Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information:

Single Crystal Synthesis and Properties of the Two-Dimensional van der Waals Frustrated Magnets, $Mn_2In_2Se_5$ and $Mn_2Ga_2S_5$

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Table S1. Single Crystal XRD data and structure refinement for $Mn_2Ga_2S_5$

Report date	2022-06-22
Identification code	Mn2Ga2S5
Empirical formula	Ga2 Mn2 S5
Molecular formula	Ga2 Mn2 S5
Formula weight	409.62
Temperature	100.0 K
Wavelength	0.71073 Å
Crystal system	Trigonal
Space group	P-3m1
Unit cell dimensions	a = 3.7096(2) Å a= 90°.
	b = 3.7096(2) Å b= 90°.
	c = 15.1880(11) Å g = 120°.
Volume	181.00(2) $Å^3$
Z	1
Density (calculated)	3.758 Mg/m^3
Absorption coefficient	12.099 mm^{-1}
F(000)	192
Crystal size	0.056 x 0.044 x 0.038 mm^3
Crystal color, habit	Orange Plate
Theta range for data collection	4.025 to 30.494°.
Index ranges	-5<=h<=5, -5<=k<=5, -21<=l<=21
Reflections collected	4696
Independent reflections	268 [R(int) = 0.0560, R(sigma) = 0.0194]
Completeness to theta = 25.000°	98.8 %
Absorption correction	Semi-empirical from equivalents
Max. and min. transmission	0.0511 and 0.0219
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	268 / 0 / 16
Goodness-of-fit on F^2	1.057
Final R indices [I>2sigma(I)]	R1 = 0.0320, wR2 = 0.0817
R indices (all data)	R1 = 0.0392, wR2 = 0.0866
Extinction coefficient	n/a

Largest diff. peak and hole	1.378 and -0.886 e.Å^{-3}

Table S2. Single crystal atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (Å^{2}x 10^3) for $Mn_2Ga_2S_5$. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

Atom	x	y	z	U(eq)
Ga(1)	6667	3333	8355(1)	20(1)
Mn(1)	3333	6667	6019(1)	23(1)
S(1)	6667	3333	6881(2)	19(1)
S(2)	0	10000	5000	24(1)
S(3)	3333	6667	8972(1)	21(1)

Table S3. Single crystal anisotropic displacement parameters (Å^{2}x 10^3) for $Mn_2Ga_2S_5$. The anisotropic displacement factor exponent takes the form: $-2\pi^2[h^2 a^{*2}U^{11} + ... + 2 h k a^* b^* U^{12}]$

Atom	U^{11}	U^{22}	U^{33}	U^{23}	U^{13}	U^{12}
Ga(1)	18(1)	18(1)	26(1)	0	0	9(1)
Mn(1)	21(1)	21(1)	28(1)	0	0	10(1)
S(1)	19(1)	19(1)	20(1)	0	0	9(1)
S(2)	26(1)	26(1)	21(1)	0	0	13(1)
S(3)	21(1)	21(1)	19(1)	0	0	10(1)

Figure S1. Powder XRD Rietveld refinement results for $Mn_2Ga_2S_5$ using TOPAS 6.

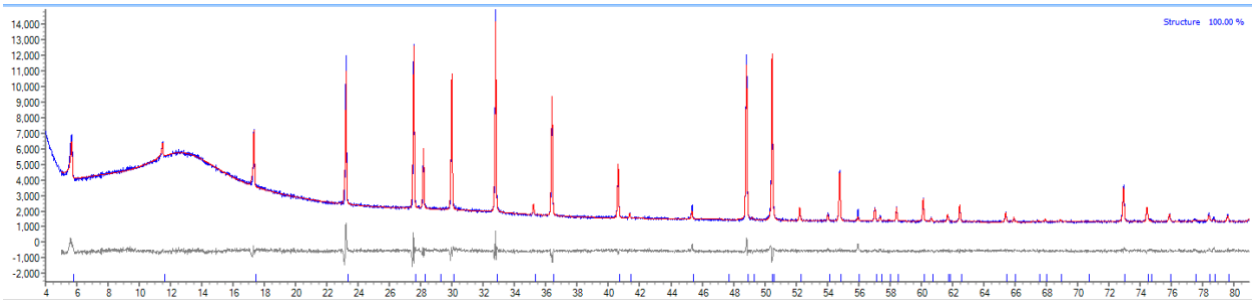

Table S4. Crystal data and refinement results for $Mn_2Ga_2S_5$.

