Supplementary Information

Direct Excitation of Dark Plasmonic Resonances in Visible Light at Normal Incidence

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Figure S1: The proposed fabrication process of the nanostructure:

(a) Negative-tone hydrogen silsesquioxane (HSQ) spin-coated onto substrate. The thickness of the HSQ layer is 85nm.

(b) HSQ nanopillars after electron-beam lithography (EBL). The unexposed portions of the HSQ are developed away using a salty developer, leaving HSQ nanopillar array.

(c) A layer of silver is deposited onto the samples using an electron-beam evaporator. The silver layer is about 10nm and covers the top of HSQ pillars and the blank area on the substrate, leaving the side of pillars bare.

(d) Another 5nm silver layer is coated cover the whole structure by Atomic Layer Deposition (ALD). The gray area in black frame represents the silver deposited in process (c). After ALD the top/bottom silver layer is 15nm and the sidewall is 5nm.
Figure S2: The comparison of the proposed 2-step silver deposition process and the simplified ALD process.

Figure S2 The reflectance of nanostructures proposed by (1) the 2-step deposition (black curve), in which $t_1=t_2=15\,\text{nm}$, $t_3=5\,\text{nm}$; (2) ALD of silver with thickness of $t_1=t_2=t_3=5\,\text{nm}$ (red curve); (3) ALD of silver with thickness of $t_1=t_2=t_3=10\,\text{nm}$ (blue curve). Seen in Fig 1b in the manuscript, $t_1$ is the thickness of the bottom Ag layer, $t_2$ is the thickness of the top Ag disks and $t_3$ is the thickness of the Ag sidewall. Height and radius of HSQ pillar is 85nm and 45nm, period of the unit cell is 120nm.

In the simplified ALD cases, the quadrupole dark modes are still supported, but much weaker than the original (black curve) quadrupole mode and the hexapole mode cannot be observed any more. The wavelength shift of resonance for case $t=10\,\text{nm}$ (blue curve) is due to the change of gap size and the top disk size caused by the incremental of sidewall thickness.