Supplementary Materials for

"Tunability of the Superconductivity of $NbSe_2$ Films Grown by Two-Step Vapour Deposition"

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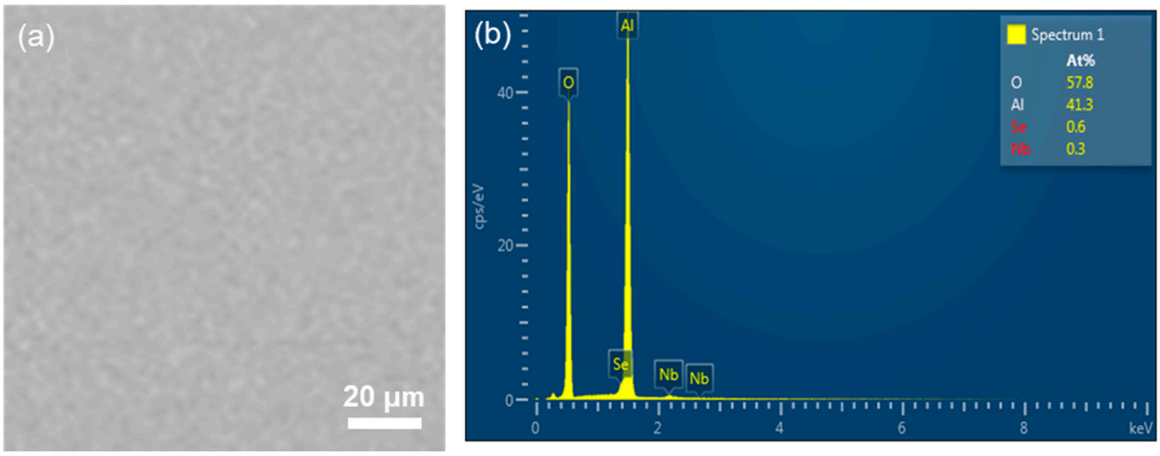
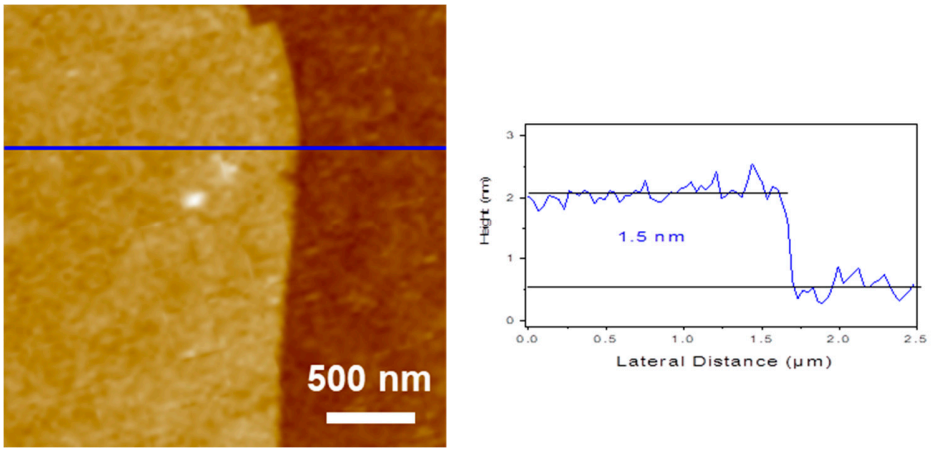
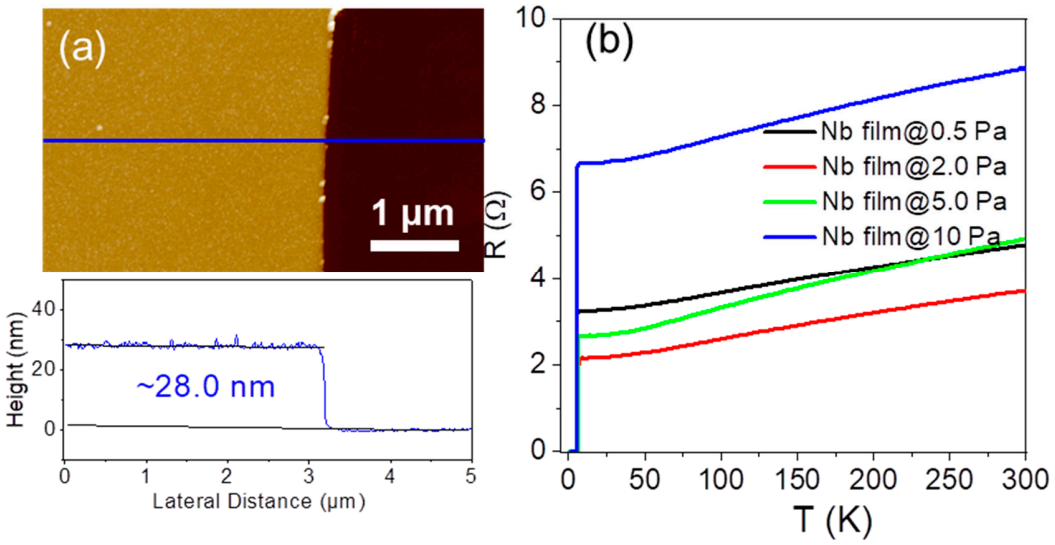