Empirical Formula	$Mn_2Ga_2S_5$
Formula weight (g/mol)	409.647
Space group	P-3m1
a lattice parameter (Å)	3.719536(18)
c lattice parameter (Å)	15.23077(12)
Cell volume ($Å^3$)	182.486(2)
Temperature (K)	295 K
λ (Å)	1.5406
Pattern range (2θ)	5-81
R_{wp}	0.0353
R_p	0.0236
R_{exp}	0.0207

Table S5. Atomic coordinates and isotropic parameters from powder XRD Rietveld refinement results for $Mn_2Ga_2S_5$.

Element	x	y	z	Occupancy	B_{eq}
Mn1	0.33333	0.66667	0.6022(2)	1	0.89(10)
Ga1	0.33333	0.66667	0.1658(2)	1	2.44(9)
S1	0.00000	0.00000	0.50000	1	2.15(10)
S2	0.33333	0.66667	0.3161(3)	1	2.15(10)
S3	0.33333	0.66667	0.8897(4)	1	2.15(10)

Figure S2. Powder XRD Rietveld refinement results for $Mn_2In_2Se_5$ using TOPAS 6.

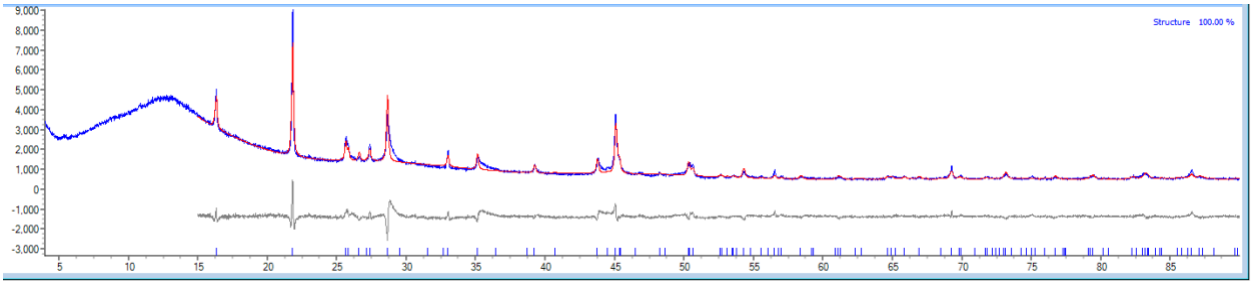

Table S6. Crystal data and refinement results for $Mn_2In_2Se_5$.

Empirical Formula	$Mn_2In_2Se_5$
Formula weight (g/mol)	2190
Space group	R-3m
a lattice parameter (Å)	4.0189(2)
c lattice parameter (Å)	48.830(4)
Cell volume (Å^3)	683.02(9)
Temperature (K)	295 K
λ (Å)	1.5406
Pattern range (2θ)	15-90
R_{wp}	0.0798
R_p	0.0557
R_{exp}	0.0315

Table S7. Atomic coordinates and isotropic parameters from powder XRD Rietveld refinement results for $Mn_2In_2Se_5$.

Element	x	y	z	Occupancy	B_{eq}
Mn1	0.00000	0.00000	0.6998(2)	0.81(2)	1.3(5)
In1	0.00000	0.00000	0.6998(2)	0.19(2)	1.3(5)
Mn2	0.00000	0.00000	0.55435(15)	0.19(2)	1.7(4)
In2	0.00000	0.00000	0.55435(15)	0.81(2)	1.7(4)
Se1	0.00000	0.00000	0.13572(19)	1	0.2(2)
Se2	0.00000	0.00000	0.39225(19)	1	0.2(2)
Se3	0.00000	0.00000	0.00000	1	0.2(2)

Table S8. Reported synthesis and magnetic properties of MTr_2Ch_4 compounds. Chemical Vapor Transport (CVT), Ferromagnetic (FM), Antiferromagnetic (AFM), Neutron Powder diffraction (NPD).

