## ELECTRONIC SUPPLEMENTARY INFORMATION (ESI)

## DNA-Driven Dynamic Assembly of MoS<sub>2</sub> Nanosheets

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**ESI Fig. 1** Raman spectrum of MoS<sub>2</sub>-bulk showing the typical in-plane vibration mode  $(E_{2g}^{1})$  at 383 cm<sup>-1</sup> and the out-ofplane mode  $(A_{1g})$  among the S atoms at 408 cm<sup>-1</sup>.



ESI Fig. 2 Schematic illustration of DNA functionalization on MoS<sub>2</sub>-NS surface employing a thiolated-DNA strand.



**ESI Fig. 3** UV absorbance spectra of (a) thiolated (1) DNA sequence in **M1**. Calculated extinction coefficient: 415600 L/(mole\*cm); Estimated concentration of (1) in **M1**: 0.54 μM. (b) thiolated (2) DNA sequence in **M2**. Calculated extinction coefficient: 301800 L/(mole\*cm); Estimated concentration of (2) in **M2**: 0.65 μM.



**ESI Fig. 4** AFM topographical images and respective height profiles of: (a) a solution containing **M1** and **M3**, average  $MoS_2$  nanosheets height = 7.5 ± 3.1 nm, and (b) a solution containing  $MoS_2$ -NS and amino-modified DNA strand (2), not capable of functionalizing  $MoS_2$ -NS, average  $MoS_2$  nanosheets height: 7.9 ± 2.8 nm. In both samples, no evidence of assembly was observed.



ESI Fig. 5 UV absorbance spectrum of thiolated (3) DNA sequence in M3. Calculated extinction coefficient: 285000 L/(mole\*cm), estimated concentration of (3) in M3=  $0.71 \mu$ M.



**ESI Fig. 6** Raman spectra of an **M2/M3** solution (a) at pH= 8, showing  $E_{2g}^{1}$ =383 cm<sup>-1</sup> and  $A_{1g}$ = 408 cm<sup>-1</sup>; (b) at pH= 5.5, showing  $E_{2g}^{1}$ = 385 cm<sup>-1</sup> and  $A_{1g}$ = 410 cm<sup>-1</sup>.



ESI Fig. 7 Raman spectrum of an M2/M3 solution after addition of (4), showing  $E_{2g}^{1}$ = 377 cm<sup>-1</sup> and  $A_{1g}$ = 402 cm<sup>-1</sup>.



ESI Fig. 8 AFM topographical image and respective height profile of an M2/M3 solution after addition of DNA strand (I); average MoS<sub>2</sub> nanosheets height: 39.6 ± 13.5 nm. No evidence of disassembly was observed.