Supporting Information

Hot carrier generation in two-dimensional silver nanoparticle array at different excitation wavelengths under on-resonant condition

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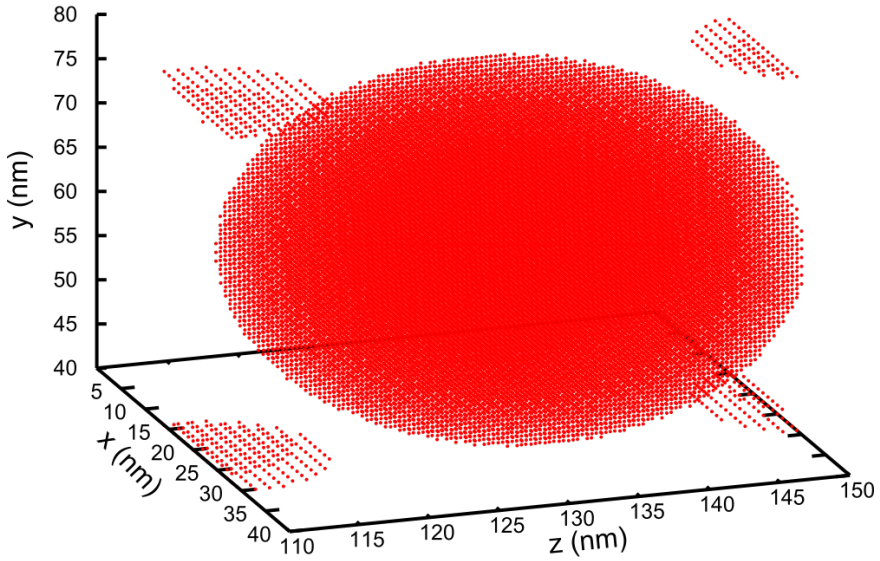
1. Simulation model for Discrete dipole approximation

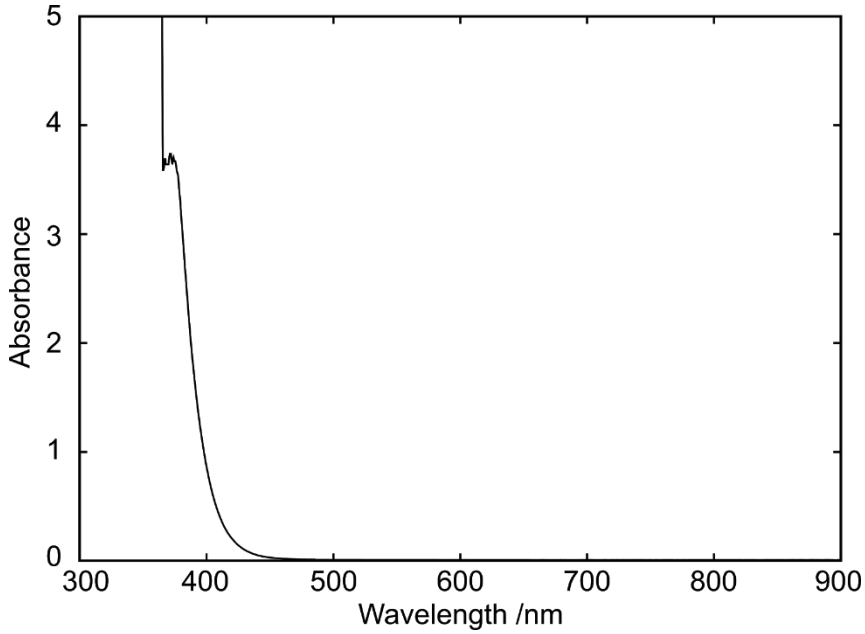
In the simulations, we hexagonally aligned 30 AgNPs (40 nm) in yz-plane with the inter-particle gap (*G*) of 1, 2, 4 and 6 nm, as shown in Fig. 4. In DDA calculation, the metallic structures are assumed to be composed by discrete dipoles. SFig. 1 shows magnified AgNP array (*G*= 4nm) composed of dipoles separated by 1 nm. Therefore, the AgNP was composed of 33401 dipoles, which resulted in 1002030 dipoles for the whole AgNP array. The optical response of the AgNP was calculated based on Mie theory, where the incident light is plane wave propagating along the x-axis. We referred the complex dielectric function of silver, which was assumed to be isotropic in x, y and z-axes, to Ref. S1.



SFigure 1. Simulation model for DDA. AgNPs are composed of dots, which represent dipoles.