Material	**Synthesis**	**Magnetic Ordering**	**Transition Temperature**	**θ_W (K)**	**f**	**Technique**	**Ref.**
$MnIn_2Se_4$	CVT	Spin glass	3.5 K	N/A	N/A	AC X_m vs. T	[1]
$MnIn_2Se_4$	Anneal	Spin glass	2.75 K	-94	34	DC X_m vs. T	[2]
$MnIn_2Se_4$	CVT	FM	7 K	N/A	N/A	DC X_m vs. T and M vs. H	[3]
$FeIn_2Se_4$	CVT	Spin glass	17 K	-183	11	DC X_m vs. T	[4]
$FeIn_2Se_4$	Directional Solidification	Spin glass	18 K	-220	12	AC X_m vs. T	[5]
$FeGa_2S_4$	CVT	Spin glass to AFM	16 K (DC) 5 K (NPD)	-158.0	10	DC X_m vs. T and NPD	[6]
$FeGa_2S_4$	Powder	AFM to Spin glass	33 K (Mössbauer), 16 K (AC)	-174	11	AC X_m vs. T, Mössbauer	[7]
$FeGa_2S_4$	CVT	Spin glass	16 K	-160	10	DC X_m vs. T	[8]
$NiGa_2S_4$	CVT	Spin glass	10 K	-80	8	DC X_m vs. T	[8]