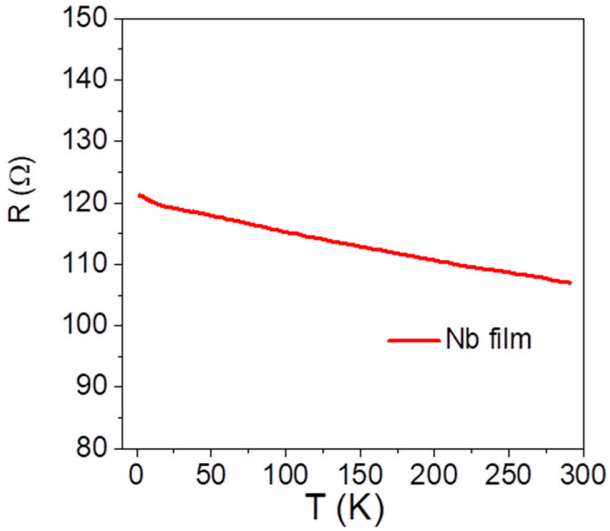
Supplementary Figure S1 Elemental analysis of as-grown $NbSe_2$ film. (a) SEM image and (b) EDS spectra of as-grown $NbSe_2$ film collected from a 100 µm × 100 µm region in Figure S1a.



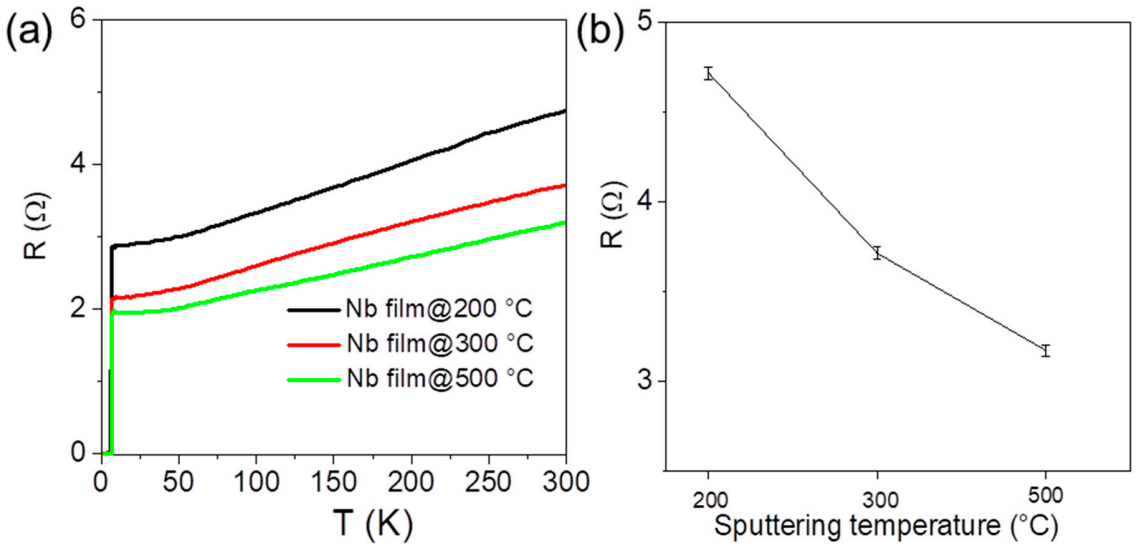
Supplementary Figure S2 AFM characterization of as-sputtered Nb film. AFM image and the corresponding height profile of a Nb film sputtered on sapphire.



