Supporting Information for

Photothermal Therapeutic Ability of Copper Open-Shell Nanostructures that are Effective in Second Biological Transparency Window Based on Symmetry Breaking-induced Plasmonic Properties

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1. Detailed characterization of CuOSN arrays using SEM images, AFM images, and XRD spectrum

To analyze their morphological properties and structural homogeneity, we observed the arrays of CuOSNs prepared on a glass plate using a scanning electron microscopy (SEM). It was confirmed from **Figure S1**(A) and (B) that the two-dimensional arrays of CuOSNs with slight roughened surface were formed by depositing Cu (thickness: 15 nm) on two-dimensional silica arrays with a quasi-hexagonal packing. Furthermore, it was confirmed that the Cu thin films were deposited onto the upper hemisphere surface of silica spheres from **Figure S1**(C) (normal SEM image) and (D) (backscattered electron image). In addition, the centerline average roughness (R_a) value of Cu thin films on the silica spheres was estimated to be 0.83 nm from the tapping mode atomic force microscope (AFM) image (**Figure S2**). Collectively, it was demonstrated that CuOSNs, with a slightly rough surface, were uniformly fabricated by depositing Cu thin films onto the upper hemispheres of silica spheres. In the XRD spectrum (**Figure S3**) of CuOSNs treated with glacial acetic acid, in addition to a broad peak around 23º, which is attributed to the amorphous SiO_2 nature of silica spheres, a weak peak was observed at 43.3º, which is attributed to the (111) plane of Cu. This observation suggests that the surface of CuOSNs treatment with glacial acetic acid is dominated by metallic Cu.

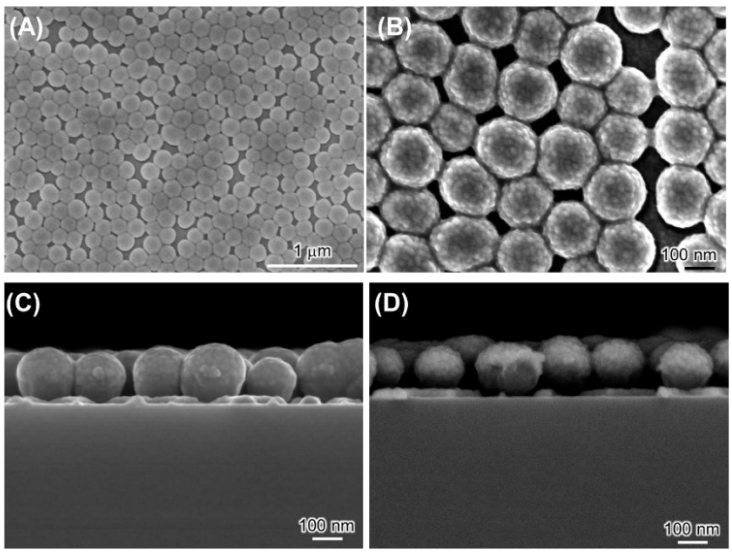

Figure S1. SEM images of arrays of CuOSNs. (A) and (B) are the top views and (C, normal image) and (D, backscattered electron image) are the cross-sectional views.

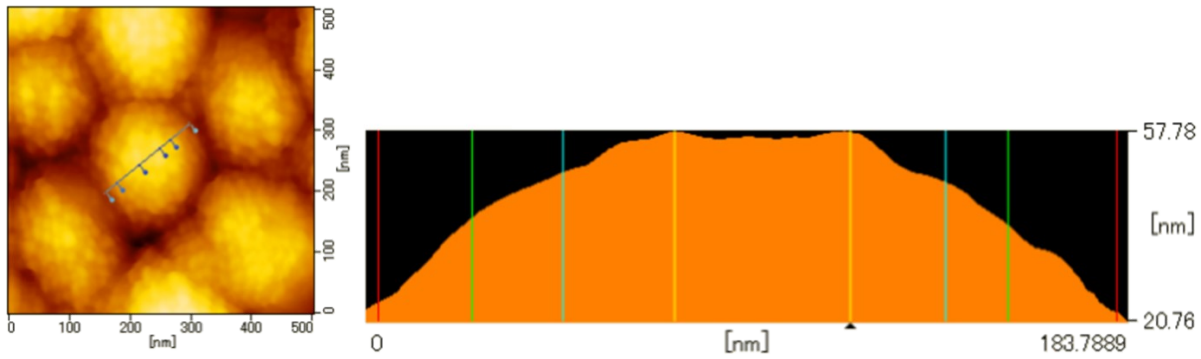

Figure S2. AFM image of arrays of CuOSNs.

