Significant Photoluminescence Enhancement in WS$_2$

Monolayers through Na$_2$S Treatment

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**Figure S1.** Optical images of samples on sapphire substrates (a) CVD-grown WS$_2$ monolayers before Na$_2$S solution treatment. (b) the same sample after 0.05 M Na$_2$S solution treatment. (c) and (d) show the corresponding AFM images, before and after chemical treatment.

**Figure S2.** (a) AFM images and (b) PL intensity mapping images of WS$_2$ monolayers treated by 0.05 M Na$_2$S solution before and after 350 °C thermal annealing in argon atmosphere for 30 min.
**Figure S3** (a) STEM image of the pristine WS$_2$ monolayers. Inset is the corresponding FFT pattern image. High-angle annular dark field (HAADF) STEM image shows (b) different density of S vacancies and (c) W vacancy.

**Figure S4** PL spectra of WS$_2$ monolayers before and after Na$_2$S treatment (0.05 M) measured at room temperature with the same laser power. (a) and (b) are inner and edge, respectively. (c) and (d) are the corresponding spectra after Na$_2$S treatment (0.05 M).
Figure S5. PL spectra measured at 77K, (a) pristine WS\textsubscript{2} monolayers excited by different laser powers from 0.1 mW to 9.7 mW. (b) PL spectra of WS\textsubscript{2} monolayers after Na\textsubscript{2}S (0.05 M) treatment with excitation laser power from 0.01 mW to 9.7 mW. (c) and (d) Deconvoluted PL spectra at high laser power at inner and edge regions.
Figure S6 WS\textsubscript{2} monolayers before and after 0.05 M Na\textsubscript{2}S solution treatment from 0 days to 15 days. Samples were exposed to ambient condition with relative humidity of 65%. (a) Optical images. (b) Corresponding PL intensity mapping images. (c) Time-dependent PL intensity, the scale bar is 9 \(\mu\)m.
Figure S7 (a) is spatial distribution of the PL intensity mapping images of WS$_2$ monolayers before and after NaCl solution treatment (0.05 M). (b) is corresponding spatial distribution of PL peak position mapping images. (c) and (d) are PL spectra at inner and edge regions of WS$_2$ monolayers before and after chemical treatment by NaCl solution, respectively. The scale bar is 4 μm.
Figure S8. XPS spectra of WS$_2$ monolayers before and after Na$_2$S treatment acquired with Al Kα line.
Figure S9 (a) and (c) AFM images of pristine and Na$_2$S treated (0.05 M) WS$_2$ monolayers, respectively. The scale bar is 5 $\mu$m. (b) and (d) Corresponding KPFM images of regions marked in Figure (a) and (c). Lines scan along the white line show a relative CPD of -178.9 mV and 52.8 mV, respectively.