# Supporting Information to

# Long-range Coupling of Toroidal Moments for the Visible

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6 pages, 5 figures

List of content:

- three dimensional stream plots of magnetic and electric fields of modes H1, H2 and H3 (Figure S1)
- modal decomposition (Figure S2)
- three dimensional stream plots of magnetic and electric fields at 2.7 and 3.1 eV (Figure S3)
- plasmons in infinite silver slabs with varying thicknesses (Figure S4)
- experimental reproducibility (Figure S5)

1. Three-dimensional stream plots of the magnetic and electric fields of modes H1, H2 and H3

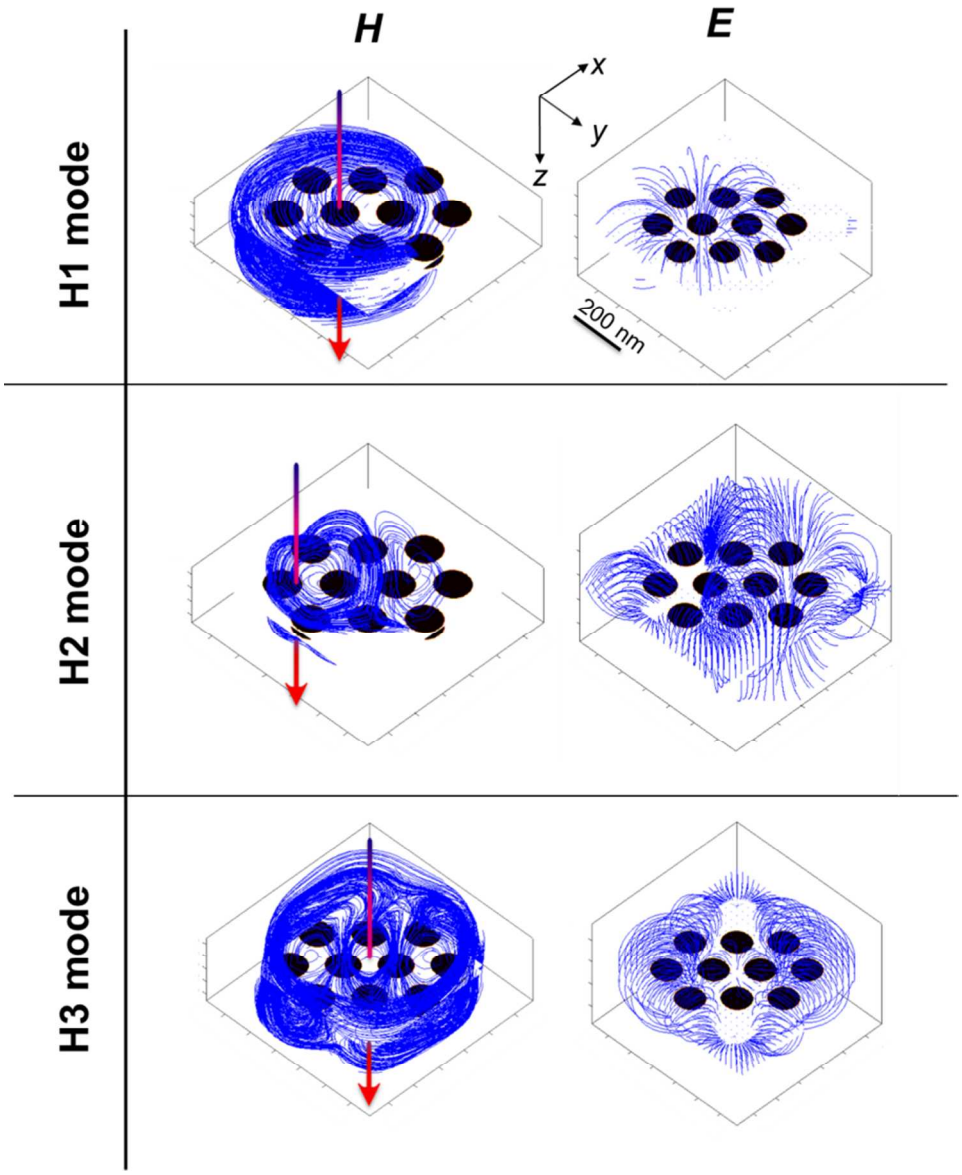


**Figure S1**. Simulated three-dimensional stream plots of the magnetic ***H*** and electric ***E*** fields of the modes H1, H2 and H3. Vertical arrows with gradient color represent the impact trajectories of electron beam. The scale bar of 200 nm applies to all the plots in *x*, *y*, *z* directions.

