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Controlling plasmon-exciton interactions through photothermal reshaping

Aiqin Hu^{1,2}, Shuai Liu^{1,3}, Jingyi Zhao¹, Te Wen¹, Weidong Zhang¹,
Qihuang Gong^{1,2}, Yongqiang Meng³, Yu Ye¹ and Guowei Lu^{1,2*}

¹State Key Laboratory for Mesoscopic Physics, Collaborative Innovation Center of Quantum Matter, Nano-optoelectronics Frontier Center of the Ministry of Education, School of Physics, Peking University, Beijing 100871, China. ²Collaborative Innovation Center of Extreme Optics, Shanxi University, Taiyuan, Shanxi 030006, China. ³School of Materials Science and Engineering, Hebei University of Science and Technology, Shijiazhuang 050018, China.

*Correspondence: G W Lu, E-mail: guowei.lu@pku.edu.cn

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Fig. S1–Fig. S8

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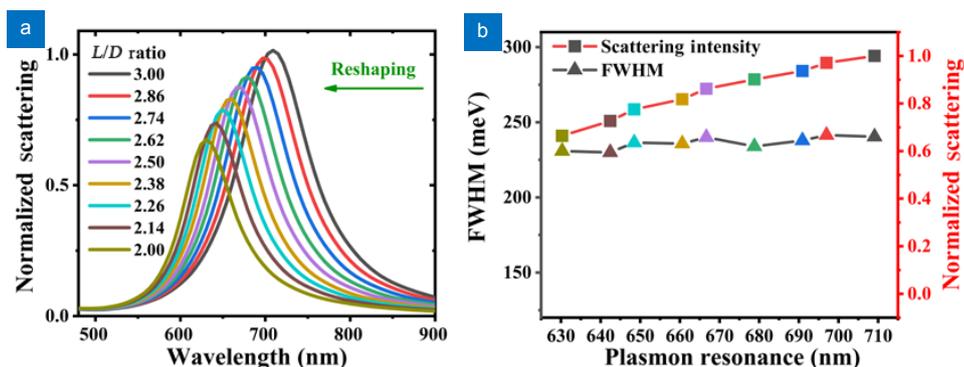


Fig. S1 | (a) Scattering spectra of a single nanoparticle with different shapes but the same volume placed on glass calculated by the FDTD method. During the reshaping, changing the length-diameter ratio of an ellipsoid without changing its volume and height to simulate the photothermal reshaping process approximately. When the particle reshaped from a rod shape to a circular disk shape, the peak of scattering spectrum blue-shifts, (b) scattering intensity reduces (red line), and scattering FWHM changes weakly (black). The dot color in (b) corresponding to the color of the scattering spectra in (a).

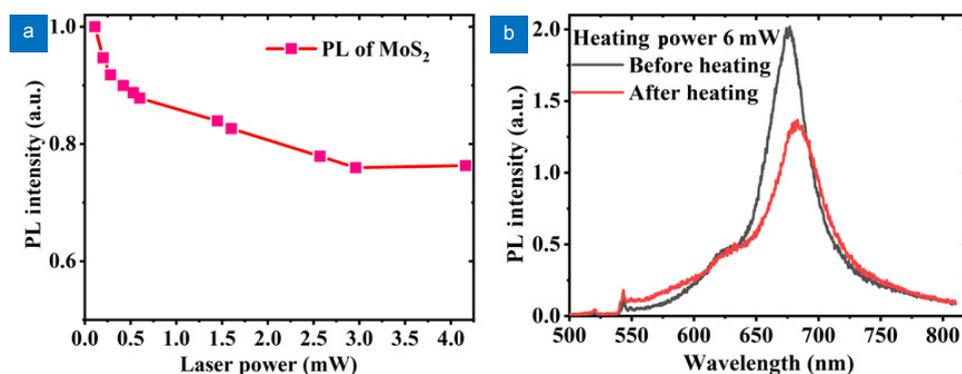


Fig. S2 | (a) PL intensity of pristine MoS₂ monolayer dependence on the laser heating power. The PL intensity reduced about 25% laser heating power from 0.11 mW to 4.2 mW. (b) PL spectra were measured before and after laser with 6 mW heating. Excitation laser wavelength: 532 nm. Excitation power: 110 μ W. Integration time: 0.5 s.

