Supplementary Information for

Observation of Superconductivity in Pressurized 2M WSe_2 Crystals

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1. Materials and methods

(1) Synthesis of 2M WSe_2 crystals

We took a topochemical method to prepare the 2M WSe_2 crystals. Firstly, the precursor K_xWSe_2 crystals were synthesized by the reaction of potassium and WSe_2 powder at 800 ^oC for two days. Then the products were soaked in distilled water to dissolve the impurities. Finally, the $K_x(H_2O)WSe_2$ crystals were stirred in the 2 mmol/L acidic $K_2Cr_2O_7$ solution for 0.5 h to deintercalate K^+ ions. The prepared 2M WSe_2 crystals were washed by distilled water for some times and dried under vacuum.

(2) Single crystal determination

Suitable crystals were chosen to perform the data collections. Single-crystal X-ray diffraction was performed on a Bruker D8 QUEST diffractometer equipped with Mo Kα radiation (λ = 0.71073 Å). The diffraction data were collected at room temperature by the ω- and φ-scan methods. The crystal structures were solved and refined using APEX3 program. Absorption corrections were performed using the multi-scan method (SADABS).

(3) Raman measurements

Raman spectra were measured using a Renishaw inVia Qontor confocal Raman microscope at excitation wavelength of 633 nm. To avoid the phase transition, the exciting laser cannot be strong and the parameter of the ND filter is set as 10%.

(4) High-pressure measurements

Electrical transport measurements were carried out using standard four-point probe method in a homemade multifunctional measurement system (2.6-300 K, JANIS Research Company Inc.; 0-9 T, Cryomagnetics Inc.). In situ high pressure angle-dispersive synchrotron XRD experiment was performed at 4W2 beamline of Beijing Synchrotron Radiation Facility (BSRF) (λ = 0.6199 Å). All high pressure measurements were conducted by diamond-anvil cell (DAC) and rhenium as gasket material. DAC made of non-magnetic Cu-Be alloy was used in the upper critical magnetic field measurement. A 4 : 1 methanol–ethanol mixture was adopted as pressure transmitting medium in XRD performance, while there was no pressure transmitting medium in resistance measurements. The R_1 fluorescence line of ruby was used to calibrate pressures in lower ranges; the Raman spectrum of diamond was employed as

pressure calibrant at higher region.

2. Supplementary tables and figures

Table S1. Crystal Data and Structural Refinement statistics for 2M WSe_2.

formula	WSe_2
F_w (g mol^{-1})	341.76
crystal system	Monoclinic
space group	*C*2/m
a (Å)	13.838(3)
b (Å)	3.291(7)
c (Å)	5.912(1)
α (°)	90
β (°)	111.468(6)
γ (°)	90
V (Å^3)	250.55(9)
Z	1
ρ_c (g·cm^{-3})	9.059
μ (mm^{-1})	74.75
λ (Mo *K*α) (Å)	0.71073
T (K)	298
F(000)	568
θ_{max} (°) / completeness (%)	25.0 / 97.4
R_{int}	0.041
R_1[a][$F^2 > 2\sigma(F^2)$]	0.030
wR_2[b](F^2)	0.095
goodness of fit	0.85
largest diff. peak and hole (e/Å^3)	2.14 and -1.09

[a]$R_1 = \sum||F_o| - |F_c||/\sum|F_o|$. [b]$wR_2=\{\sum[w(F_o^2 - F_c^2)^2]/\sum[w(F_o^2)^2]\}^{1/2}$, $w = 1/[\sigma^2(F_o^2) + (0.1000P)^2]$ where $P = (F_o^2 + 2F_c^2)/3$.

Table S2. Fractional atomic coordinates and isotropic or equivalent isotropic displacement parameters (Å^2) for 2M WSe_2.

atom	*x*	*y*	*z*	U_{iso}*/U_{eq}
W	0.25685 (5)	1/2	0.19725 (10)	0.0296 (4)
Se1	0.14190 (13)	0	0.3296 (3)	0.0305 (6)
Se2	0.39518 (13)	0	0.2055 (3)	0.0317 (5)

Table S3. Atomic displacement parameters (Å^2) for WSe_2.

