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Electronic Supplementary Information

Ultrasensitive molecular sensing of few-layer niobium diselenide

Qian Lv,[a†] Xi Wu,[b†] Junyang Tan,[b] Bilu Liu,[b] Lin Gan,[b] Jia Li,*[b] Zheng-Hong Huang,[ac] Feiyu Kang[bc] and Ruitao Lv*[ac]

[a] State Key Laboratory of New Ceramics and Fine Processing, School of Materials Science and Engineering, Tsinghua University, Beijing 100084, China

[b] Shenzhen Geim Graphene Center, Tsinghua Shenzhen International Graduate School, Tsinghua University, Shenzhen 518055, China

[c] Key Laboratory of Advanced Materials (MOE), School of Materials Science and Engineering Tsinghua University, Beijing 100084, China

[†] These authors contributed equally to this work.

*Email: lvruitao@tsinghua.edu.cn; li.jia@sz.tsinghua.edu.cn

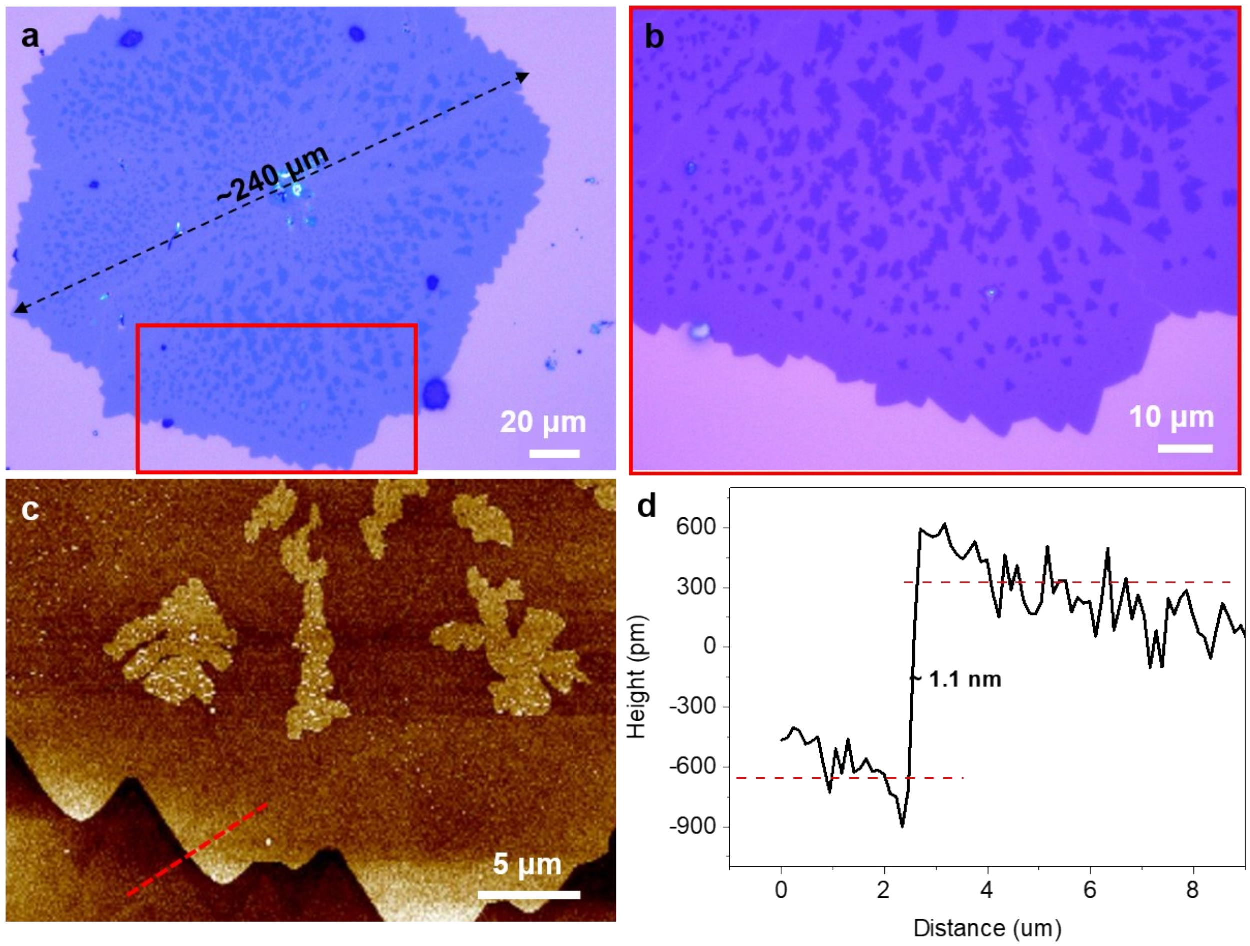

Fig. S1. (a, b) Typical optical images of as-grown large-area $NbSe_2$. (c) Atomic force microscope (AFM) image and (d) the corresponding height profile of as-grown $NbSe_2$. The small irregular areas on top are the second $NbSe_2$ layers.

