Nominal Kagome Antiferromagnetic Mn_3Sn: Effects of excess Mn and its novel synthesis method

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S1.1. Synthesis of Mn_3Sn single crystals

Except for initial trials using the Sn self-flux method, Bi flux was utilized to recrystallize Mn_3Sn from a precursor or to synthesize Mn_3Sn directly from Mn and Sn elements. High-purity Mn (99.95%, Alfa Aesar), Sn (99.99+%, Alfa Aesar), and Bi (99.997%, Alfa Aesar) were prepared in various atomic ratios and temperature conditions (see **Table S1**). They were loaded into an alumina crucible, sealed in a quartz ampoule under a partial Ar atmosphere, and subjected to different heating processes. The resulting Mn_3Sn crystals generally exhibited hexagonal needle- or rod-like morphologies, as displayed in **Fig. S1**. We optimized single-crystalline Mn_3Sn samples by recrystallizing the precursors with an atomic ratio of 3:1:4 (that is, Mn_3Sn:Bi = 1:4).

S1.2. Identification

We conducted X-ray diffraction (XRD, PANalytical Empyrean) to examine the sample quality and identify the crystallographic axes. Energy dispersive X-ray spectroscopy (EDS) using a scanning electron microscope (SEM, Hitachi S-4800) was employed to determine the chemical composition of the grown single crystals.

S1.3. Characterization of magnetization

To evaluate the magnetic properties of nominal samples, we used a Magnetic Property Measurement System (MPMS, Quantum Design). A home-built resistivity measurement system with a cryocooler and a Physical Property Measurement System (PPMS, Quantum Design) were used to examine the electrical transport properties. The heat capacity of the sample was also examined by utilizing the PPMS.

S1.4. STM measurements

Scanning tunneling microscopy (STM) experiments were conducted using a closed-cycle cryogen-free STM machine (PanScan Freedom, RHK Technology). The Mn_3Sn single crystal was cleaved in an ultra-high vacuum (UHV) chamber under a pressure below 1×10^{-10} Torr. The cleaved sample exhibited a flat surface with a metallic luster and was immediately transferred to the STM head for measurement. A mechanically sharpened iridium (Ir) wire was used as the STM tip. The tip stability and shape were verified on Cu(111), which was prepared through multiple cycles of Ar sputtering and annealing at 793 K. To obtain differential conductance (dI/dV) spectra, we used a standard lock-in technique with a modulation frequency of $f = 732$ Hz and an amplitude of $V_{mod} = 20$ mV. All STM data were acquired at 15 K.