	U_{11}	U_{22}	U_{33}	U_{12}	U_{13}	U_{23}
W	0.0379 (6)	0.0319 (6)	0.0204 (6)	0	0.0124 (4)	0
Se1	0.0359 (10)	0.0342 (10)	0.0227 (10)	0	0.0124 (7)	0
Se2	0.0386 (10)	0.0335 (11)	0.0245 (10)	0	0.0134 (7)	0

Table S4. Selected bond distances (Å) and bond angles (°) for 2M WSe_2

Bond distance (Å)		Bond distance (Å)	
W—Se2^{i}	2.5112 (13)	W—W^{ii}	2.8034 (10)
W—Se2	2.5112 (13)	W—W^{iv}	2.8034 (10)
W—Se2^{ii}	2.5353 (18)	Se1—W^{v}	2.6002 (13)
W—Se1^{i}	2.6002 (13)	Se1—W^{iii}	2.6302 (17)
W—Se1	2.6003 (13)	Se2—W^{v}	2.5112 (13)
W—Se1^{iii}	2.6301 (17)	Se2—W^{ii}	2.5353 (18)
bond angle (°)		bond angle (°)	
Se2^{i}—W—Se2	81.88 (5)	Se2^{ii}—W—W^{ii}	55.85 (3)
Se2^{i}—W—Se2^{ii}	112.51 (5)	Se1^{i}—W—W^{ii}	139.40 (6)
Se2—W—Se2^{ii}	112.51 (5)	Se1—W—W^{ii}	90.94 (4)
Se2^{i}—W—Se1^{i}	97.20 (4)	Se1^{iii}—W—W^{ii}	137.07 (3)
Se2—W—Se1^{i}	162.70 (7)	Se2^{i}—W—W^{iv}	56.66 (4)
Se2^{ii}—W—Se1^{i}	83.91 (6)	Se2—W—W^{iv}	102.69 (4)
Se2^{i}—W—Se1	162.70 (7)	Se2^{ii}—W—W^{iv}	55.85 (3)
Se2—W—Se1	97.20 (4)	Se1^{i}—W—W^{iv}	90.94 (4)
Se2^{ii}—W—Se1	83.91 (6)	Se1—W—W^{iv}	139.40 (6)
Se1^{i}—W—Se1	78.52 (5)	Se1^{iii}—W—W^{iv}	137.07 (3)
Se2^{i}—W—Se1^{iii}	82.75 (6)	W^{ii}—W—W^{iv}	71.89 (3)
Se2—W—Se1^{iii}	82.75 (6)	W^{v}—Se1—W	78.52 (5)
Se2^{ii}—W—Se1^{iii}	159.17 (6)	W^{v}—Se1—W^{iii}	100.00 (5)
Se1^{i}—W—Se1^{iii}	80.00 (5)	W—Se1—W^{iii}	100.00 (5)
Se1—W—Se1^{iii}	80.00 (5)	W—Se2—W^{v}	81.88 (5)
Se2^{i}—W—W^{ii}	102.69 (4)	W—Se2—W^{ii}	67.49 (5)
Se2—W—W^{ii}	56.66 (4)	W^{v}—Se2—W^{ii}	67.49 (5)

Symmetry codes: (i) *y*-1, *x*, *z*; (ii) -*y*+1, *x*-*y*+1, *z*; (iii) -*x*+*y*-1, -*x*, *z*; (iv) -*x*+*y*, -*x*+1, *z*; (v) -*y*, *x*-*y*+1, *z*; (vi) -*x*+*y*, -*x*, *z*; (vii) -*y*, *x*-*y*, *z*.