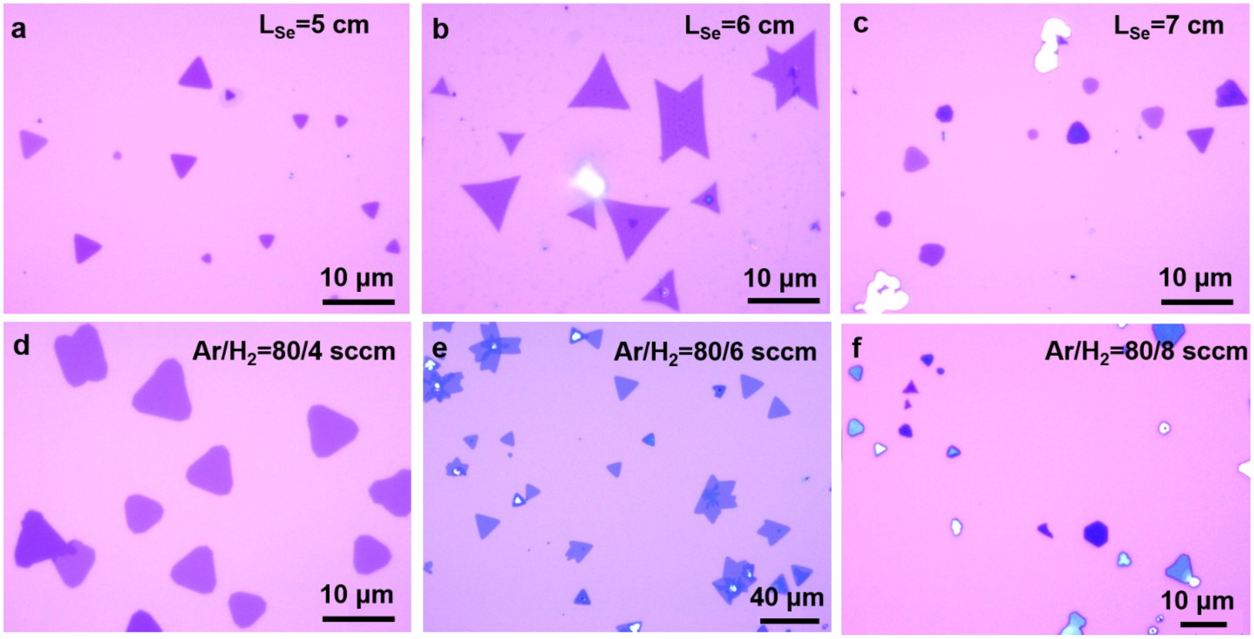

Fig. S2. Optical images of $NbSe_2$ flakes synthesized at different conditions: (a-c) at different L_{Se} values. Here L_{Se} denotes the distance of Se source from the left end of the furnace (as shown in Figure 1). (d-f) $NbSe_2$ flakes synthesized with different H_2 contents in the carrier gas (Ar/H_2 mixture). Here Ar/H_2=80/4 denotes that the Ar flow and H_2 flow are 80 sccm and 4 sccm, respectively. The others are named in the same rule.

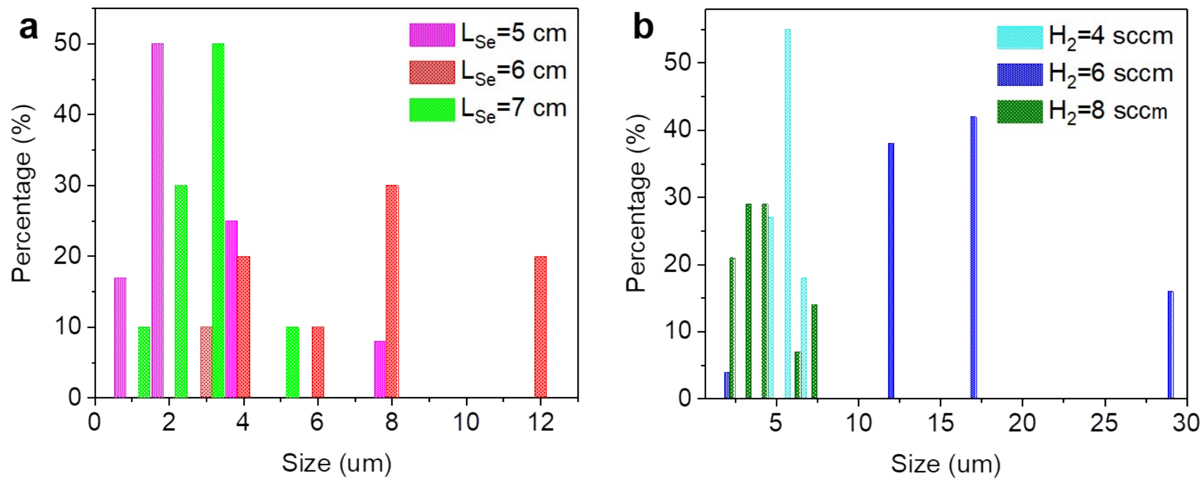

Fig. S3. Size distributions of as-grown $NbSe_2$ with different (a) L_{Se} values and (b) hydrogen flows.

