

Omnidirectional and band-tunable light absorption in free-standing dielectric-metal core-shell resonator arrays

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Highly efficient light absorption is required in many optical applications, e.g. photo-thermal conversion, photocatalytic reaction and photovoltaic cells.¹ It is also critical to develop light absorbers that allow omnidirectional absorption of optical energy within desirable spectral ranges to avoid, for example, the need for active tracking in solar energy conversion applications.

Here we report free-standing periodic arrays of dielectric-metal core-shell nanoparticles, which demonstrate near-unity light absorption at tunable frequencies within a wide incident angle range. Specifically, the core-shell structures are based on free-standing monolayers that are self-assembled from silica (SiO₂) nanospheres (diameter ~ 400 nm). Coating both sides of the monolayers with 40 nm thick silver (Ag) films form a hexagonally patterned array of SiO₂-Ag core-shell nanoparticles (Fig.1). Angle-resolved optical measurements (Fig.2a and 2b) show that over a 100-nm spectral range (550-650 nm) the arrays achieve high (>80%) and polarization-independent light absorption within a broad angle range from -40° to 40°. In addition, the spectral position and range of absorption can be easily adjusted by altering the core diameter and shell thickness.²

Numerical simulations (Fig.2c) show that such high light absorption is enabled by the hybridization of multiple optical modes, which consists of localized surface plasmon resonances (LSPRs) that are excited within the Ag shells, surface plasmon polaritons (SPPs) that propagate across core-shell arrays and whispering gallery modes (WGMs) that are localized inside each core-shell resonators.^{3,4} The excitation of both WGMs and LSPRs are insensitive to incident angles, which results in the omnidirectional light absorption of the resonator arrays.

¹ T. V. Teperik, F. J. García De Abajo, A. G. Borisov, M. Abdelsalam, P. N. Bartlett, Y. Sugawara, and J. J. Baumberg, *Omnidirectional Absorption in Nanostructured Metal Surfaces*, Nat. Photonics **2**, 299 (2008).

² P. Gu, M. Wan, W. Wu, Z. Chen, and Z. Wang, *Excitation and Tuning of Fano-like Cavity Plasmon Resonances in Dielectric-Metal Core-Shell Resonators*, Nanoscale **8**, 10358 (2016).

³ R. M. Cole, Y. Sugawara, J. J. Baumberg, S. Mahajan, M. Abdelsalam, and P. N. Bartlett, *Easily Coupled Whispering Gallery Plasmons in Dielectric Nanospheres Embedded in Gold Films*, Phys. Rev. Lett. **97**, 1 (2006).

⁴ T. A. Kelf, Y. Sugawara, R. M. Cole, J. J. Baumberg, M. E. Abdelsalam, S. Cintra, S. Mahajan, A. E. Russell, and P. N. Bartlett, *Localized and Delocalized Plasmons in Metallic Nanovoids*, Phys. Rev. B - Condens. Matter Mater. Phys. **74**, 1 (2006).

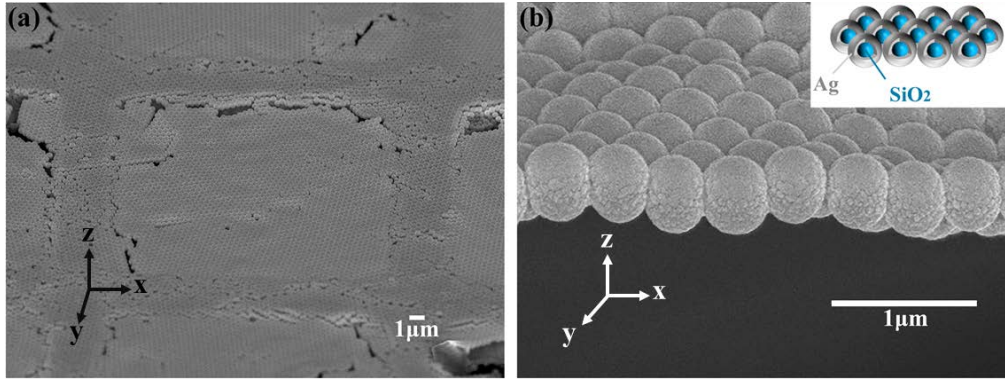


Figure 1: Scanning electron microscopy (SEM) images of free-standing $\text{SiO}_2\text{@Ag}$ core-shell resonator array: (a) the surface morphology of free-standing $\text{SiO}_2\text{-Ag}$ core-shell nanosphere arrays on copper grids, where the nanosphere monolayer is suspended in air due to electrostatic attraction; (b) the enlarged SEM image of cross-section of the array. The inset here shows the schematic of the core-shell structure.

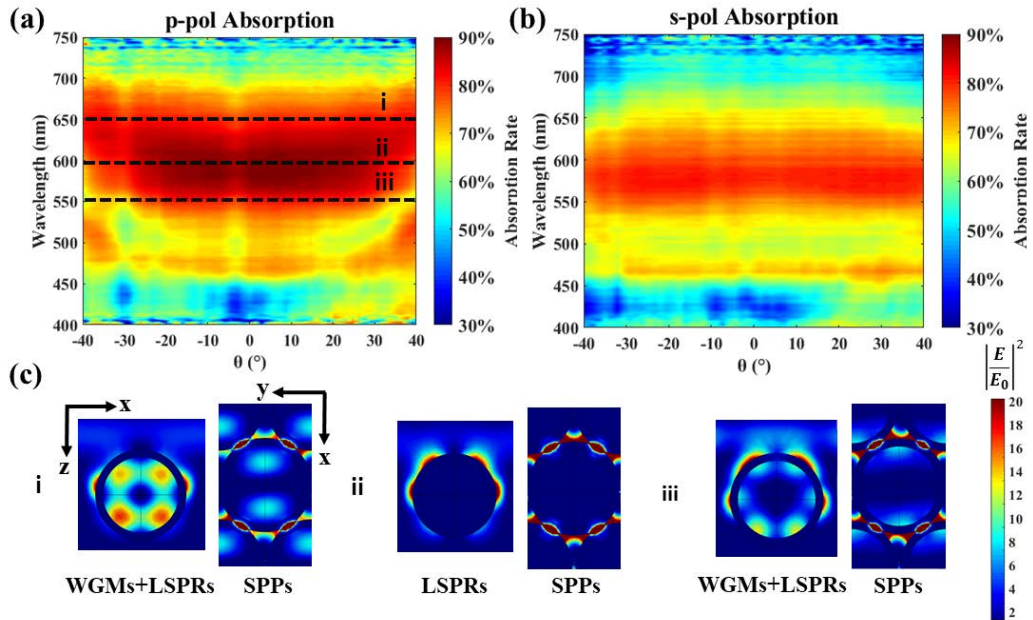


Figure 2: Experimentally acquired angle-resolved absorption spectra under p-polarized (a) or s-polarized illumination (b), where p- or s- polarization refer to electric field perpendicular or parallel to the incident plane respectively). (c) Numerically modelled electric field enhancement $|E/E_0|^2$ of **i**, **ii** and **iii** modes labelled in panel (b).