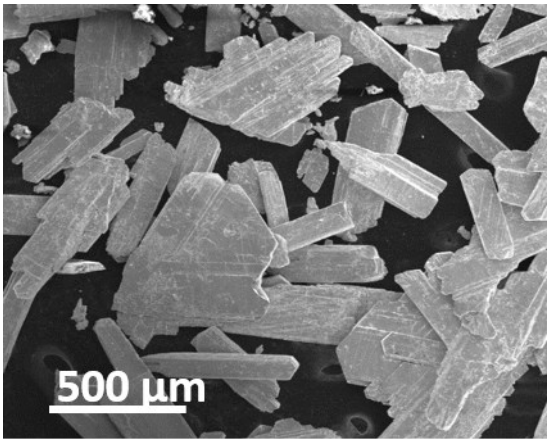

Figure S1. The scanning electron microscopy image of 2M WSe_2 crystals.

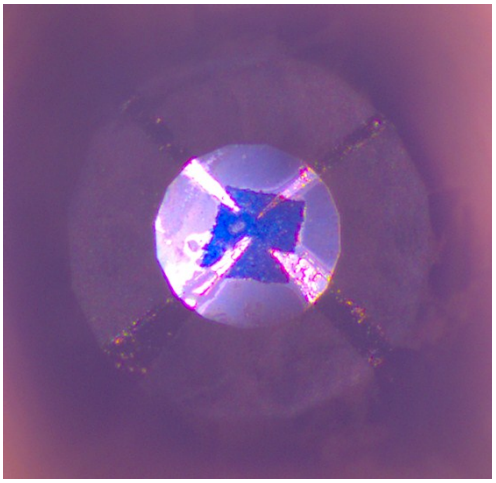

Fig S2. The micrograph of sample in contact with four electrodes inside the DAC

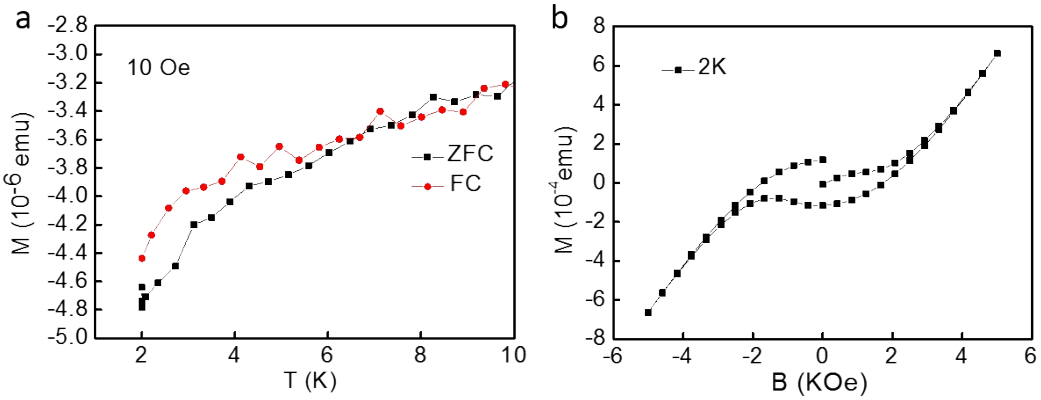

Figure S3 (a) temperature dependence of magnetization in the ZFC and FC modes for 2M WSe_2 under 8.1 GPa. (b) Magnetization vs magnetic field at 2K.

