## **Supporting Information for**

Photoluminescence Manipulation of WS<sub>2</sub> Flakes by an individual Si nanoparticle

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Figure S1. Characterization of WS<sub>2</sub> flakes. (a)-(d) The corresponding SEM images of WS<sub>2</sub> flakes displayed in Figure 1. Scale bar,  $5\mu$ m. (e), (f) PL emission spectra of the 300 nm SiO<sub>2</sub>/Si substrate and Si NPs, respectively.



**Figure S2.** PL spectra of (a) 1L-, (b) 2L-, (c) 3L-, (d) 4L-, (e) ML-WS<sub>2</sub> flakes on a 300 nm SiO<sub>2</sub>/Si substrate at various excitation laser power. The arrows indicate the changes of A exciton peaks. (f) The wavelength changes of A exciton peaks of WS<sub>2</sub> flakes at various excitation laser power. In our measurements, 100% excitation equals to 2 mW excitation laser power.



Figure S3. Raman spectra of (a) 1L-, (b) 2L-, (c) 3L-, (d) 4L-, (e) ML-WS<sub>2</sub> flakes on a 300 nm SiO<sub>2</sub>/Si substrate at various excitation laser power. In our measurements, 100% excitation equals to 2 mW excitation laser power. (f) The changes of Raman modes ( $E_{2g}^{1}$  and  $A_{1g}$ ) of WS<sub>2</sub> with different thickness at the excitation laser power of 2 mW.



Figure S4. The typical morphology and structure characterization of Si NPs prepared by fs-LAL. (a) SEM image, (b) XRD pattern, (c) TEM image, (d) high-resolution TEM image, (e) selected-area electron diffraction (SAED) pattern of Si NPs.



**Figure S5. The PL mapping of Si/WS<sub>2</sub> heterostructures. (a), (d), (g)** The bright-field optical microscopy images of Si/1L-WS<sub>2</sub>, Si/3L-WS<sub>2</sub> and Si/ML-WS<sub>2</sub>, respectively. The white rectangular box represents the scanning range of PL mapping. **(b), (e), (h)** The corresponding SEM images of Si/1L-WS<sub>2</sub>, Si/3L-WS<sub>2</sub> and Si/ML-WS<sub>2</sub>, respectively. **(c), (f), (i)** The corresponding PL mapping images of Si/1L-WS<sub>2</sub> (620 nm), Si/3L-WS<sub>2</sub> (650 nm) and Si/ML-WS<sub>2</sub> (650 nm), respectively. PL mapping step size, 200 nm.



Figure S6. Optical constant of WS<sub>2</sub> flakes. (a) The scattering spectra of WS<sub>2</sub> flakes of different layer numbers. The orange and olive arrows indicate A and B excitonic resonances in WS<sub>2</sub> flakes. (b) Real and (c) Imaginary parts of dielectric function for 1L-WS<sub>2</sub> (blue line) and the bulk (red line), respectively. (d), (e) The simulated reflectance of WS<sub>2</sub> flakes of different thicknesses (1 nm, 10 nm and 20 nm) on the 300 nm SiO<sub>2</sub>/Si substrate and SiO<sub>2</sub> substrate, respectively.



**Figure S7. PL enhancement in Si/4L-WS**<sub>2</sub> heterostructures. (a), (f) The dark-field optical microscopy images of Si NPs deposited on 4L-WS<sub>2</sub>. The selected heterostructures are marked with white dashed circles and numbered from 1 to 3. Insets are the corresponding SEM and bright-field optical microscopy images. The scale bar is  $10 \,\mu\text{m}$ . (b) SEM images of the typical heterostructures selected in (a). The scale bar is  $500 \,\text{nm}$ . (c) The measured scattering spectra of the heterostructures compared with that of the bare 4L-WS<sub>2</sub>. The vertical dashed line located at around 640 nm indicates the A excitonic resonance. (d) PL spectra of the typical heterostructures compared with the bare 4L-WS<sub>2</sub>. (e) PL enhancement of the heterostructure varies with the scale of Si NPs.



Figure S8. The AFM images including height profiles of WS<sub>2</sub> flakes displayed in Figure 5a and 5b. Scale bar, 2µm.



Figure S9. Multipolar contributions of Si NPs with different sizes.



Figure S10. The decay kinetics obtained from Si/WS<sub>2</sub> heterostructures compared with bare WS<sub>2</sub>. (a) The time-resolved spectra of Si/1L-WS<sub>2</sub> heterostructures shown in Figure 3a and 3b. (b) The time-resolved spectra of Si/2L-WS<sub>2</sub> heterostructures shown in Figure 4a. (c) The time-resolved spectra of Si/3L-WS<sub>2</sub> heterostructures shown in Figure 4f. (d) The time-resolved spectra of Si/4L-WS<sub>2</sub> heterostructures shown in Figure S7. (e) The time-resolved spectra of Si/ML-WS<sub>2</sub> heterostructures shown in Figure 5a and 5b.