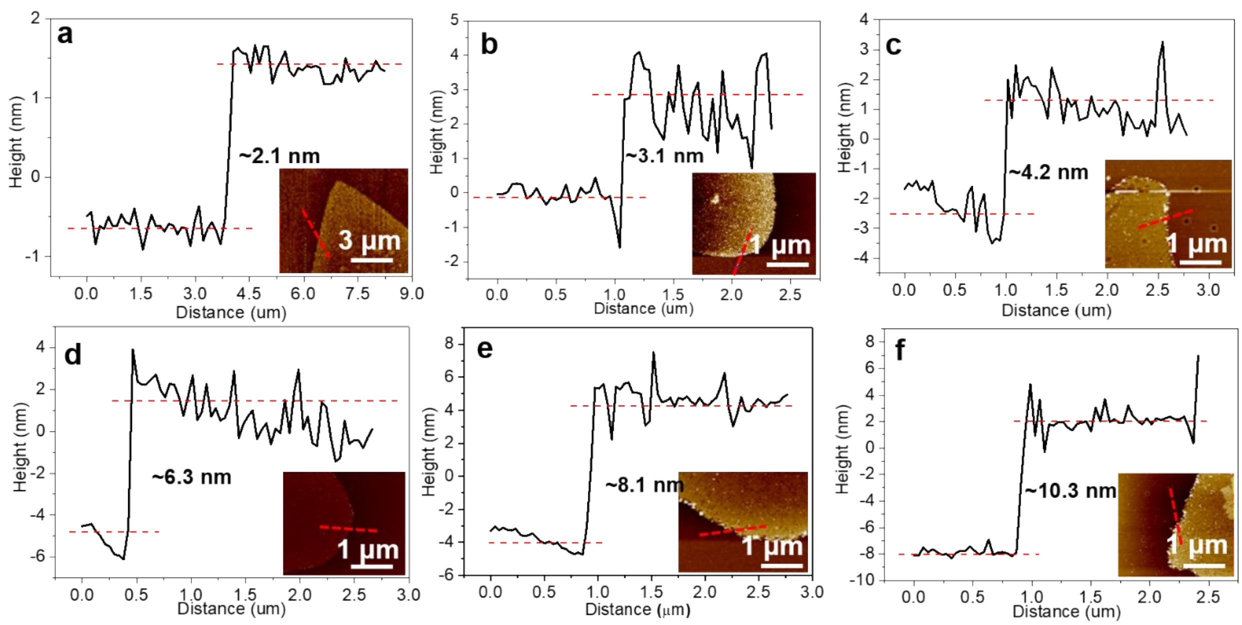

Fig. S4. AFM height profiles and the images (inset of each profile of each panel) of as-grown $NbSe_2$ with tunable thicknesses.

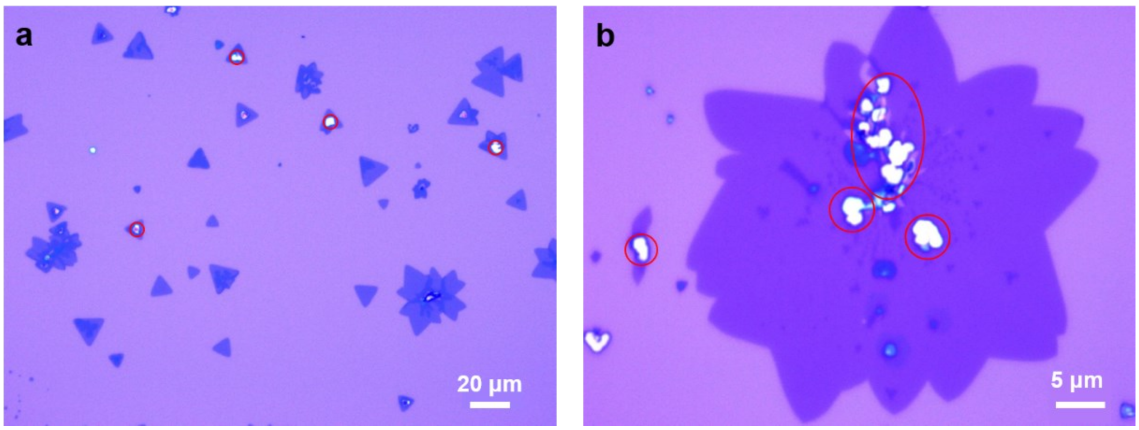

Fig. S5. Optical images showing the residual Nb_2O_5 particles on as-grown $NbSe_2$ flakes marked by red circles.