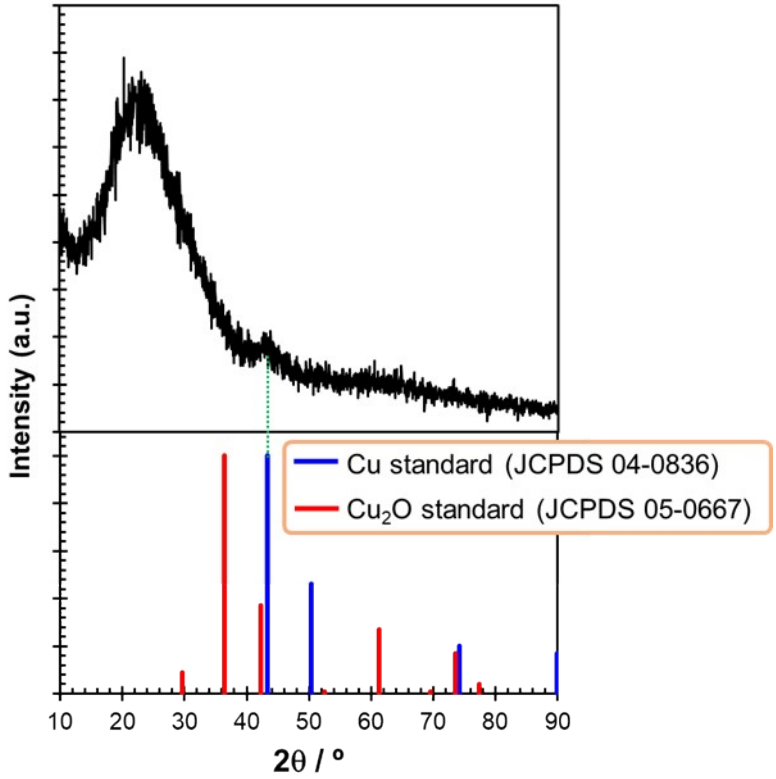

Figure S3. XRD spectrum of CuOSNs after treatment with glacial acetic acid. The lower panel shows the Cu and Cu_2O standard data.

