Unveiling longe-range coupling of toroidal moments by EELS and EFTEM

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In addition to electric and magnetic multipoles, there is a third independent family of elementary electromagnetic sources, namely dynamic toroidal multipoles [1]. Toroidal dipoles exhibit the peculiar character of possessing odd parity under space and time inversion in contrast to electric and magnetic dipoles. This characteristic feature makes toroidal moments particularly interesting for applications in solid state physics [2]. Similar to the coupling of electric or magnetic dipoles which tailors the optical responses, the coupling among toroidal moments is expected to form exotic new optical states. However, the fundamental toroidal dipole - dipole coupling is still open for theoretical and experimental exploration.

In a previous study we have demonstrated that toroidal resonances exist in the optical wavelength range. These resonances were excited in small holes arranged in the shape of a benzene molecule in a free-standing silver thin film [3]. Here, we propose a plasmonic decamer cavity structure in thin silver films to realise transverse coupling between toroidal dipoles (Figure 1). Coupling phenomena were investigated both experimentally and theoretically, by means of electron energy-loss spectroscopy (EELS) and energy-filtered transmission electron microscopy (EFTEM), together with finite-difference time-domain (FDTD) calculations [4].

We observe pronounced coupling effects of toroidal moments in plasmonic nanocavities, even at a separation of hundreds of nanometres. The coupling phenomena give rise to hybridised modes of toroidal moments with either parallel or antiparallel alignment (Figure 2). While increasing the number of antiparallel aligned toroidal moments, the energy of the hybridised modes increases. The underlying mechanism was further theoretically analysed and qualitatively described: in the regime of long-range coupling, the antisymmetric mode possesses a positive contribution from the interaction energy, whereas the symmetric mode yields a negative contribution from the interaction energy. This work further enhances the understanding of fundamental electromagnetic phenomena in the third family of electromagnetic sources, and paves the way for further research and exploitation in the fields of metamaterials, and light-matter interaction.

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References

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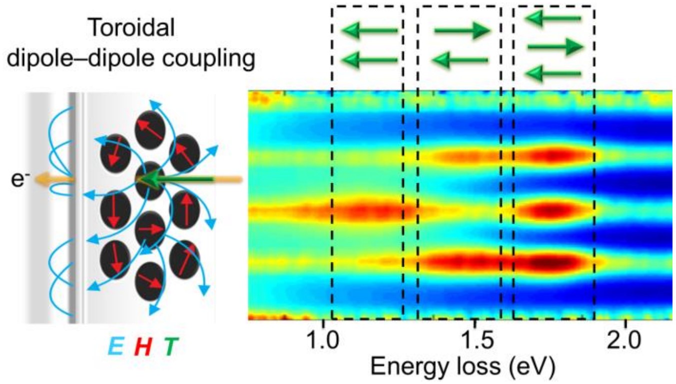

Figure 1. Schematic illustration of toroidal dipole - dipole coupling. Fast electrons induce simultaneous toroidal dipoles (green) in a plasmonic decamer nanocavity, with the induced electric (blue) and magnetic fields (red) sketched. The hybridisation of the toroidal dipole - dipole coupling is observed in the electron energy-loss spectra (right part), which are marked with the corresponding coupling alignments, respectively. Reprinted with permission from ref. [4]. Copyright (2018) American Chemical Society.

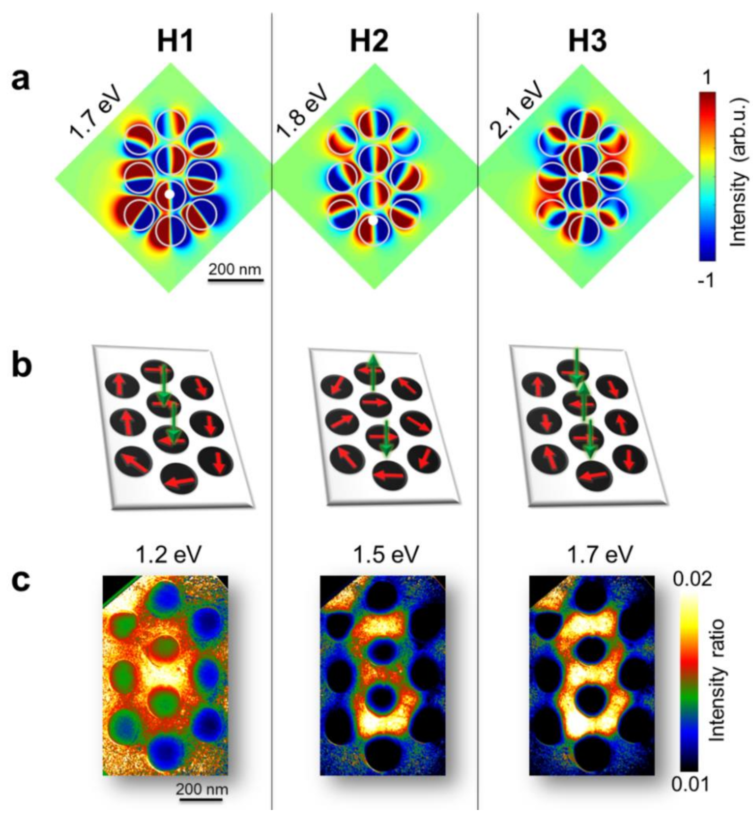

Figure 2. (a) Simulated magnetic fields along the electron trajectory of the decamer structure at resonance energy of the three coupled toroidal moments H1 (1.7 eV), H2 (1.8 eV), and H3 (2.1 eV), respectively. The grey circles denote the nanoholes. The impact locations of the electron probe used for the calculations are indicated by the white dots in the corresponding figures. (b) Schematic illustration for the H1, H2 and H3 modes in the plasmonic decamer nanocavity. Red and green arrows represent magnetic and toroidal dipoles, respectively. (c) Normalised EFTEM images of the decamer nanocavity at energy losses of 1.2 ± 0.1, 1.5 ± 0.1 and 1.7 ± 0.1 eV, respectively.