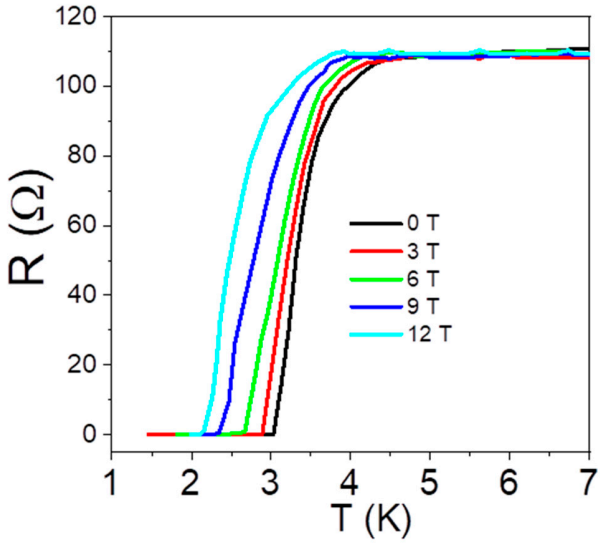
Supplementary Figure S3 AFM characterization of a 28 nm Nb film and transport measurement of as-sputtered Nb films at the different sputtering pressure. (a) AFM image and the corresponding height profile of an as-sputtered Nb film; (b) Superconductivity of as-sputtered Nb films at the different sputtering pressure. Note: The superconducting transition critical temperature (T_c): is the temperature where the sheet resistance drops to 10% of its normal state.



Supplementary Figure S4 Transport property of a 5.5 nm Nb. Temperature dependent resistance of a 5.5 nm Nb film, the film shows semimetal behavior (dR/dT < 0) and without the appearance of superconductivity.



Supplementary Figure S5 Transport measurement of as-sputtered Nb films at the different sputtering temperature. (a) Superconductivity of as-sputtered Nb films at different sputtering temperature; (b) Resistance of as-sputtered Nb films at the different sputtering temperature (5 different sites are tested for every sample at room temperature).



Supplementary Figure S6 Superconducting resistivity transition of the $NbSe_2$ film under an in-plane magnetic field.