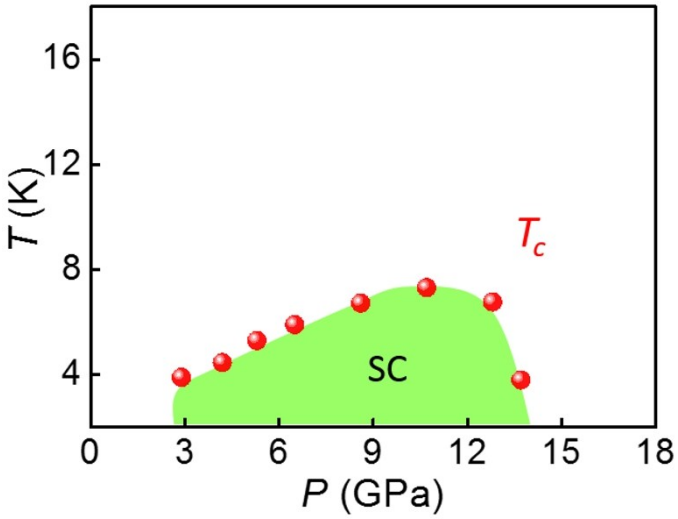

Figure S4. The pressure dependent temperature phase diagram of 2M WSe_2

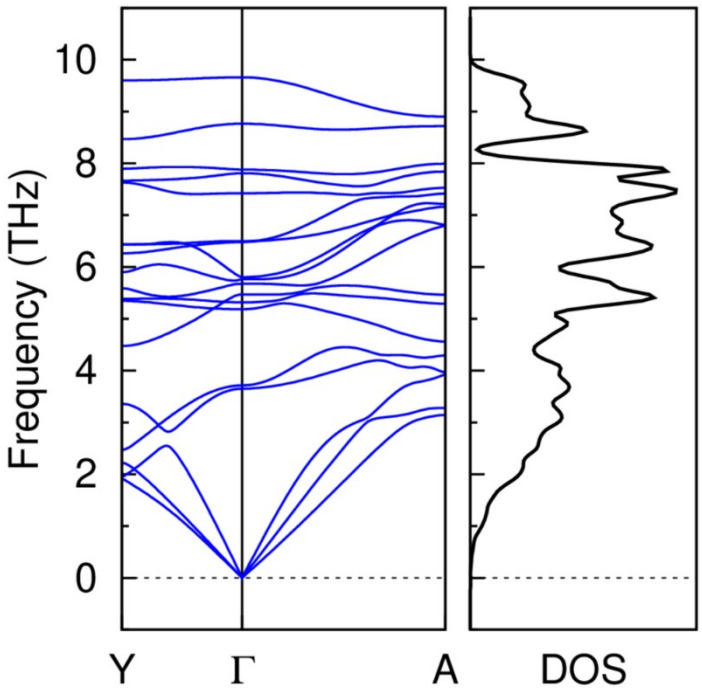

Figure S5. Phonon calculation of 2M WSe_2 at 13.4 GPa.

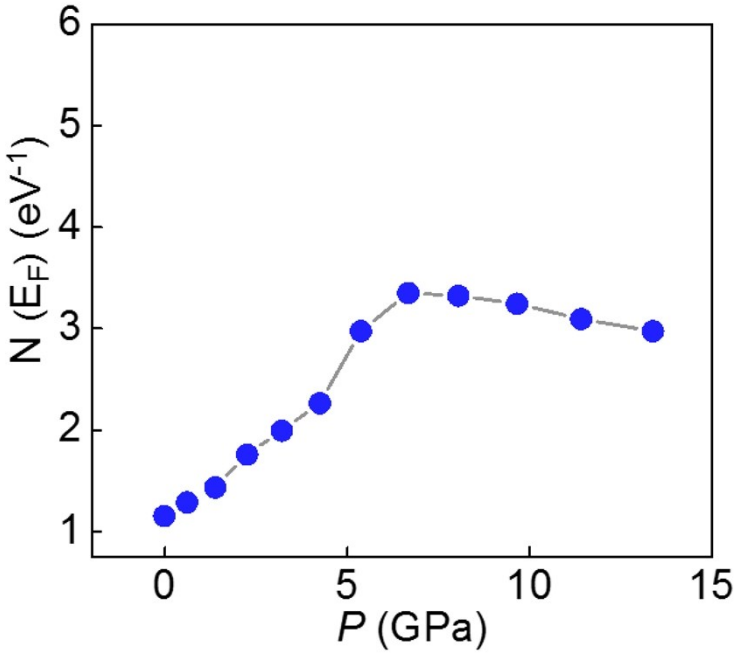

Figure S6. The density of states (DOS) at the Fermi level calculated under pressure from 0 to 14GPa.