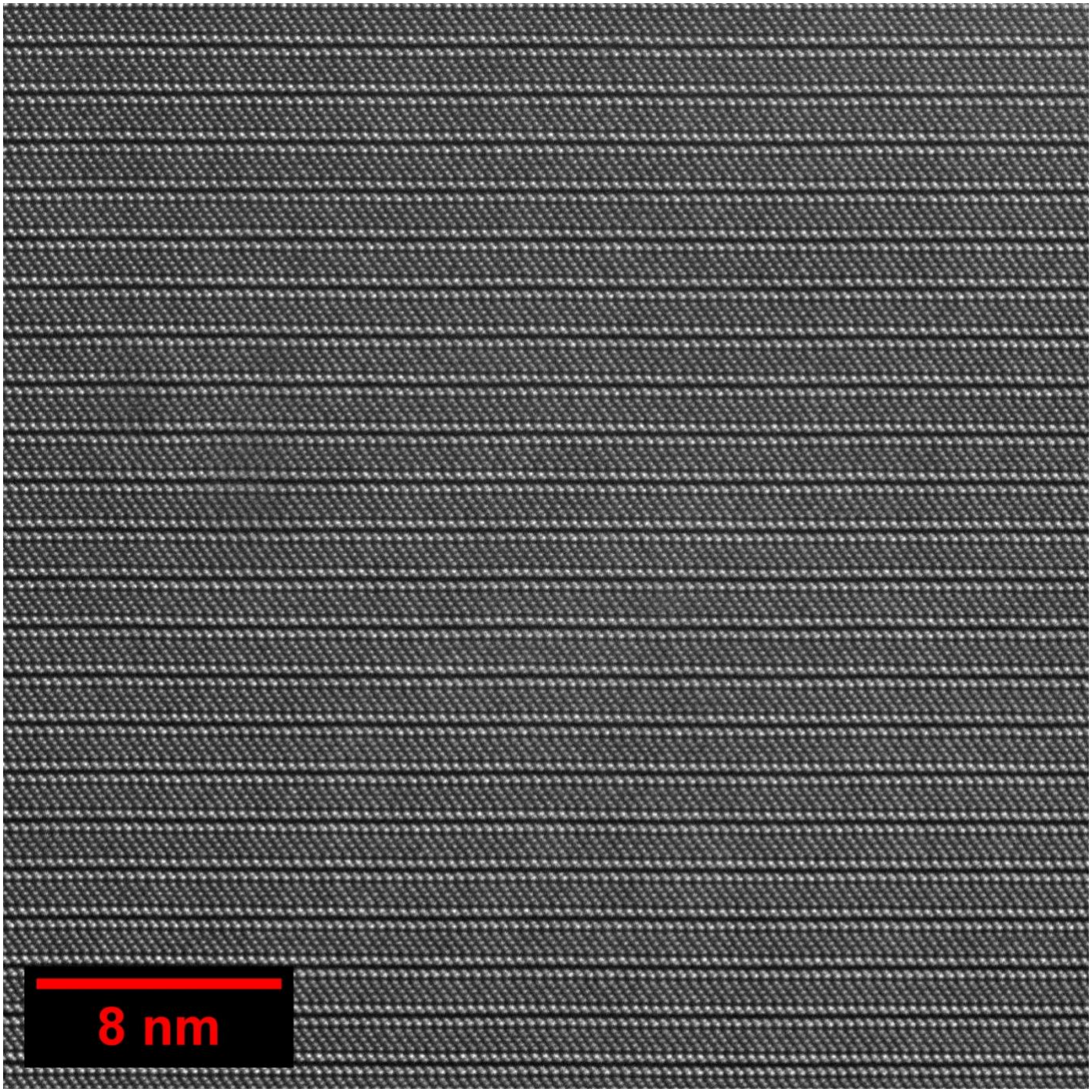

Figure S3. HAADF-STEM image of $Mn_2In_2Se_5$ viewed along the [1 0 0] direction.

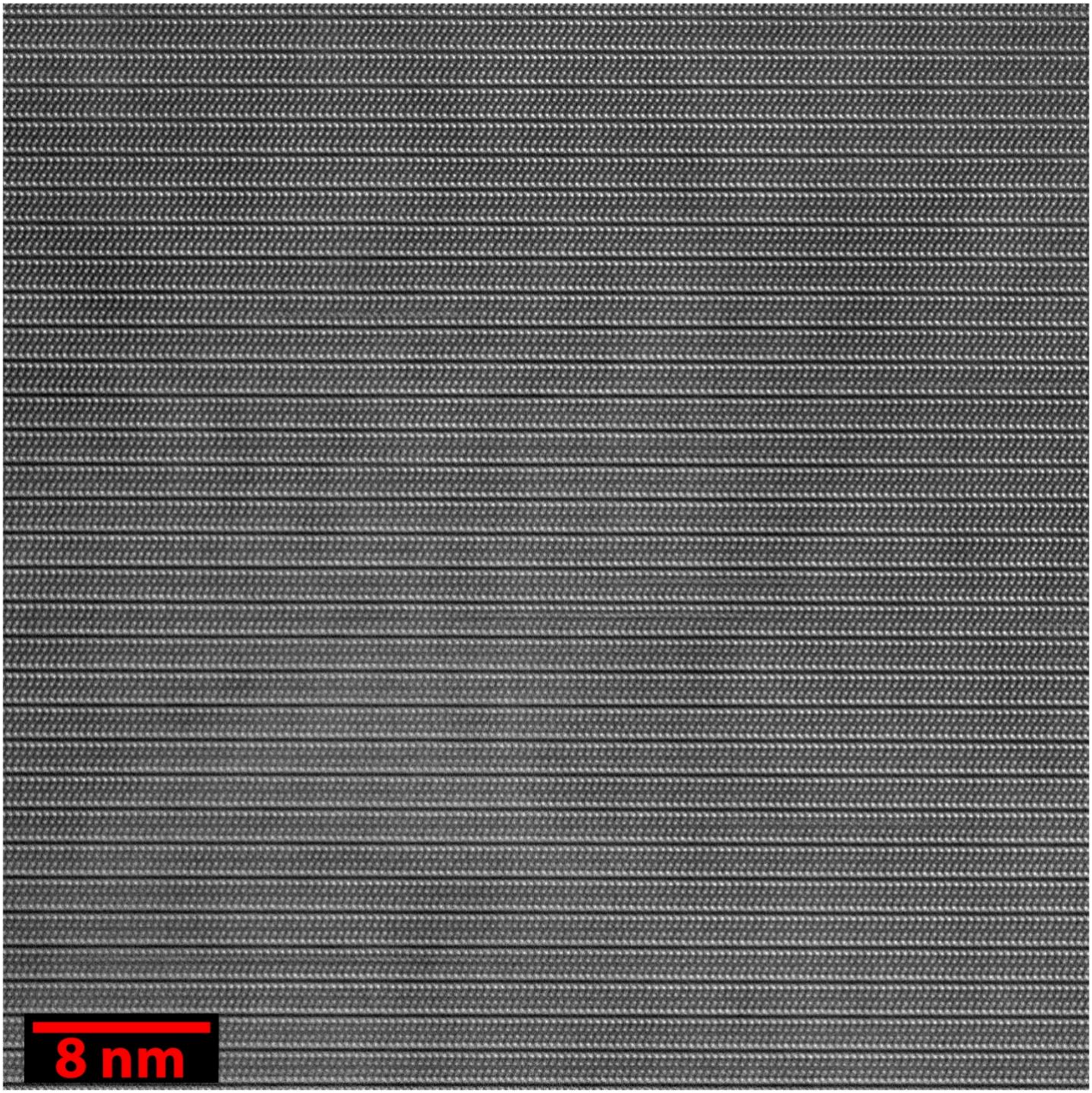

Figure S4. HAADF-STEM image of $Mn_2Ga_2S_5$ viewed along the [1 0 0] direction.