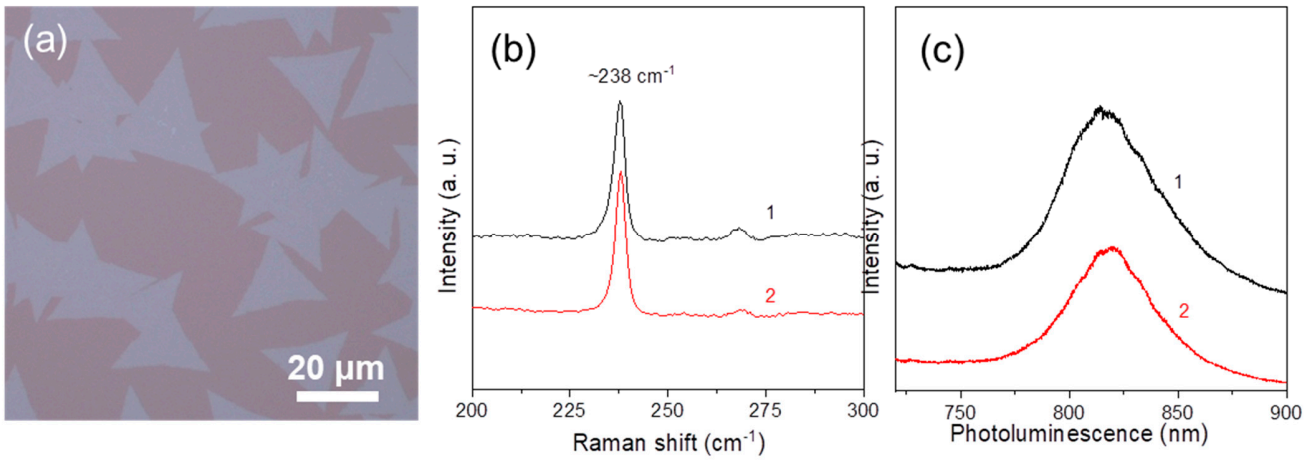
As described by the Ginzburg-Landau (GL) theory of 2D superconductors, coherence length and effective thickness can be further derived from $H_{c2\parallel}$ and $H_{c2\perp}$ with the following relations:

$$\boldsymbol{H_{c2,\perp} = \frac{\Phi_0}{2\pi\xi_{GL}^2(0)}\left(1-\frac{T}{T_c}\right)}, \qquad \boldsymbol{H_{c2,\parallel} = \frac{\Phi_0\sqrt{12}}{2\pi\xi_{GL}^2(0)d_{SC}}\left(1-\frac{T}{T_c}\right)^{\frac{1}{2}}}$$

where $\xi_{GL}(0)$ is the zero-temperature GL in-plane coherence length, Φ_0 is the magnetic flux quantum, and d_{SC} is the out-of-plane superconducting effective thickness. From the fitting, the coherent length is determined to be ~6.5 nm. An effective thickness of ~3.2 nm is obtained, which is smaller than $\xi_{GL}(0)$ and revealed 2D behavior of the superconductivity in the as-grown $NbSe_2$ film.

Supplementary Table S1 The T_c of monolayer$NbSe_2$ by different preparation methods.

Preparation method	Layers/ L	Protective layer	T_c/ K	Refs.
Tape exfoliation	1	Thin h-BN layer	~ 2.9	Nat. Phys. 2016, 12, 139. [14]
MBE	1	~ 10 nm amorphous Se layer	~ 0.50	Nat. Phys. 2016, 12, 92. [33]
MBE	1	20 nm amorphous Se layer	~ 0.65	Nano lett. 2017, 17, 6802. [20]
MBE	1	~ 20 nm amorphous Se layer	~ 1.0	Nature Physics, 2019, 15, 904. [1]
Electrochemical exfoliation	1	Thin h-BN layer	~3.2	Nat. Mater. 2021, 20, 181 [27]
Salt-assisted CVD	1	Graphene	~ 1.0	Nat. Commun. 2017, 8, 394 [22]
Two-step vapour deposition method	1	None	~ 2.0	This work



Supplementary Figure S7 Monolayer $MoSe_2$ crystals grown by Two-Step Vapor Deposition. (a) Optical microscopic image, (b) Raman spectra and (c) photoluminescence (PL) spectra of monolayer $MoSe_2$ crystals.

[1] Zhao, K. ; Lin, H. ; Xiao, X. ; Huang, W. ; Yao, W. and Yan, M. Disorder-induced multifractal superconductivity in monolayer niobium dichalcogenides. Nat. Phys. 2019,15, 904-910