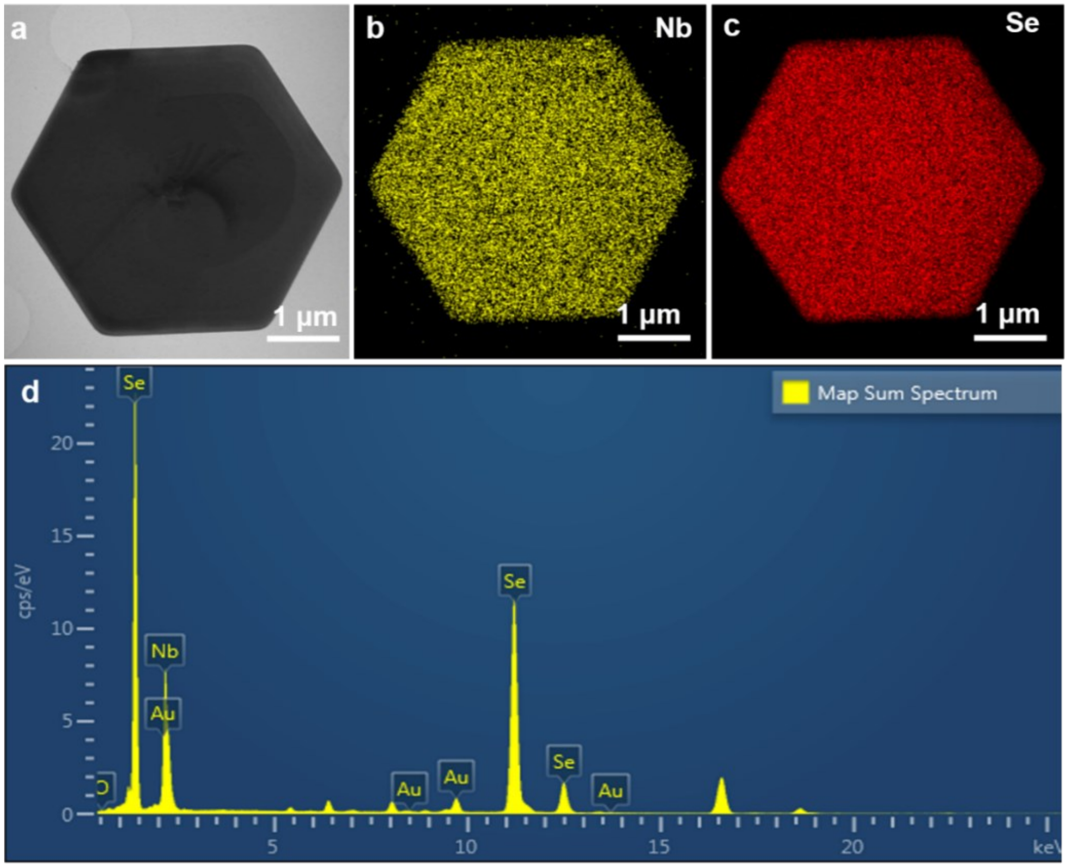

Fig. S6. Energy-dispersive X-ray spectrometer (EDS) elemental mapping of as-grown $NbSe_2$. (a) Bright field image of $NbSe_2$ on Au quantifoil grid collected in the scanning-transmission electron microscope (STEM) mode and the corresponding EDS mappings of (b) Nb and (c) Se, respectively. (d) EDS spectra of $NbSe_2$.

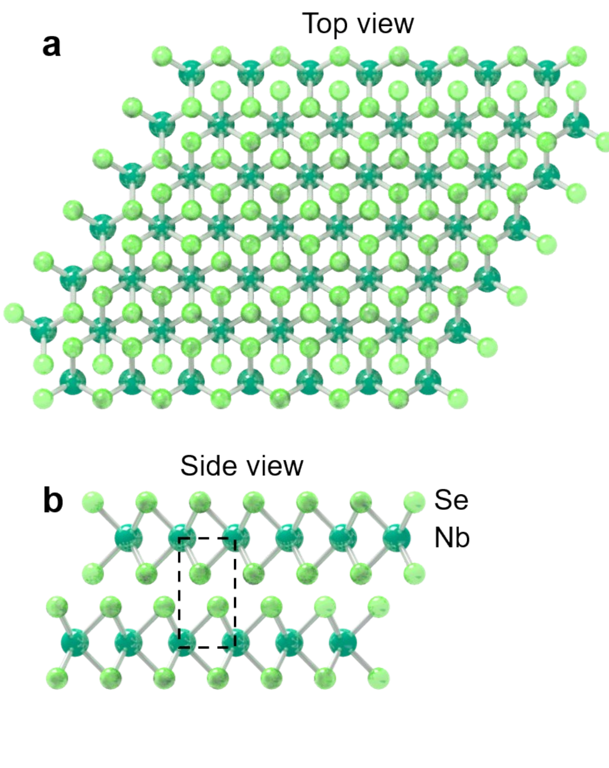

Fig. S7. Atomic structures of (a, b) 2H$_a$-NbSe$_2$ with A'B stacking mode and (c, d) 3R-NbSe$_2$ with AA stacking mode at top and side views.