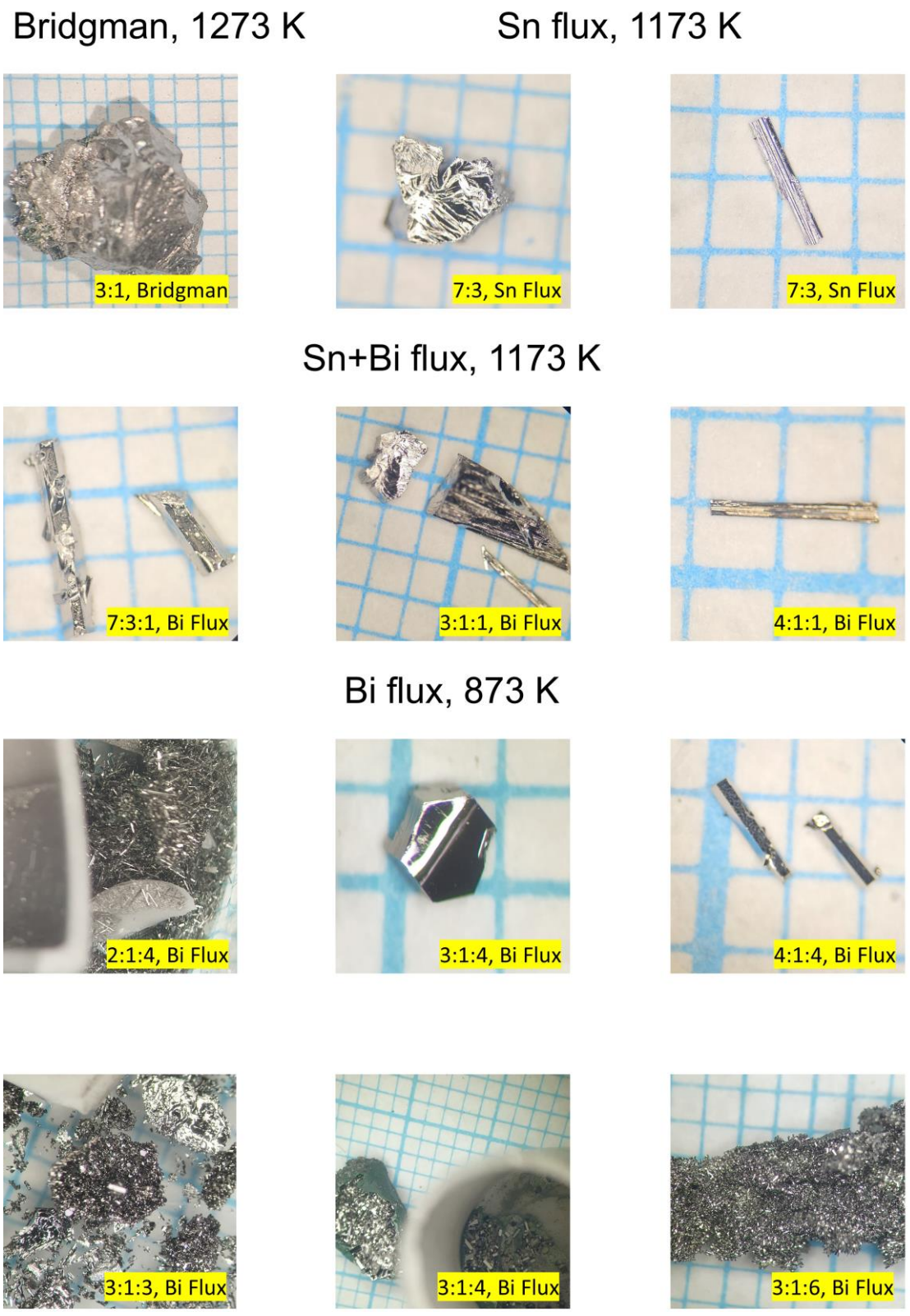

Fig. S1. Optical microscopy of Mn_3Sn single crystals grown under the synthesis conditions described in **Table S1**.

Table S1. Synthesis conditions of single-crystalline Mn_3Sn samples.

Atomic ratio Mn	Sn	Bi	heating (K)	cooling (K)	growth time (h)	method
3	1	0	1000	-	-	Bridgman method, 2mm/h
7	3	0	1100	900	200	Sn flux method
7	3	1	1100	900	200	Bi flux method
3	1	1	1100	900	200	Bi flux method
4	1	1	1100	900	200	Bi flux method
2	1	4	800	600	200	Bi flux method
3	1	2	800	600	200	Bi flux method + Mn_3Sn precursor
3	1	4	800	600	200	Bi flux method + Mn_3Sn precursor
3	1	6	800	600	200	Bi flux method + Mn_3Sn precursor
4	1	4	800	600	200	Bi flux method