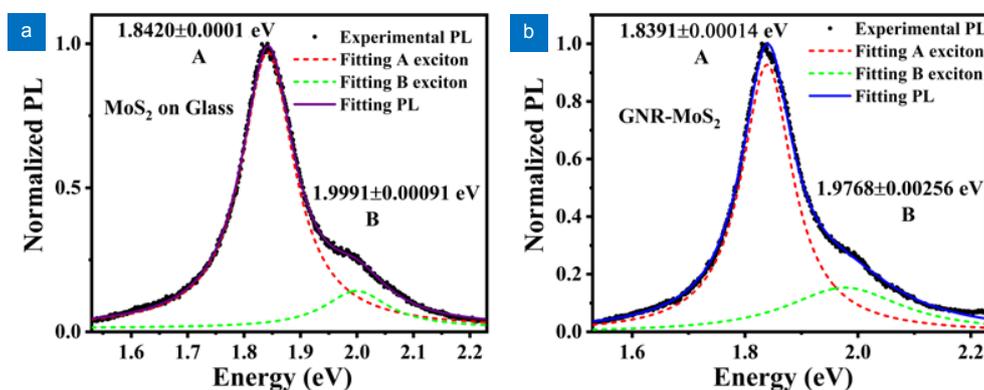


Fig. S3 | Normalized PL spectra from a pristine MoS₂ monolayer on a glass substrate (a) and GNR-MoS₂ hybrid (b). (a) The measured spectra (black dashed lines) were fitted to Lorentz-shape, corresponding to the MoS₂ A and B exciton peaks at 1.8420, and 1.9991 eV, respectively (red and green dashed lines). The purple solid black lines represent the sums of the two exciton components. (b) The measured spectra (black dashed lines) were fitted to Lorentz-shape with peaks at 1.8391, and 1.9768 eV, respectively (red and green dashed lines). The blue solid black lines represent the sums of the two exciton components.

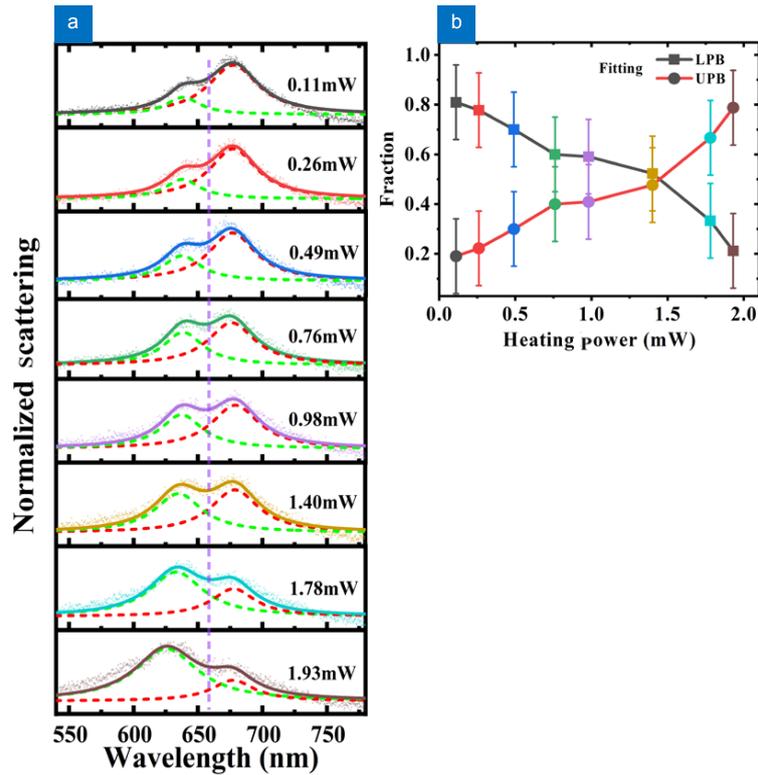


Fig. S4 | Fitting the scattering spectra. (a) The dot curves are scattering spectra of individual GNR coupled to the monolayer MoS₂ flake, and the corresponding solid lines are the fittings with two Lorentz line shape. The vertical purple dashed line in (a) mark A exciton absorption peak. (b) The intensity for the UEB (red) and LEB (black) changes with the heating power. The dot color in (b) corresponding to the color of the scattering spectra in (a).