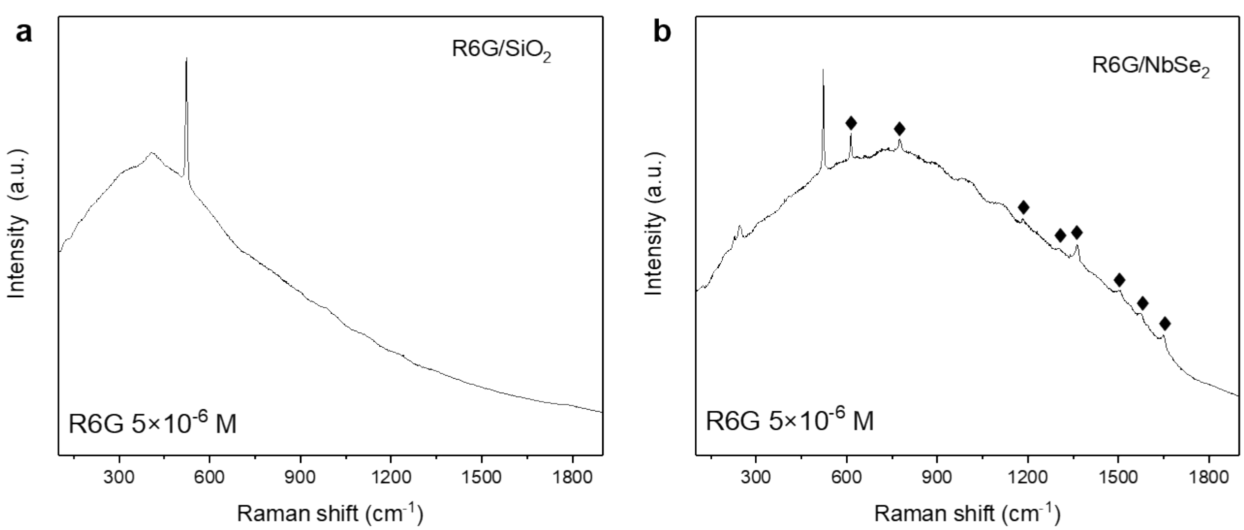

Fig. S8. Comparison of Raman spectra of R6G on bare SiO_2/Si and $NbSe_2$.

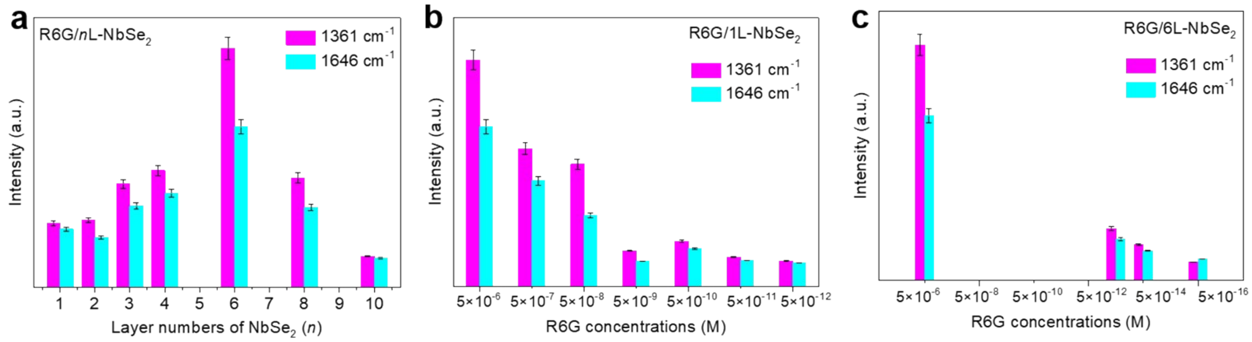

Fig. S9. (a) Raman intensities at 1361 and 1646 cm^{-1} of R6G on $NbSe_2$ with different layers, (b, c) Raman intensities at 1361 and 1646 cm^{-1} of R6G on (b) 1L-$NbSe_2$ and (c) 6L-$NbSe_2$ with different concentrations.

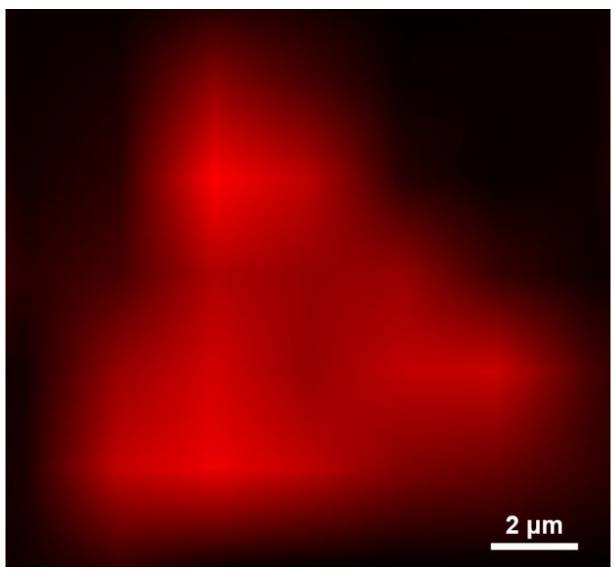

Fig. S10. Raman intensity mapping of R6G (5×10^{-6} M) at 1646 cm^{-1} on $NbSe_2$.

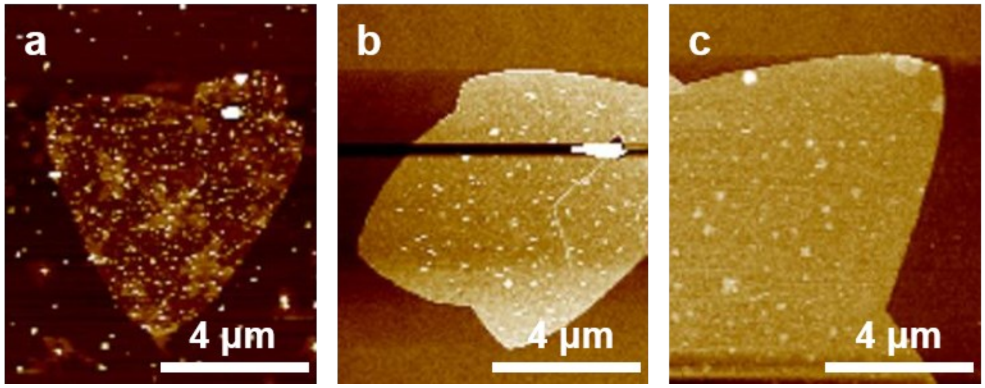

Fig. S11. AFM images of R6G on $NbSe_2$ with different concentrations: (a) 5×10^{-5} M, (b) 5×10^{-6} M and (c) 5×10^{-11} M.