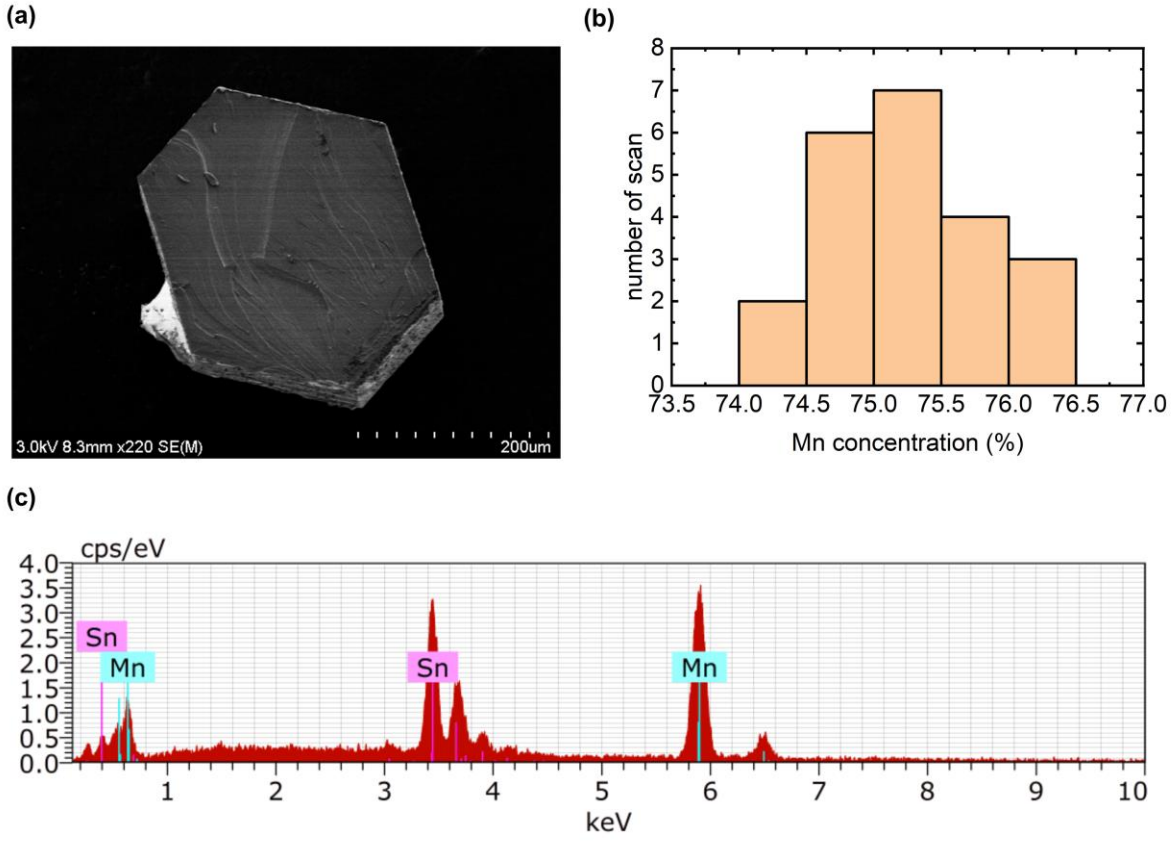

Fig. S2. (a) SEM image of the cross-sectioned Mn_3Sn single crystal. It reproducibly exhibits terrace-like surfaces. (b) Histogram of collected EDS data for the specific cross-sectioned sample. (c) Obtained EDS signals and the corresponding atoms matched to the peaks.

Fig. S2 presents the chemical composition of a single-crystalline Bi flux-grown Mn_3Sn sample. A total of 22 data points was collected to determine the average chemical composition, where detailed Mn values are provided in Table S2. The averaged composition was found to be $Mn_{75.3(6)}Sn_{24.7(6)}$, which corresponds to the $Mn_{3+x}Sn_{1-x}$ formulation with x = 0.012 (i.e., $Mn_{3.012}Sn_{0.988}$). In this context, we confirmed the chemical composition of the sample as $Mn_{3.01}Sn_{0.99}$ with x = 0.01, indicating a slight excess of Mn but a near-stoichiometric nature of the sample.

Table S2. EDS data points collected from a single-crystalline Mn_3Sn sample. The data are sorted in ascending order of Mn atomic composition and were used to generate the histogram shown in Fig. S2b.

Mn (%)	#1	#2	#3	#4	#5	#6	#7
74.0-74.5	74.28	74.46					
74.5-75.0	74.59	74.61	74.79	74.82	74.91	74.99	
75.0-75.5	75.07	75.22	75.28	75.3	75.37	75.43	75.44
75.5-76.0	75.68	75.69	75.85	75.87			
76.0-76.5	76.15	76.36	76.39				