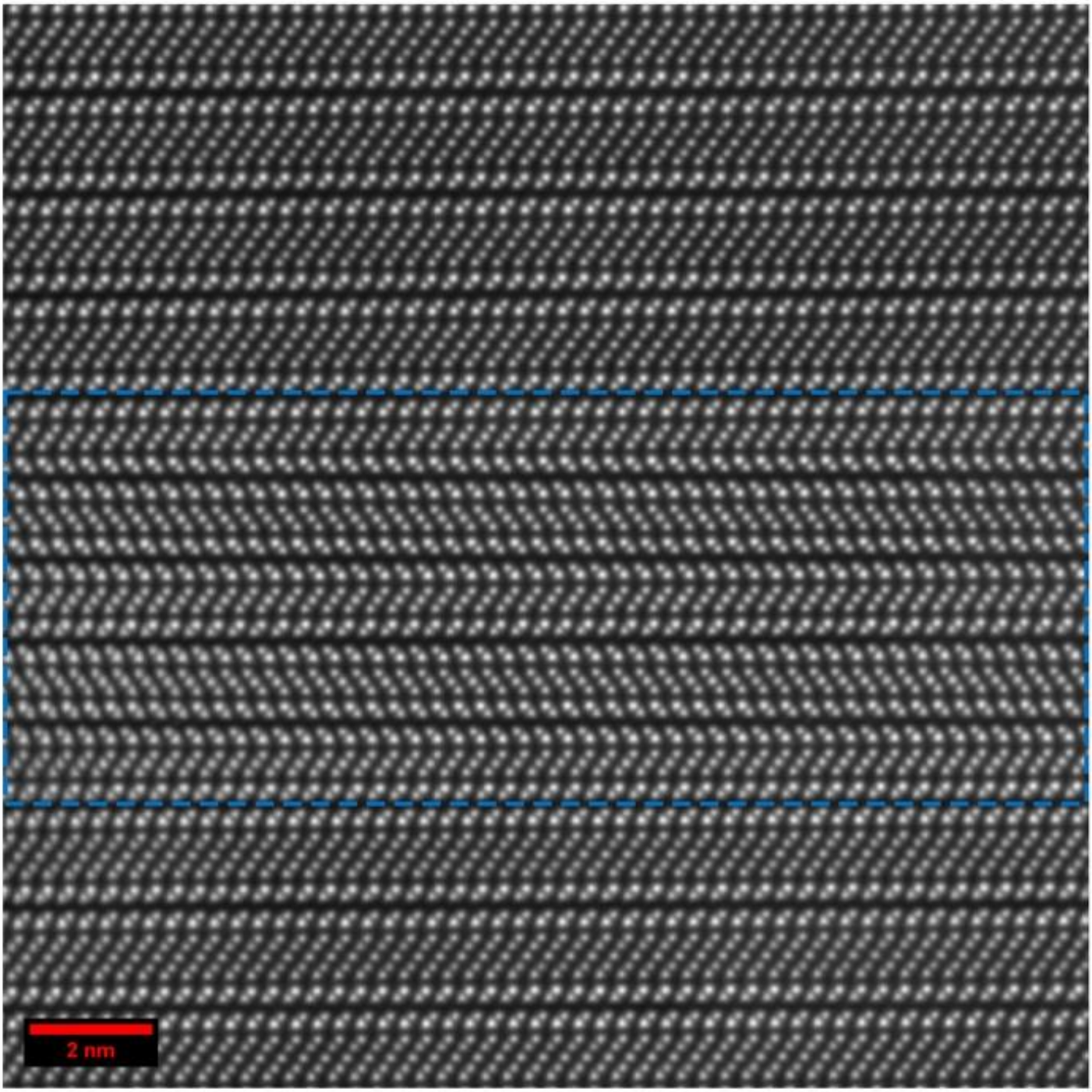

Figure S5. HAADF-STEM image of impure $Mn_2In_2Se_5$ crystals before CVT purification viewed along the [1 0 0] direction. Intergrowth layers of $MnIn_2Se_4$ shown in blue box.