2. Absorption spectrum of *p*-ATP

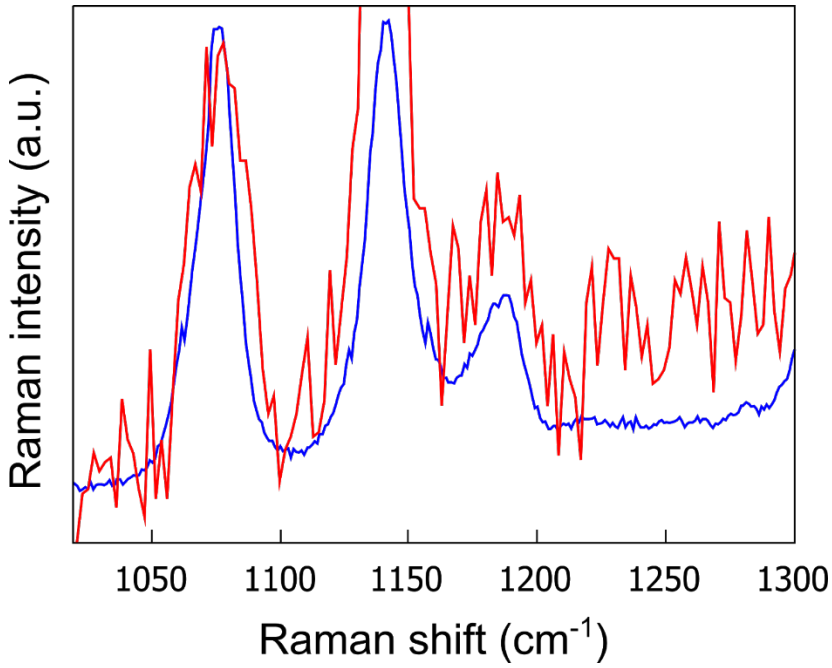
Absorption spectrum of *p*-ATP was measured, which is shown in SFig. 2. Almost no absorption was observed at >450 nm although ultraviolet light is strongly absorbed.



SFigure 2. Absorption spectrum of *p*-ATP (c.a. 1 wt% in ethanol).

3. The comparison of peak width at 1080 cm^{-1} between 532 nm and 458 nm

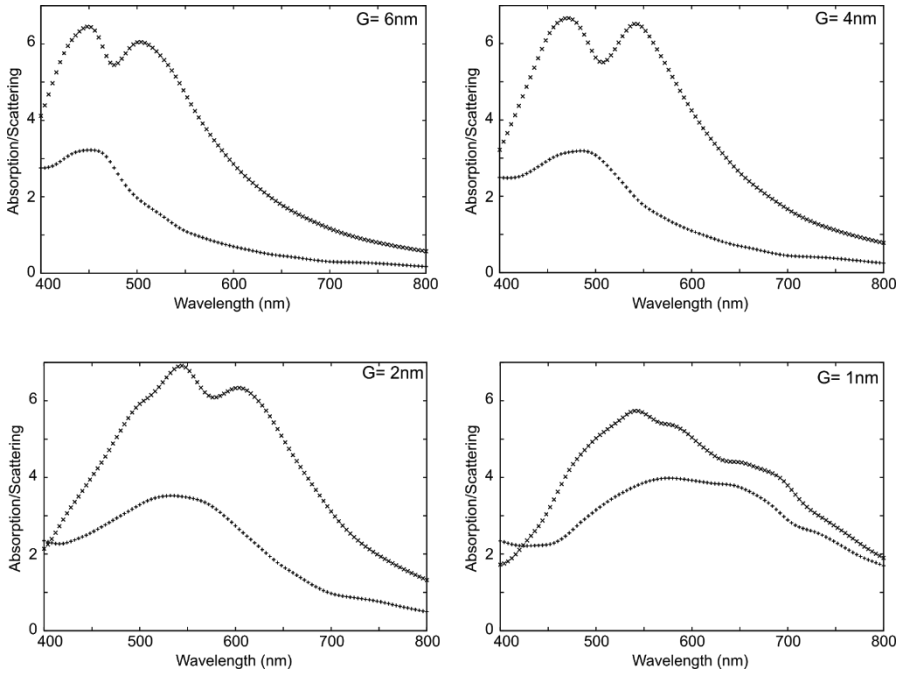
We compared the peak width at 1080 cm^{-1} at the same height between 532 nm and 458 nm.



SFigure 3. Comparison of peak width between 532 nm (blue) and 458 nm (red)

3. Absorption and Scattering components in Extinction of AgNP arrays

We calculated the extinction spectra of AgNP arrays at the inter-particle gaps of 6, 4, 2 and 1 nm with DDA, which are shown in Fig. 4. Extinction includes the absorption and scattering components, and the components can be calculated individually in DDA. SFigure 3 shows the absorption and scattering spectra of AgNP arrays at the inter-particle gaps of 6, 4, 2 and 1 nm, respectively.



SFigure 4. Absorption (+) and Scattering (x) spectra of AgNP (40 nm) arrays at the inter-particle gaps of 6, 4, 2 and 1 nm.

References

(S1) A. D. Rakić, A. B. Djurišić, J. M. Elazar, M. L. Majewski, Optical properties of metallic films for vertical-cavity optoelectronic devices. *Appl. Opt.*, 1998, **37**, 5271-5283.