## 2. Modal decomposition

To further support our statements, we have calculated the induced current and charge density with space dependence, and used them to calculate the individual moments and the corresponding far-field radiation. Please note that the radiation was calculated excluding the matter. Hence, the actual radiation should be less intense. The contribution of the toroidal dipole moment in the z-direction is significantly dominant ($T_z$, green curve in Figure S2 (b) plotted on a log scale). These peaks marked by the red arrows (1.75, 2.0 and 2.2 eV) correspond to three coupled toroidal modes (1.7, 1.8, and 2.1 eV in Figure 2b). Since EELS is a near-field measuring technique, slight energy differences are expected when compared to the far-field radiation.

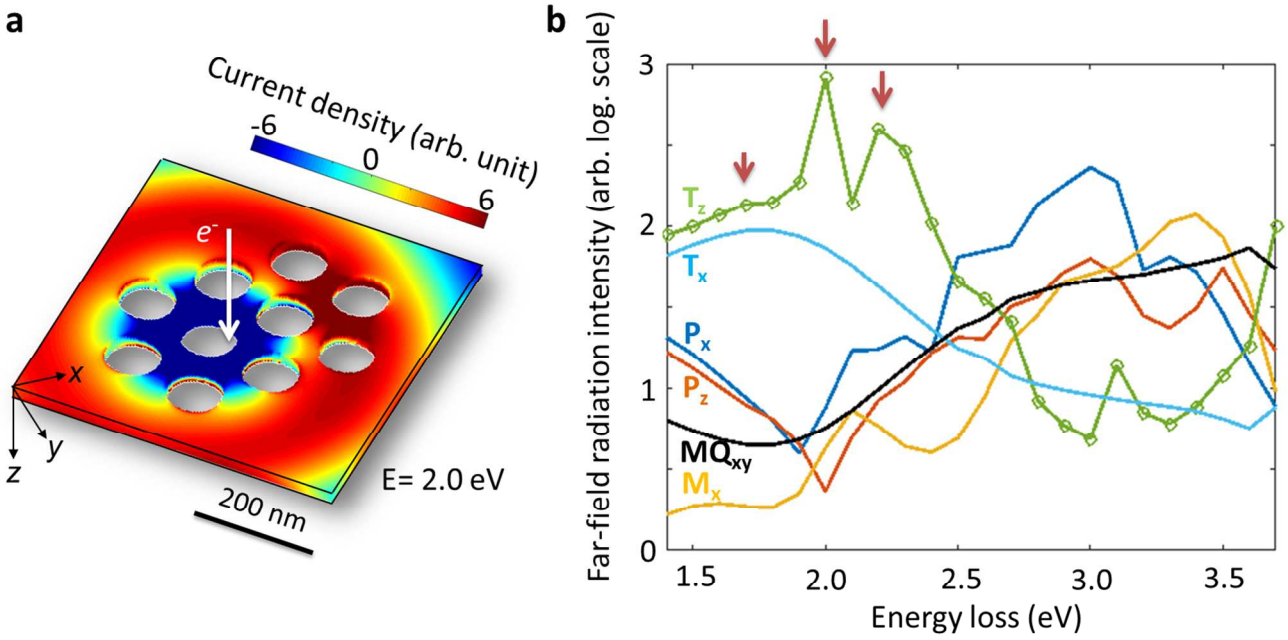


**Figure S2**. (a) Current density on the surface of the plasmonic decamer nanocavity induced by a fast electron (white arrow) at 2.0 eV. (b) Corresponding far-field radiation (scattered power) for 6 multipole moments induced in the structure. The peaks of the toroidal moment contribution $T_z$ are marked by red arrows. Notations: $P_x$–electric dipole moment along *x*-axis; $P_z$–electric dipole moment along *z*-axis; $M_x$–magnetic dipole moment along x-axis; $T_z$– toroidal dipole moment along *z*-axis; $T_x$– toroidal dipole moment along *x*-axis; $MQ_{xy}$–magnetic quadrupole moment in *x*-*y* plane.