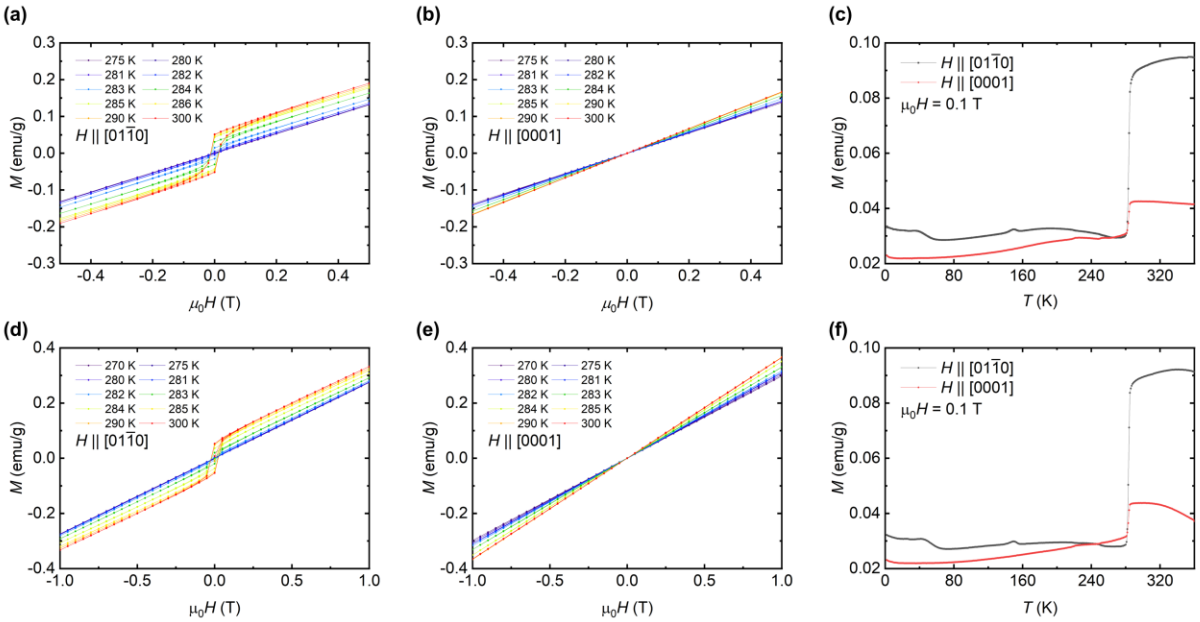

Fig. S3. Magnetic behavior of Mn_3Sn single crystal grown at the same batch. Magnetic hysteresis measured along the $[01\bar{1}0]$ direction and (b) [0001] direction. (c) Temperature dependence of the magnetization. (d-f) Same graphs to (a-c) but measured on different Mn_3Sn single crystal. Note that the crossover point can differ subtly by measurements.