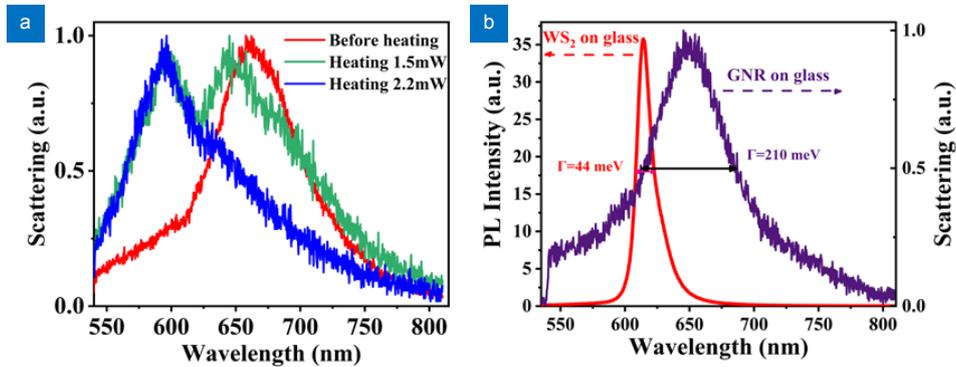


Fig. S5 | Plasmon-exciton coupling in GNRs-WS₂ hybrid. (a) Scattering spectra of GNR-WS₂ hybrid before heating (black curve), after heating with laser power ~1.5 mW (red curve), after heating with laser power ~2.2 mW (blue curve). (b) PL spectra of pristine WS₂ on the glass (red curve) with FWHM ~44 meV and the GNR on the glass (purple curve) with FWHM ~210 meV.

From Fig. S5, a clear anti-crossing dispersion is obtained, which can be fit to the semi-classical coupled oscillator model^{1,2}, as given

$$\omega_{\pm} = \frac{\omega_{\text{pl}} + \omega_{\text{ex}}}{2} \pm \frac{1}{2} \sqrt{\Omega_{\text{R}}^2 + \delta^2}, \quad (1)$$

where ω_{\pm} : the high-energy branch and low energy branch frequency, ω_{ex} : exciton transition frequency, ω_{pl} : the LSP resonant frequency of GNR, $\delta = \omega_{\text{pl}} - \omega_{\text{ex}}$ is the detuning between the plasmon and exciton. A vacuum Rabi splitting of $\Omega_{\text{R}} = 162$ meV was obtained when the detuning is zero. It is larger than the peak width of the exciton ($\gamma_{\text{ex}} = 44$ meV) (Fig. S5(b)) and smaller than the scattering peak width of the uncoupled GNR plasmon ($\gamma_{\text{pl}} = 210$ meV). And it does rigorously sat-

isfy the criterion for strong coupling ($\Omega_R > (\gamma_{pl} + \gamma_{ex})/2$)³. Thus it can indicate that there exists a strong light-matter interaction between single plasmonic nanoresonators and WS₂ monolayer at room temperature.

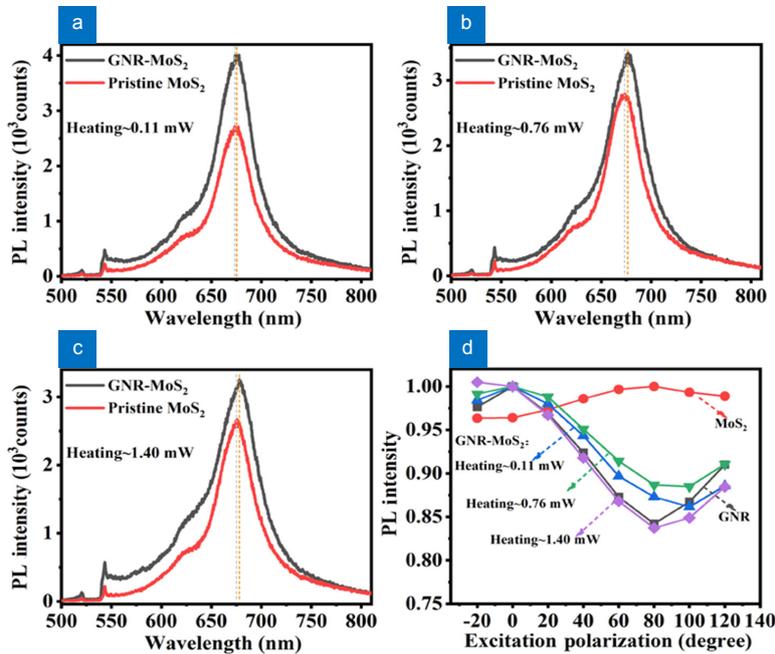


Fig. S6 | Photoluminescence spectra of MoS₂ monolayer without (red) and with GNR (black) during the heating ~0.11 mW (a), ~0.76 mW (b) and ~1.40 mW (c). The grown and orange dashed line in mark A exciton absorption peak of MoS₂ without and with GNR, respectively. Excitation laser wavelength: 532 nm. Excitation power: 110 μ W. Integration time: 0.5 s. (d) The PL intensity as a function of the excitation polarization, single GNR on glass (black), pristine MoS₂ monolayer on glass (red), GNR-MoS₂ hybrid with heating power ~0.11 mW (blue), ~0.76 mW (green) and ~1.40 mW (purple). Zero degree presents the excitation polarization along with the longitudinal directions of the GNR.