## 3. Three dimensional stream plots of magnetic and electric fields at 2.7 and 3.1 eV

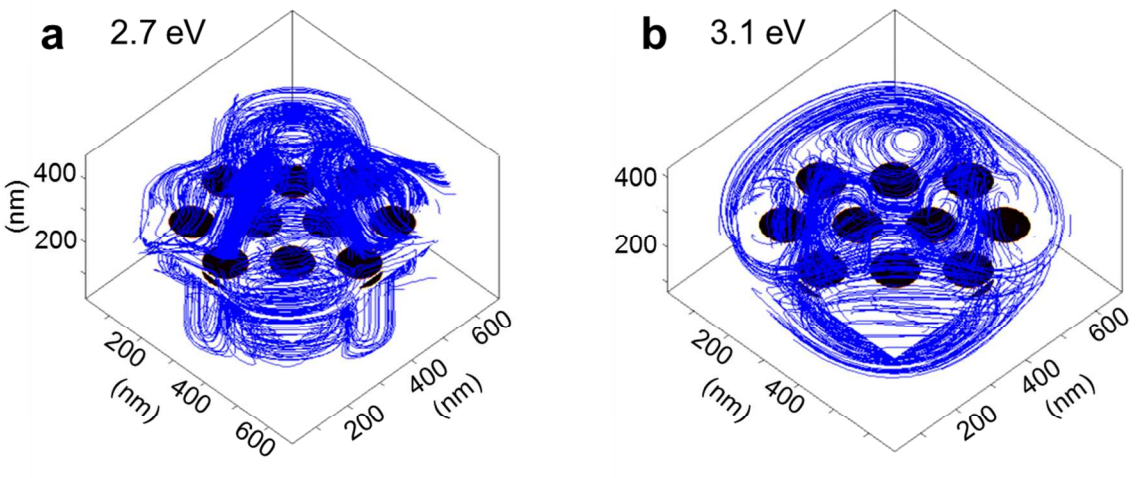


**Figure S3**. Simulated three-dimensional stream plots of the magnetic fields ***H*** at (a) 2.7 and (b) 3.1 eV, respectively. The electron position is at the center of the structure.

## 4. Influence of thickness variation on surface plasmon polaritons in infinite silver slabs

The thickness map of the investigated decamer cavity structure (Figure 2a) is shown in Figure S3a. The thickness between the nanoholes is approximately constant, which is 67 nm calculated from $t/\lambda = 0.8$ with the mean free path $\lambda \approx 84$ nm. However, the thickness varies slightly along the decamer long axis by about ~ 16 nm. The resonance of the infinite silver slab is calculated as function of slab thickness in steps of 10 nm from 20 to 80 nm in Figure S3b.

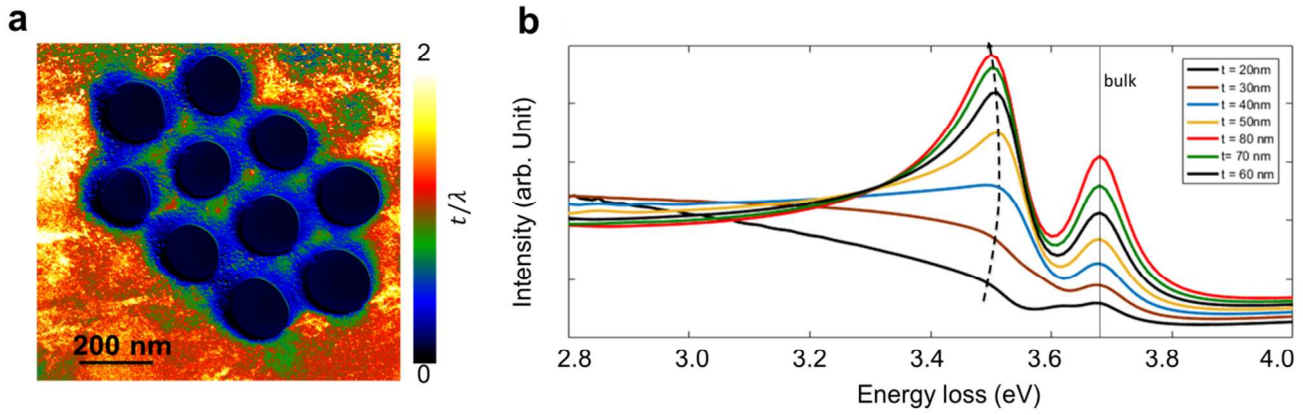


**Figure S4**. (a) Thickness map of the plasmonic decamer cavity structure shown in Figure 2a. (b) Simulated EEL spectra of infinite silver thin films with thicknesses from 20 to 80 nm at incremental steps of 10 nm. The black dashed line indicates the energy shift of the surface plasmon polariton mode at about 3.5 eV. The black solid line shows the location of silver bulk plasmon.