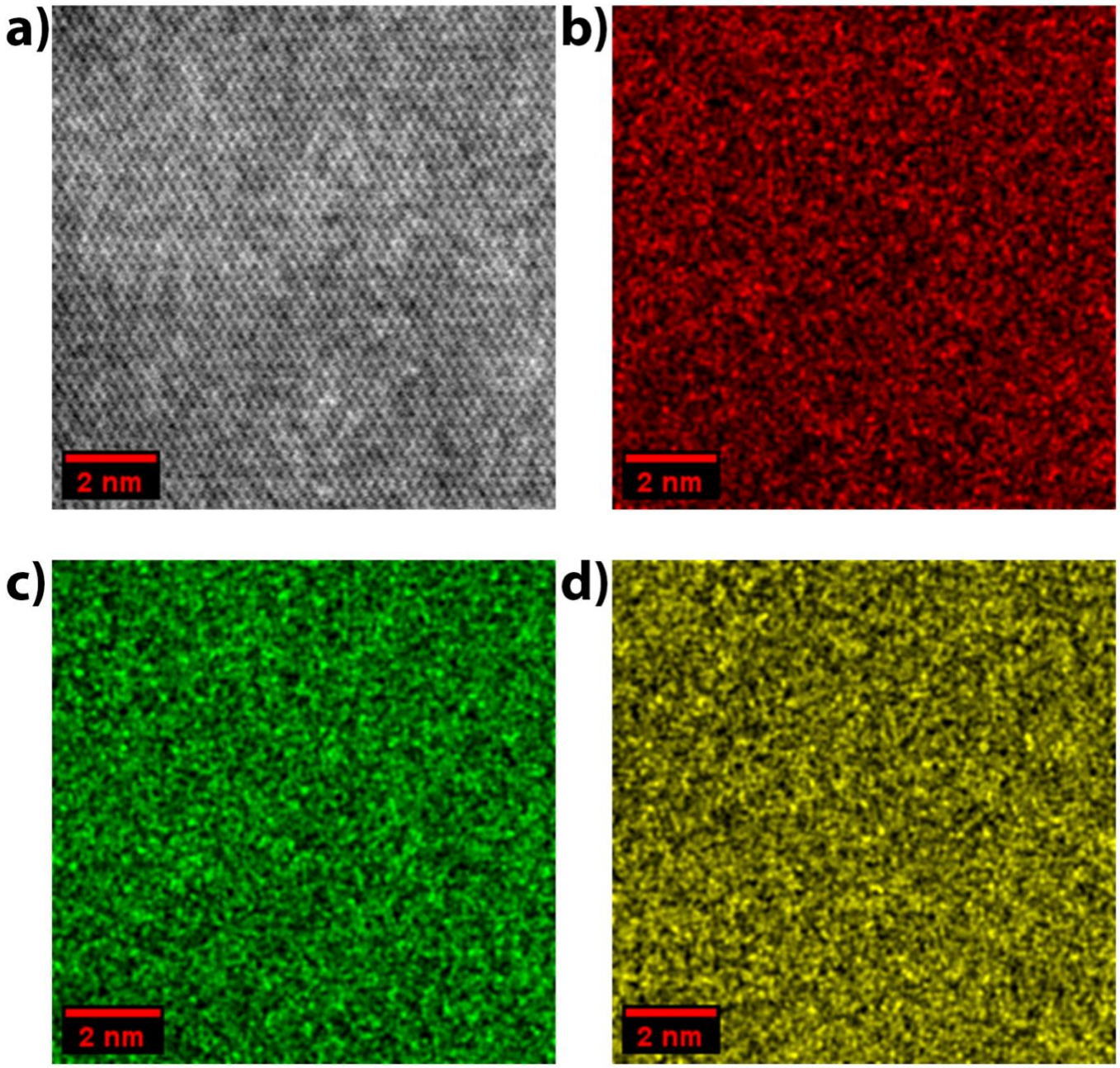

Figure S6. HAADF-STEM image (a) and Energy Dispersive X-ray Spectroscopy (EDX) element maps for manganese (b), indium (c), and selenium (d) of $Mn_2In_2Se_5$ viewed along the [0 0 1] direction.

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