**1. Experiment**

The cylindrical nanostructure samples were prepared by using an electron beam lithography (EBL) system in the Cornell Nanofabrication Facility on quartz substrates. AJEOL JBX-9600 FS (JEOL, Japan) system was used to expose the pattern with 20 nm resolution achieved at 100kV accelerating voltage. Two layers of positive photoresist: 2% poly methyl methacrylate (PMMA) in Anisole was used followed by 1% of PMMA in methyl isobutyl ketone (MBIK) were spin coated on silica substrate to a total thickness of 90 nm and each baked at 170°C for 15 minutes. A regular silver two dimensional array pattern with a fixed period of 150 nm in both directions and thickness of 40 nm was produced by a lift-off technique with increasing electron beam exposure dose. The structure contained individual Ag cylinders with varying size in the 30 – 100 nm range. The period of the pattern was 150 nm in both directions and the array size was 100 µm x 100 µm. In single particle arrays, the particles remained separated, as the spacing decreased with increasing size of the particles. The SEM images were obtained using Zeiss Ultra SEM without any sample preparation, and the height of the nanostructures was determined by atomic force microscope (AFM) measurements to be 30+/- 10 nm nm. The structures were deposited on quartz and coated with a 10 nm Al₂O₃ layer to protect against oxidation.

The extinction spectra were obtained by using the SEE2100 micro-spectrometer in transmission mode, across the spectral range of 380-1000 nm. The spectra were collected by 50x objective from small regions containing only a single type of pattern, with a numerical aperture of 0.7. The unpolarised and polarized illumination from a mercury lamp was used.

**Simulations**

The calculations were performed at a supercomputer cluster with 15 nodes, with each node based on an Intel Xeon chip E5530 @ 2.4GHz, with 8MB cache and 16 GB of RAM. The Comsol program version 3.5a, with the RF module, in its three-dimensional version was used for all simulations. The Comsol program is based on the Finite Element Method (FEM), and the RF module applies it to electromagnetic calculations. The FEM scheme used in this work employs non-orthogonal grids which resolve the problem of “spurious” solutions, frequent in calculations with rectangular grids. A typical geometry and mesh in the simulations are shown in Figure S1.

In the simulations a cube 150 x 150 x 150 nm was first defined. Within that cube a silver cylinder was located, with rounded or sharp edges (blue), and a protecting layer of Al₂O₃ (red), surrounding the cylinder and covering the substrate below. The refractive index of the silver used in this work was taken from the *Handbook of Optical Constants of Solids* edited by E. D. Palik, Academic Press, New York, 1985. The refractive index of the protecting layer was taken as 1.77 and the value of 1.55 was used for the substrate and 1 for air above the sample. The calculations were carried out for the boundary condition of a plane polarized electromagnetic wave at normal incidence arriving from above, and the upper and lower boundaries of the simulated regions were set to absorbing. Periodic or absorbing boundaries were also set at the cube sides, periodic for arrays of cylinders as in real nanolithography structures and absorbing for single cylinders simulated in the present work. A typical simulation of a single structure took about 2 hours.
We have tested the accuracy of our simulations in two ways. Firstly, we simulated the absorption of plane polarised light by a single 200 nm silver sphere by using the FEM method as described earlier and compared it with known analytical solution by using the Mie expansion, which is essentially exact. Figure S2 indicates that the two calculations are producing very close results. Secondly, we addressed the issue of singularities by checking whether the calculations for a typical geometry with sharp edges converge for the number of elements used. Figure S3 illustrates the differences observed when the number of elements used is increased from 2889 to 10698. As the change of simulated absorbance is insignificant we can conclude that the two peaks observed in the spectrum have been well rendered. The convergence of our numerical procedure indicates that we have been able to simulate the sample absorbance with good accuracy.

Figure S2. Absorbance of radiation by a silver sphere, calculated by the Mie expansion (red) and by FEM (black).
3. Assignment of multipolar character of the observed EM modes

In order to understand the multipolar character of the observed EM modes one needs to note that the colour scale in the figures illustrating EM field distributions indicates its direction. In this colour scale the regions marked with blue colour have electric field component pointing in the negative direction (i.e. to the left) and those marked red in the positive direction (to the right). Thus the EM field distribution in Figure 2a (repeated here for convenience in Figure S4 a) corresponds to a predominantly dipolar mode as shown in Figure S4 b. Likewise, the EM field distribution in Figure 3 b (repeated here as Figure S4 c) has a quadrupolar character (see Figure S4 d), and we should note a very distinctive contribution of a dipole. The distribution shown earlier in Figure 2 d (repeated here as Figure S4 e) has a higher multipolar character, similar to three dipoles one above the other and pointing in opposite direction (hexapolar), as shown in Figure S4 f. Of course none of the observed EM field distributions represents a pure multipole. For example the distribution in Fig S4 a shows a very weak higher multipolar (hexapolar) component, in addition to the dominant dipole.

Figure S3. Comparison of two FEM simulations with different numbers of elements. Red- 2889 elements, black – 10698 elements.
Figure S4. Illustration of the multipolar character of the selected EM modes. a) mode in Figure 2b, b) schematics of the corresponding dipole, c) mode in Figure 3b, d) schematics of the corresponding quadrupole, e) mode in Figure 2d, f) schematics of the corresponding higher multipole (hexapole).