## 5. Experimental reproducibility

Here we present the experimental EELS results of another plasmonic decamer structure in Figure S4. Three resonances at 1.23, 1.56 and 1.82 eV are pronounced and well visible. Moreover, the locations of these modes fit very well with the characteristic locations of the hybridized modes H1, H2 and H3 which indicates the reproducibility of the toroidal moment coupling phenomena in plasmonic decamer cavities.

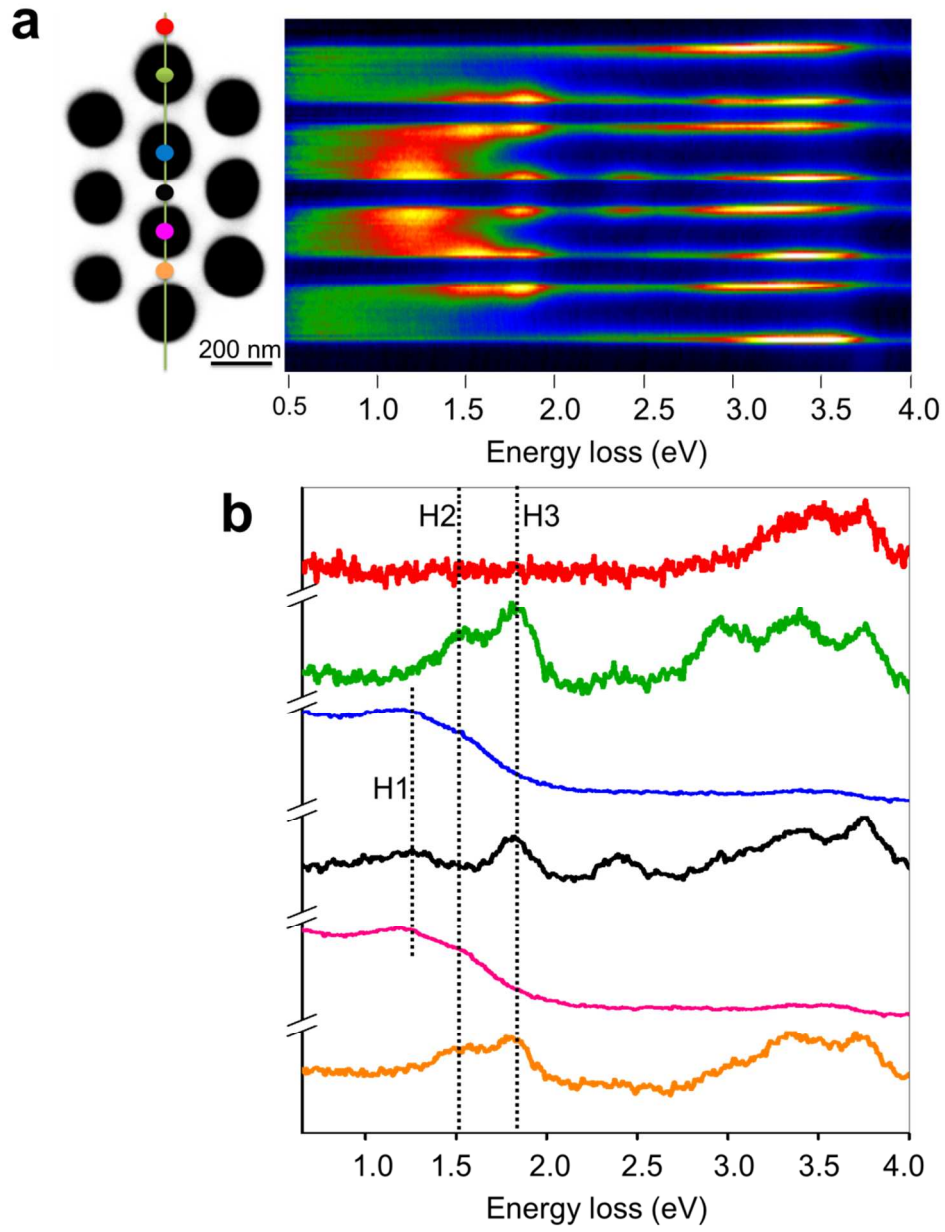


**Figure S5**. (a) High-angle annular dark-field image and EELS line scan of a fabricated decamer nanocavity. The experimental ZLP-subtracted EEL spectra are plotted as a function of impact location along the long axis of the cavity (green line). (b) Experimental ZLP-subtracted EEL spectra extracted from the 6 color marked locations in (a). Dotted lines indicate the peaks of the modes H1, H2 and H3.