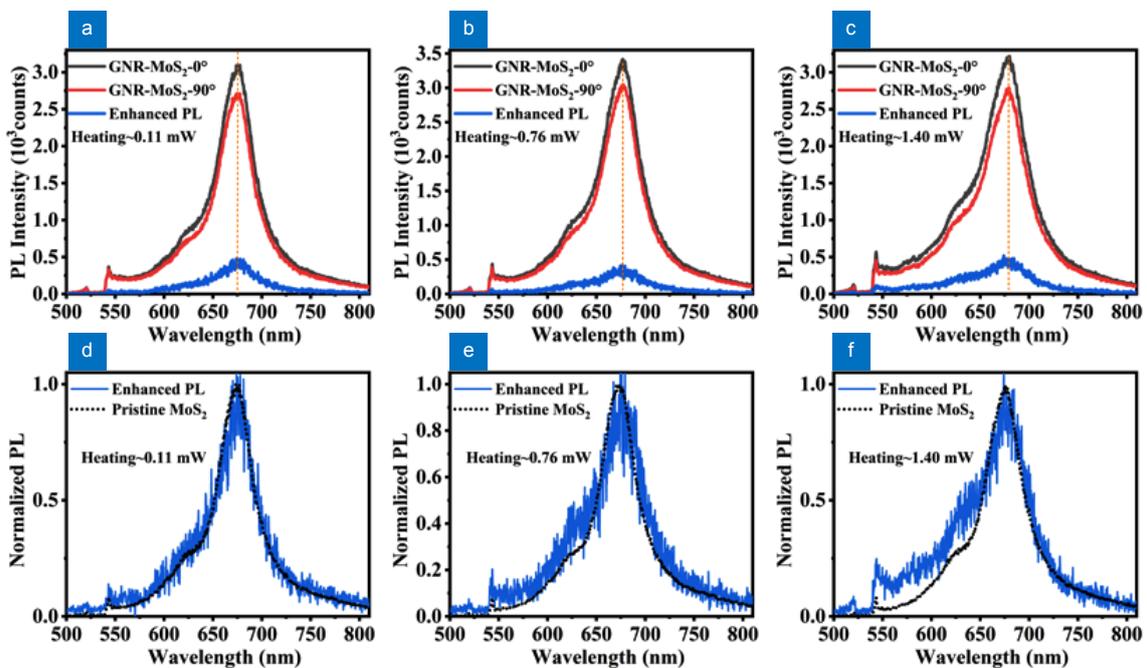


Fig. S7 | The enhanced PL changes during the photothermal reshaping of heating power ~0.11 mW (a), ~0.76 mW (b) and ~1.40 mW (c). The enhanced PL was defined as the PL of excitation polarization at zero degrees minus PL of excitation polarization at ninety degrees. (d-f) Normalized enhanced PL (red line) and normalized PL of pristine MoS₂ (black dashed line).

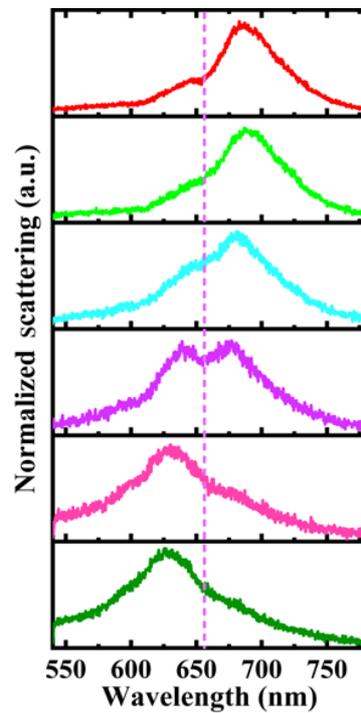


Fig. S8 | Plasmon-exciton coupling in another GNRs–MoS₂ hybrid. The vertical purple dashed line in (a) mark A exciton absorption peak.

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