinal bonding (Figure 1e) and anti-bonding (Figure 1f) rim modes and a transverse (Figure 1g) rim mode on the pillars [3]. We will discuss how this hybridization is affected by the coupling of four SRRs into a toroidal structure. We will also present the full spatially-resolved polarimetry state of the CL emission from the SPR modes present in the single SRR and in the array of four SRRs, linking this to the nature of the dipoles associated with each mode.

References

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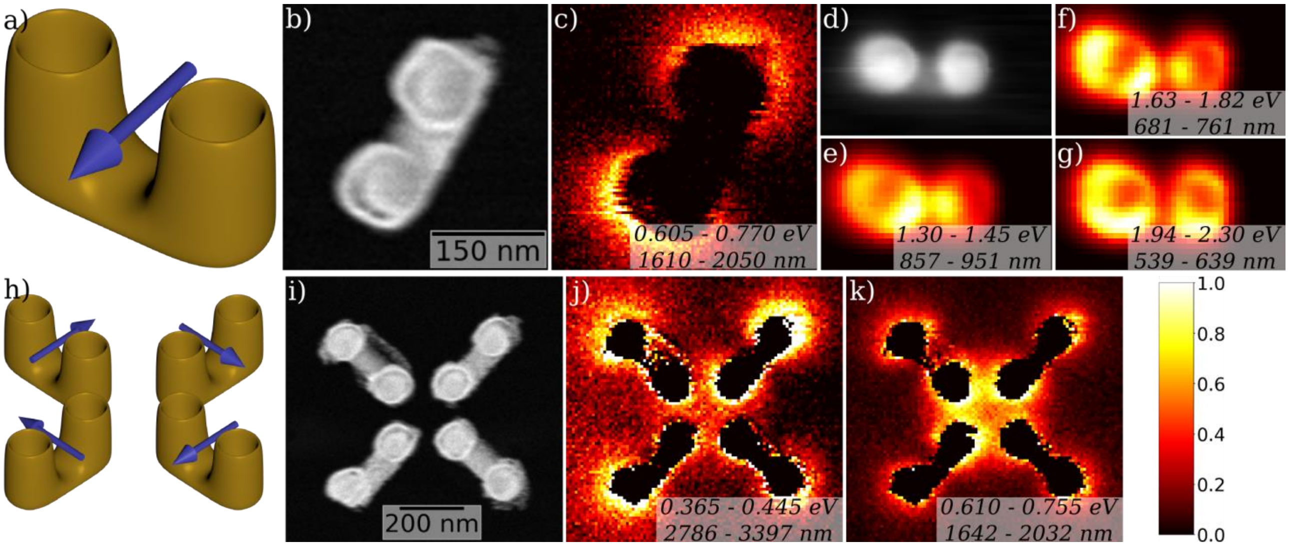

Figure 1: a) Magnetic dipole moment (blue arrow) in an upright SRR; b) high-angle annular dark-field (HAADF) image of upright SRR; c) EELS map of magnetic dipole mode (0.6875 eV); d) SEM image of the same upright SRR; e) CL map of the longitudinal bonding rim mode (1.375 eV); f) CL map of the longitudinal anti-bonding rim mode (1.725 eV); g) CL map of a transverse rim mode (2.120 eV); h) Array of four upright SRRs creating a toroidal dipole moment; i) HAADF image of four SRRs; j) EELS map of magnetic dipole mode (0.405 eV); k) EELS map of toroidal dipole mode (0.6825 eV). Colourbar represents normalized intensity scale for each SPR map.

Funding

This work was supported by the Natural Sciences and Engineering Research Council of Canada (NSERC) under the Discovery Grant Program (ICB, EPB, GAB); The Netherlands Organization for Scientific Research (SM, AP); and the European Research Council (ICB, SM, AP).