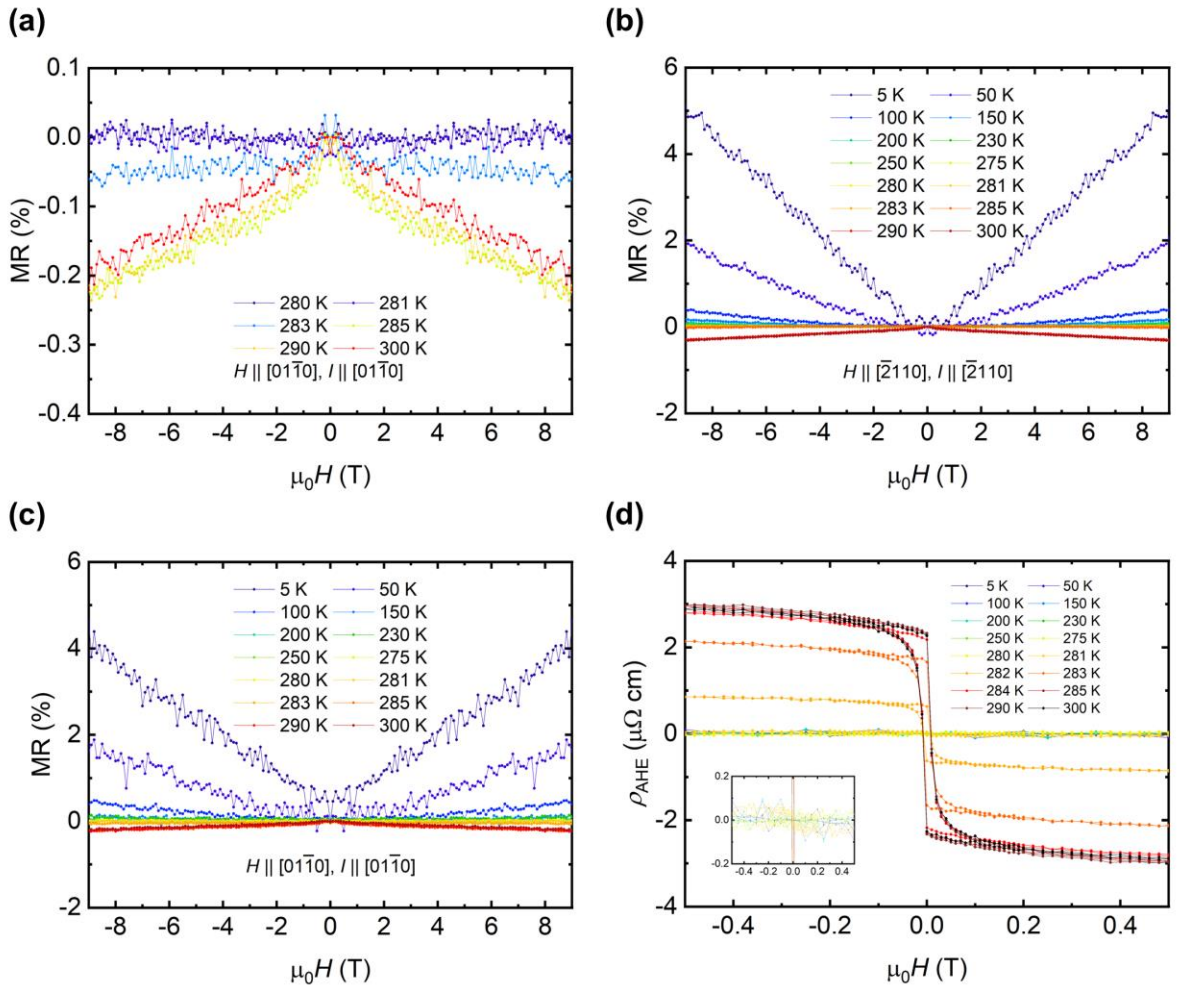

Fig. S4. (a) Chiral anomaly-driven negative magnetoresistance in the longitudinal sample along the $[01\bar{1}0]$ direction. (b-c) Raw data of the longitudinal magnetoresistance measured along the (b) $[\bar{2}110]$ and (c) $[01\bar{1}0]$ directions. (d) Raw data of the Hall resistivity between 5 K and 300 K. (Inset) Enlarged data with the absence of hysteresis under T = 280 K.