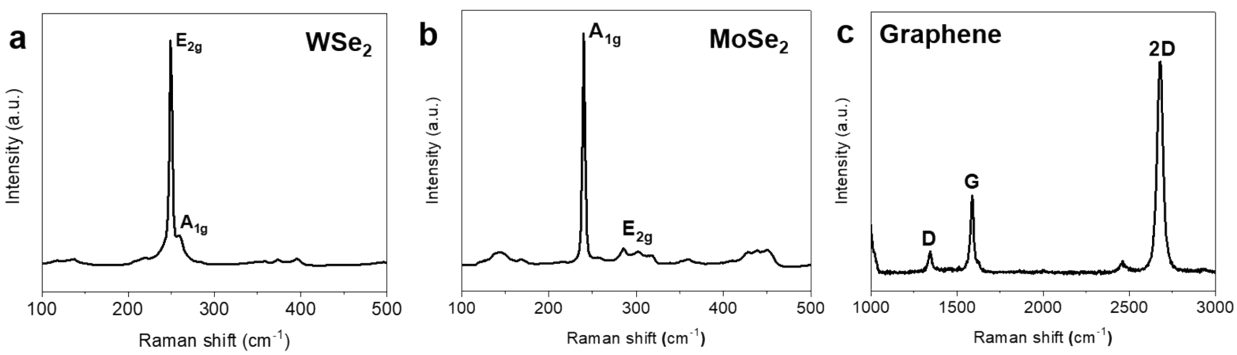

Fig. S12. Characterizations of different 2D materials grown by atmospheric pressure chemical vapor deposition (AP-CVD). Raman spectra of (a) WSe_2, (b) $MoSe_2$ and (c) graphene.

Table S1. Comparison of molecular sensing properties of different materials as substrates

Types	Substrates	Probe molecules	Limits of detection/M	Laser excitation wavelength/nm	Data source
Non-noble metal-based materials	$NbSe_2$	R6G	$\mathbf{5\times10^{-16}}$	532	**This work**
	NbS_2	MeB	1×10^{-14}	633	1
	1T'-WTe_2	R6G	4×10^{-14}	532	2
	1T'-$MoTe_2$	R6G	4×10^{-13}	532	2
	MoS_2/graphene	R6G	5×10^{-12}	532	3
	1T-MoS_2	R6G	1×10^{-9}	532	4
	Few-layer MoS_2	R6G	1×10^{-9}	532	5
	$W_{18}O_{49}/MoS_2$	R6G	1×10^{-9}	532	6
	1T'-ReS_2	R6G	1×10^{-9}	532	7
	NG[a]	R6G	1×10^{-9}	514	8
	$MnPS_{2.4}Se_{0.6}$	R6G	1×10^{-8}	532	9
	1T-$MoSe_2$	R6G	1×10^{-8}	532	10
	Oxygen-incorporated MoS_2	R6G	1×10^{-7}	532	11
	2H-MoS_2	R6G	5×10^{-6}	532	3
Noble-metal-based materials	Au/h-BN/Ag	R6G	1×10^{-12}	514	12
	Ag NP-VE/OCT[b]	R6G	1×10^{-9}	633	13
	Au@Ag/3D-Si	R6G	1×10^{-8}	532	14

Note: [a)] NG: nitrogen-doped graphene; [b)] Ag NP-VE/OCT: Ag nanoparticles-vitamin E-1-octanol.

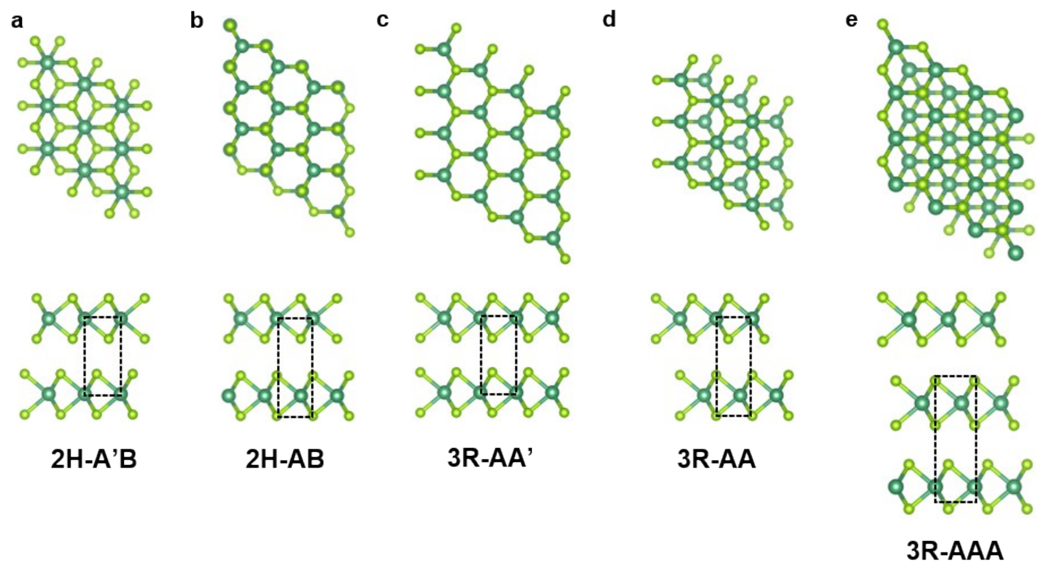

Fig. S13. Structural polytypes of few-layer $NbSe_2$ with different stacking modes. (a) 2H-A'B. (b) 2H-AB. (c) 3R-AA' phase. (d) 3R-AA. (e) 3R-AAA.