2. XPS spectra of CuHS/MHA/SiO_2 before and after modification with PEG

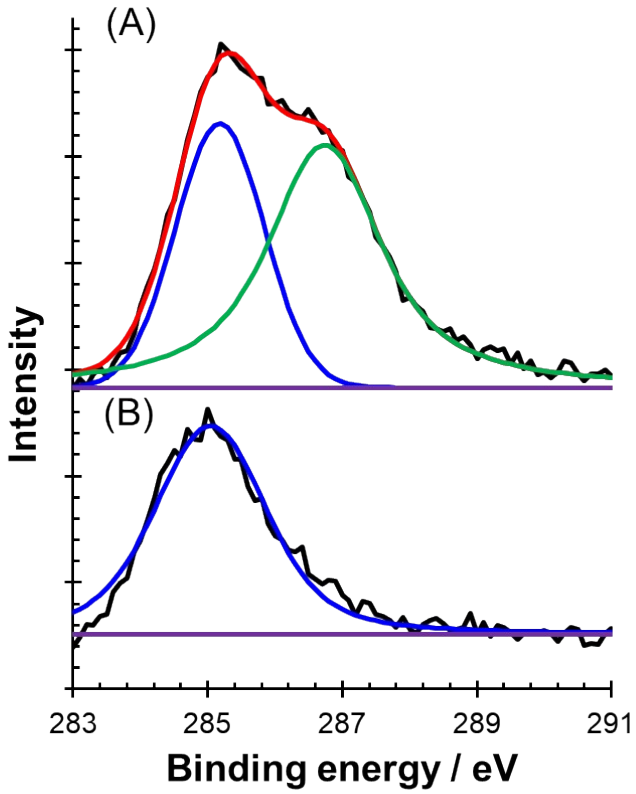

Figure S4. XPS spectra of CuHS/MHA/SiO_2 before and after modification with PEG. (A) C 1s narrow spectrum of CuHS/MHA/SiO_2 after modification with PEG. The peak is found to be composed of two peaks by deconvolution. (B) C 1s narrow spectrum composed of one peak for CuHS/MHA/SiO_2 before modification with PEG.

3. Optical and morphological properties of AuOSNs/MHA/SiO_2

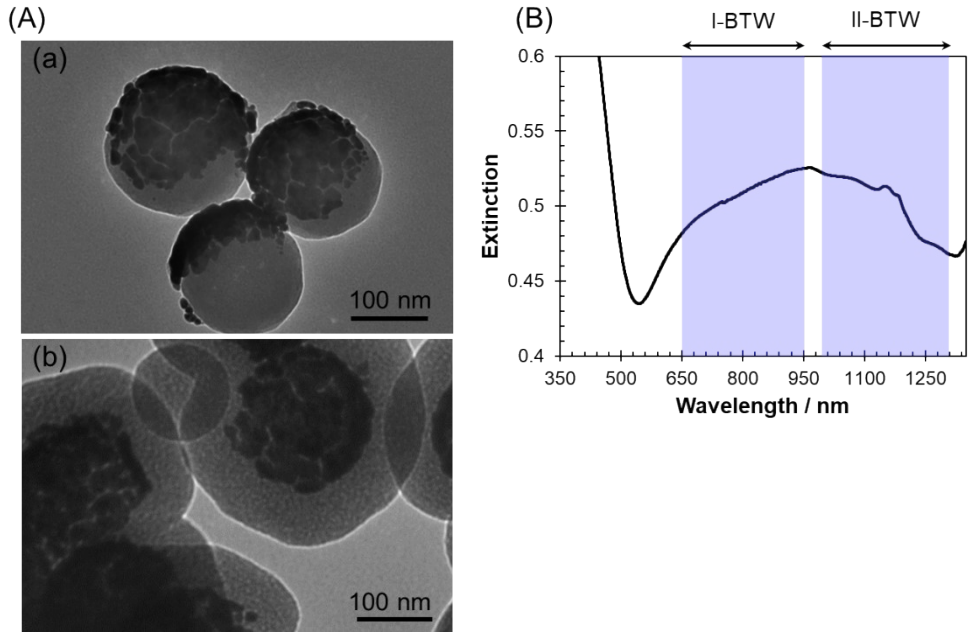

Figure S5. Morphological and optical properties of AuOSNs/MHA/SiO_2. (A) TEM images of (a) AuOSNs/MHA and (b) AuOSNs/MHA/SiO_2 and (B) an extinction spectrum of colloidal aqueous solution of AuOSNs/MHA/SiO_2.