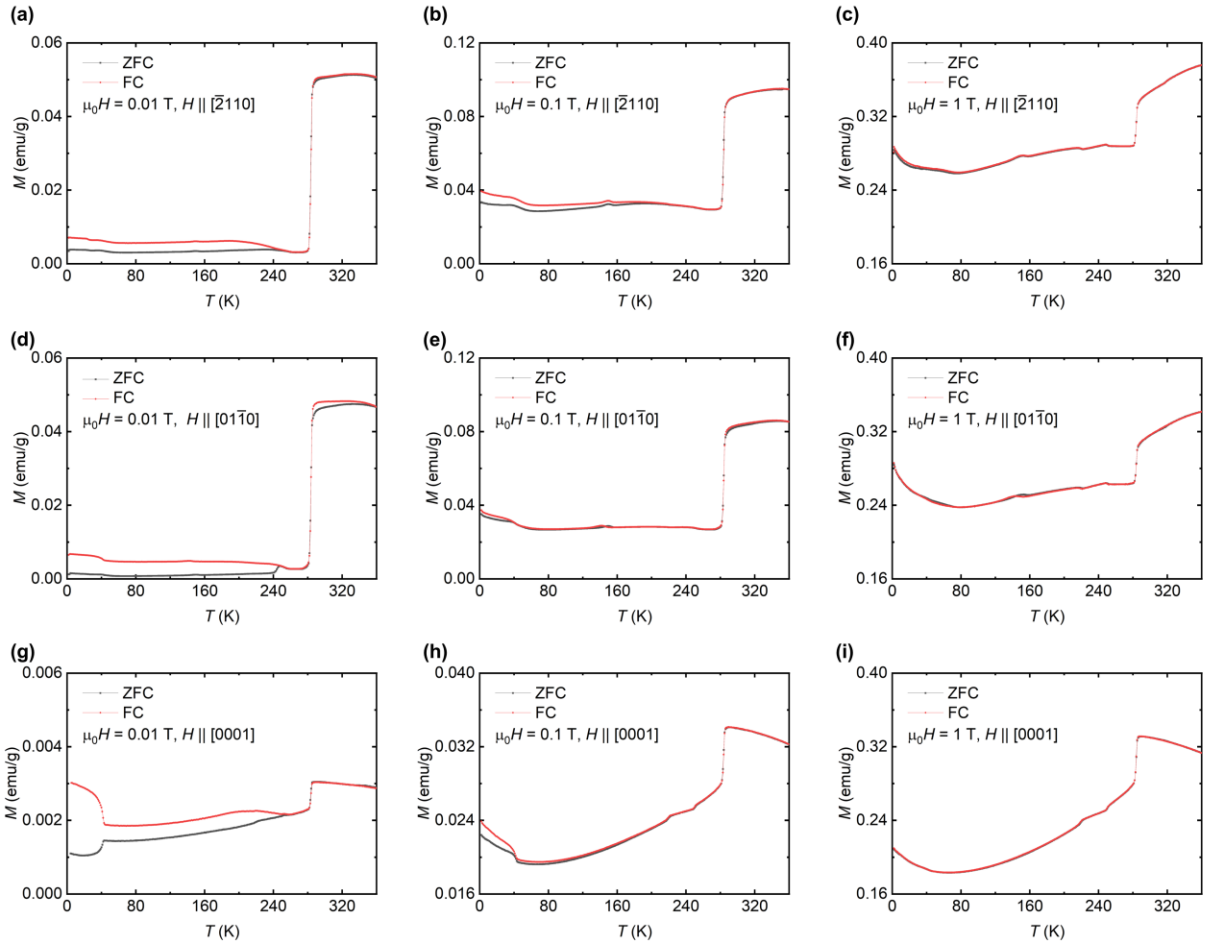

Fig. S5. Isofield magnetization with ZFC and FC curves by applying magnetic fields along the (a-c) [$\bar{2}$110] direction, (d-f) [01$\bar{1}$0] direction and (g-i) [0001] direction. Note that the sample in (a-c) is used to show the magnetic behaviors of Mn_3Sn single crystals in this and is different from the sample used to describe (d-f) and (g-i), which is used in the main text.

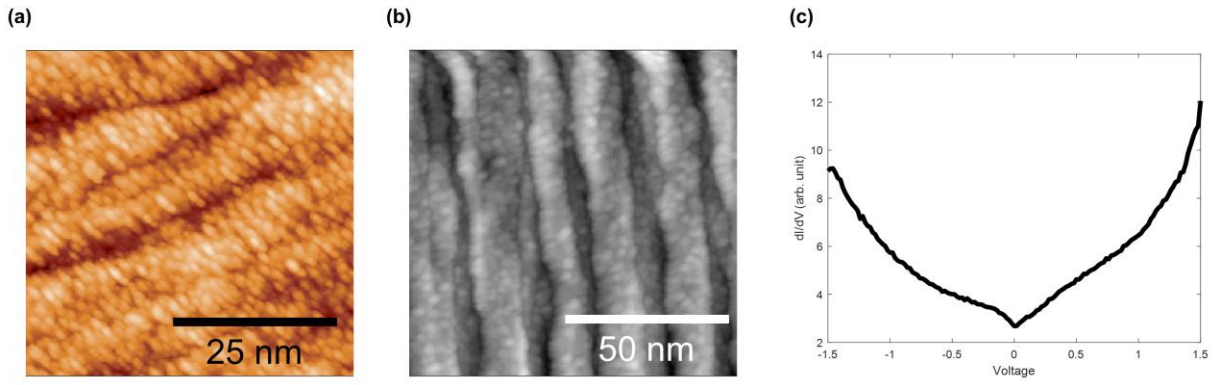

Fig. S6. Topography measured under the same conditions (V_s = – 1.0 V and I_t = – 200 pA) but with different sizes (a) 50 nm × 50 nm (b) 100 nm × 100 nm. (c) dI/dV curve for other area in the cleaved surface of Mn_3Sn.