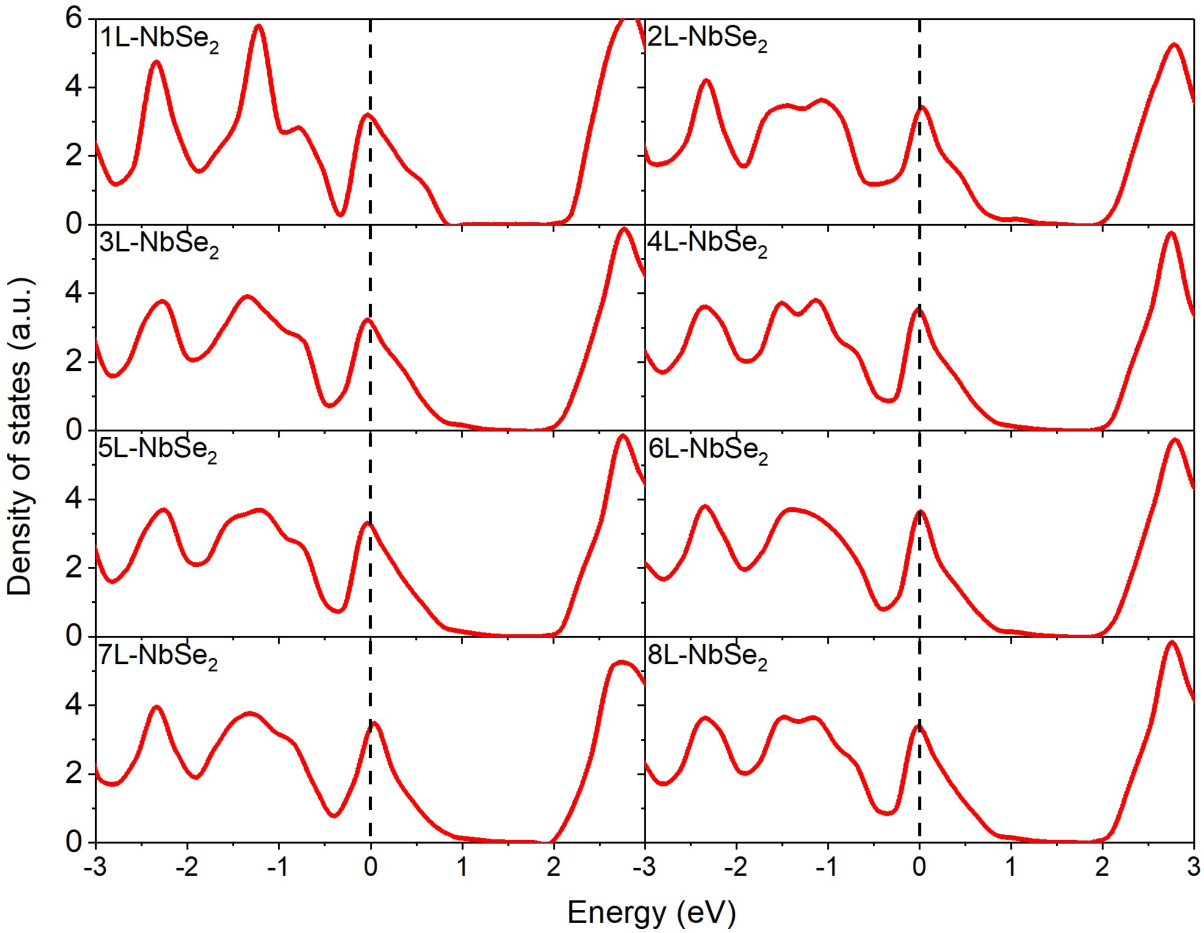

Fig. S14. Density of states (DOS) of the outmost layer $NbSe_2$ with $2H_a$-A’B stacking mode.

