**Supplementary Materials**

**Designing an efficient self-assembled plasmonic nanostructures from spherical shaped nanoparticles**

Vasanthan Devaraj 1,Ϯ, Jong-Min Lee 1,2,Ϯ , Ye-ji Kim 3 , Hyuk Jeong 1 and Jin-Woo Oh 1,3,4,\*

1Bio-IT Fusion Technology Research Institute, Pusan National University, Busan, 46241, Republic of Korea; devarajvasanthan@gmail.com

2School of Nanoconvergence Technology, Hallym University, Chuncheon, 24252, Republic of Korea; jongminlee1984@gmail.com

3Department of Nano Fusion Technology and BK21 Plus Nano Convergence Division, Pusan National University, Busan, 46241, Republic of Korea; kkyeaj0608@gmail.com

4Department of Nanoenergy Engineering, Pusan National University, Busan, 46241, Republic of Korea

\*Correspondence: ojw@pusan.ac.kr

ϮV.D. and J.-M.L contributed equally.

Diagram

Description automatically generated

**Figure S1.** Modelling information which is used throughout our simulations.

Chart, radar chart, histogram

Description automatically generated

**Figure S2.** Simulated broadband near field spectra |E/E0| for dimer nanostructures with NP shapes of a sphere (**a**), disk (**b**), and cube (**c**) as a function of gap size.

Chart, histogram

Description automatically generated

**Figure S3.** Simulated broadband near field spectra |E/E0| for trimer nanostructures with NP shapes of a sphere (**a**), disk (**b**), and cube (**c**) as a function of gap size.

Diagram

Description automatically generated

**Figure S4.** Schematic description of surface charge distributions reveals dipolar or quadrupolar modes in a plasmonic cavity region.

Chart, line chart

Description automatically generated

**Figure S5.** Data covering the FW3QM region in evaluating the broadband spectral performance of plasmonic nanostructures.

Chart, histogram

Description automatically generated

**Figure S6.** Simulated broadband gap-size dependent near field spectra |E/E0| for sphere-based dimer nanostructures with different NP diameter: 60 nm (**a**), 70 nm (**d**), 80 nm (**c**) and 90 nm (**d**).

Chart, histogram

Description automatically generated

**Figure S7.** Simulated broadband gap-size dependent near field spectra |E/E0| for sphere-based trimer nanostructures with different NP diameter: 60 nm (**a**), 70 nm (**d**), 80 nm (**c**) and 90 nm (**d**).

Table

Description automatically generated

**Table 1.** FW3QM calculation information for dimer nanostructures for different gap sizes.

Table

Description automatically generated

**Table 2.** FW3QM calculation information for trimer nanostructures for different gap sizes.