4. Calculation of time constants for heat transfer from the systems

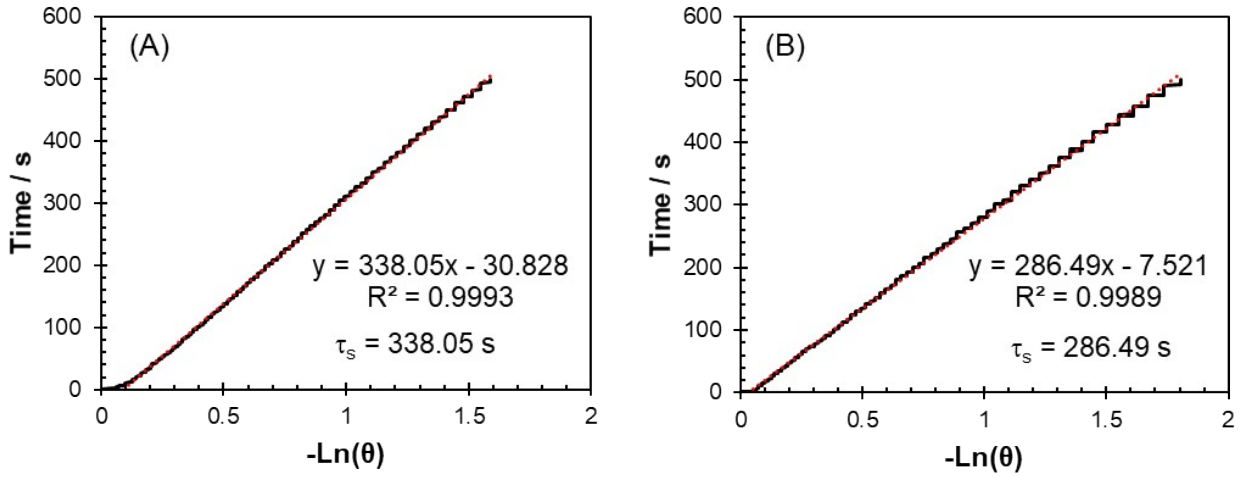

Figure S6. Time constants for heat transfer from the systems were calculated by applying the linear regression to the relationship between the cooling periods versus negative natural logarithm of driving force temperature, which is obtained from **Figure 8**(A): (A) CuOSNs/MHA/SiO_2 and (B) AuOSNs/MHA/SiO_2 under 1060 nm laser irradiation.

5. Extinction spectra of CuOSNs/MHA/SiO_2 before and after laser irradiation

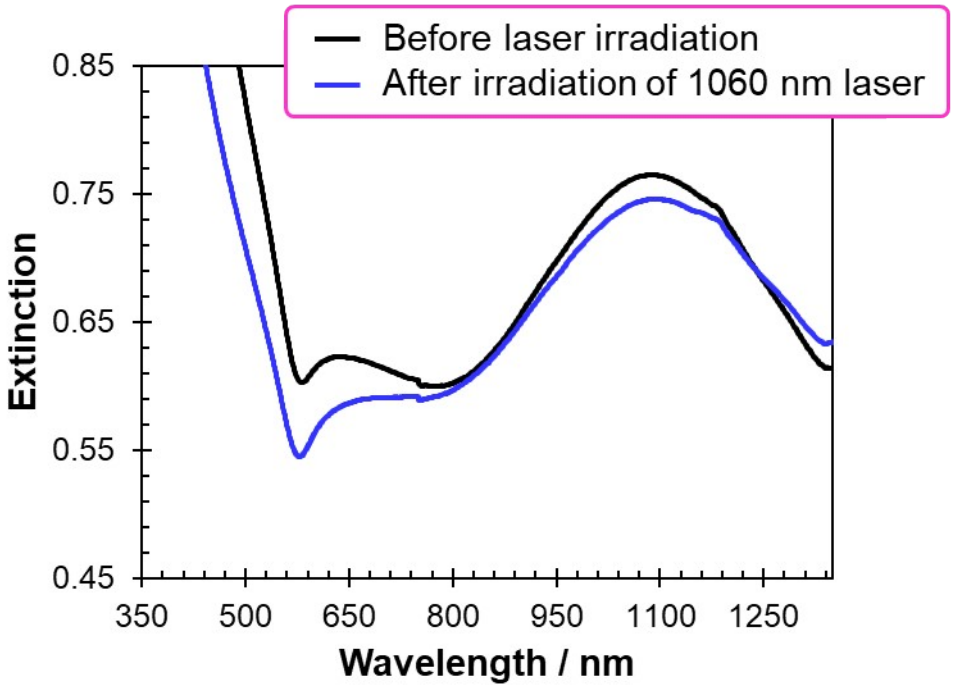

Figure S7. Extinction spectra of colloidal aqueous solution of CuOSNs/MHA/SiO_2 before and after irradiation with 1060 nm laser.