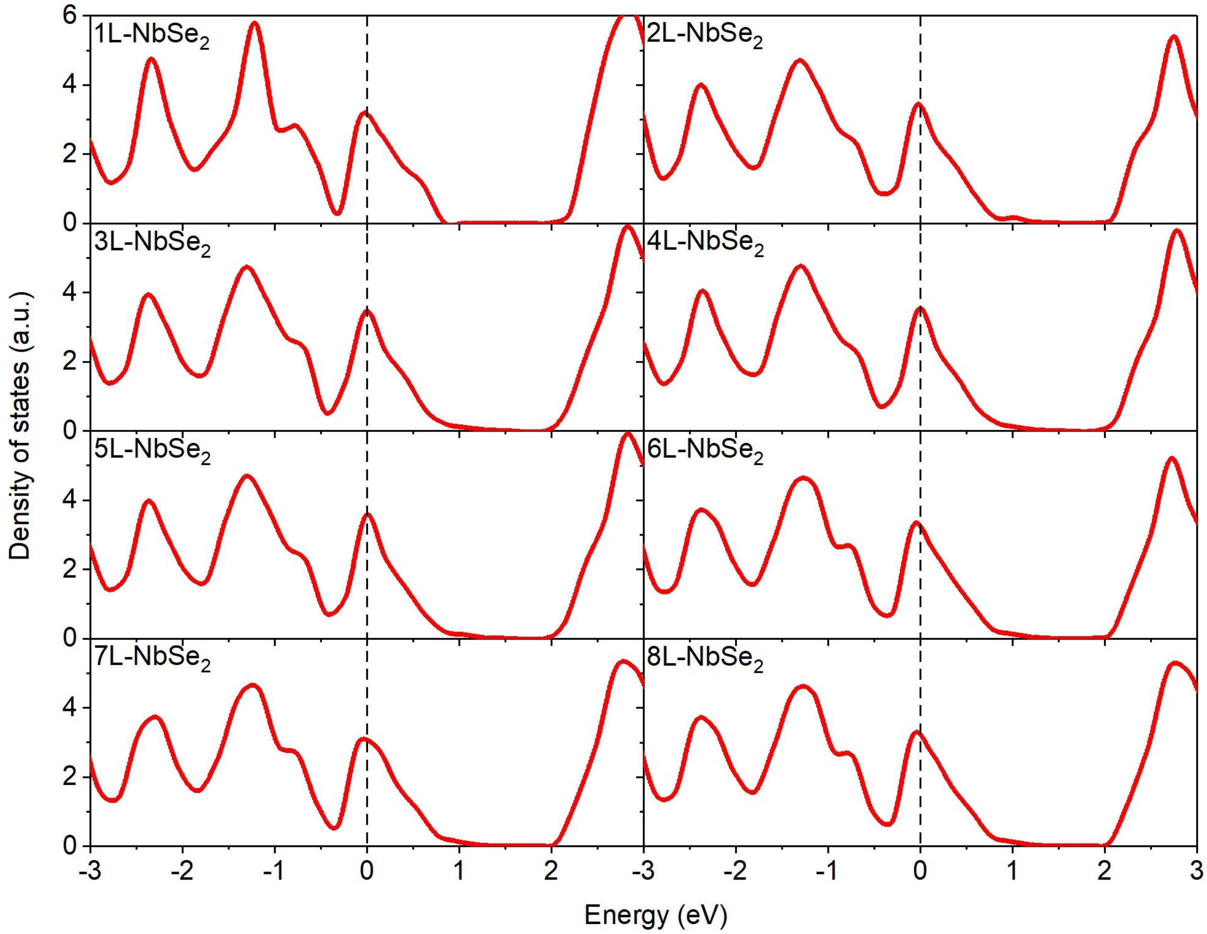

Fig. S15. DOS of the outmost layer of $NbSe_2$ with 3R-AA stacking mode.

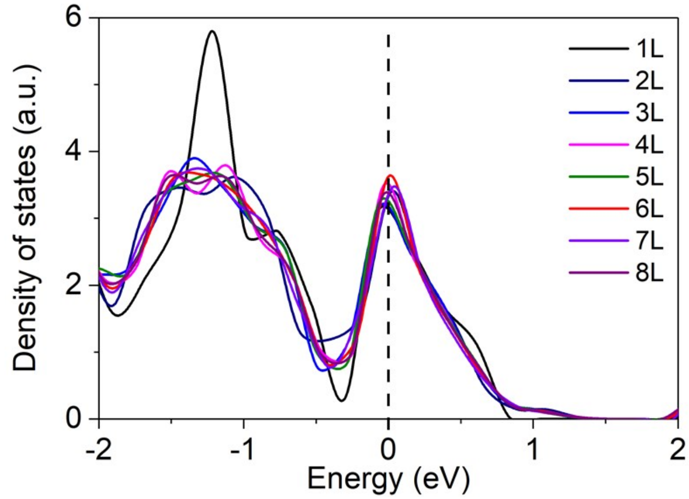

Fig. S16. DOS of the outmost layer of $NbSe_2$ with $2H_a$-A'B stacking modes.

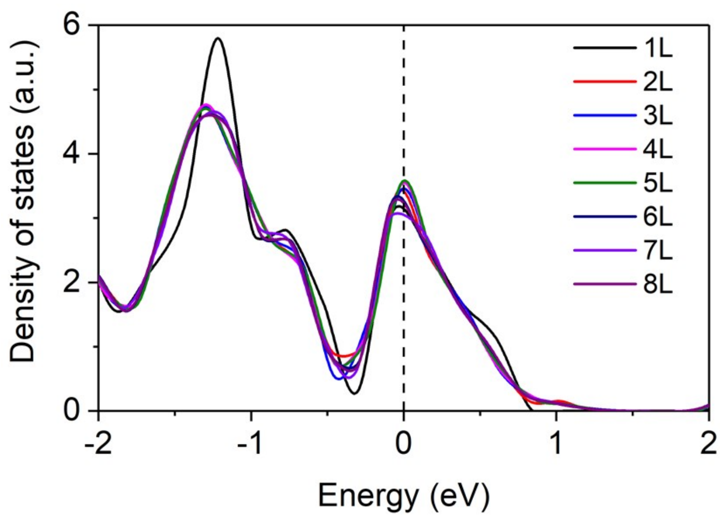

Fig. S17. DOS of the outmost layer of $NbSe_2$ with 3R-AA stacking modes.

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