

Supporting Information

Room-Temperature Ferromagnetism in $Mg_{1-x}Mn_{2+x}As_2$ with Layered Structure

Xiao-Cun Liu,^{*†} Min Zhu^{‡,§}, Sheng-Qing Xia^{*‡}

[†] *School of Civil Engineering, Shandong Jiaotong University, Jinan, Shandong 250023, People's Republic of China*

[‡] *State Key Laboratory of Crystal Materials, Institute of Crystal Materials, Shandong University, Jinan, Shandong 250100, People's Republic of China*

[§] *School of Materials Science and Engineering, Lanzhou University of Technology, Lanzhou, Gansu 730050, People's Republic of China*

Corresponding Authors:

***E-mail: liuxiaocunde@163.com**

***E-mail: shqxia@sdu.edu.cn**

Contents

Table S1. Refined atomic coordinates and isotropic displacement parameters for $\text{Mg}_{1-x}\text{Mn}_{2+x}\text{As}_2$ ($x = 0.17, 0.49, 0.69$).

Figure S1. (a) Powder X-ray diffraction of titled compounds $\text{Mg}_{1-x}\text{Mn}_{2+x}\text{As}_2$ ($x = 0.17, 0.48, 0.69$). The theoretical calculated patterns of $\text{Mg}_{0.83(3)}\text{Mn}_{2.17}\text{As}_2$ are provided for comparison as well. The small peak marked with * at about 44.4° for $\text{Mg}_{0.52(2)}\text{Mn}_{2.48}\text{As}_2$ indicates possible As impurity. (b) Calculated lattice parameters from the PXRD results vs. x .

Figure S2. EDS analysis on the composition of single crystals for $\text{Mg}_{0.83(2)}\text{Mn}_{2.17}\text{As}_2$.

Figure S3. EDS analysis on the composition of single crystals for $\text{Mg}_{0.52(2)}\text{Mn}_{2.48}\text{As}_2$.

Figure S4. EDS analysis on the composition of single crystals for $\text{Mg}_{0.31(3)}\text{Mn}_{2.69}\text{As}_2$.

Figure S5. Schematic diagram of the spin configuration for MH curve at 5 K.

Table S1. Refined atomic coordinates and isotropic displacement parameters for $\text{Mg}_{1-x}\text{Mn}_{2+x}\text{As}_2$ ($x = 0.17, 0.49, 0.69$).

Atoms	Wyckoff	Occupancy	x	y	z	U_{eq}^a (\AA^2)
$\text{Mg}_{0.83(2)}\text{Mn}_{2.17}\text{As}_2$						
Mg 1	$1a$	0.83(2)	0	0	0	0.0199(16)
Mn1	$1a$	0.17(2)	0	0	0	0.0199(16)
Mn2	$2d$	1	1/3	2/3	0.6304(2)	0.0143(5)
As1	$2d$	1	1/3	2/3	0.23653(14)	0.0130(4)
$\text{Mg}_{0.52(2)}\text{Mn}_{2.48}\text{As}_2$						
Mg 1	$1a$	0.52(2)	0	0	0	0.0222(13)
Mn1	$1a$	0.48(2)	0	0	0	0.0222(13)
Mn2	$2d$	1	1/3	2/3	0.6317(2)	0.0148(5)
As1	$2d$	1	1/3	2/3	0.23523(15)	0.0130(4)
$\text{Mg}_{0.31(3)}\text{Mn}_{2.69}\text{As}_2$						
Mg 1	$1a$	0.31(3)	0	0	0	0.0285(17)
Mn1	$1a$	0.69(3)	0	0	0	0.0285(17)
Mn2	$2d$	1	1/3	2/3	0.6324(4)	0.0203(8)
As1	$2d$	1	1/3	2/3	0.2340(3)	0.0195(7)

^a U_{eq} is defined as one third of the trace of the orthogonalized U^{ij} tensor.

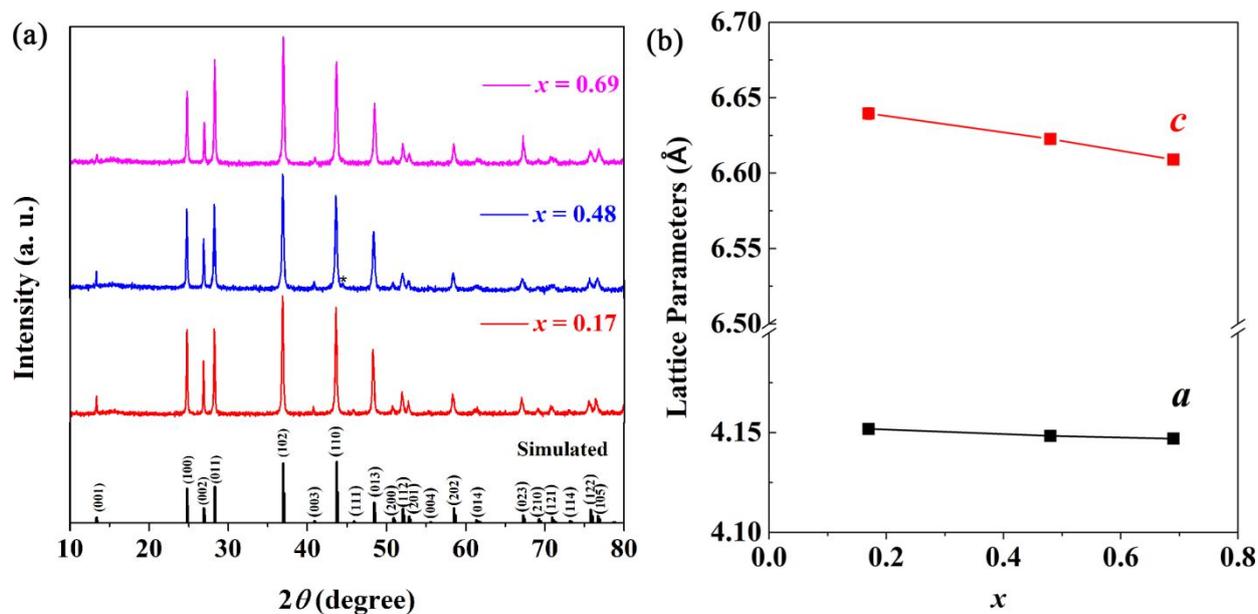


Figure S1. (a) Powder X-ray diffraction of titled compounds $\text{Mg}_{1-x}\text{Mn}_{2+x}\text{As}_2$ ($x = 0.17, 0.48, 0.69$). The theoretical calculated patterns of $\text{Mg}_{0.83(3)}\text{Mn}_{2.17}\text{As}_2$ are provided for comparison as well. The small peak marked with * at about 44.4° for $\text{Mg}_{0.52(2)}\text{Mn}_{2.48}\text{As}_2$ indicates possible As impurity. (b) Calculated lattice parameters from the PXRD results vs. x . The calculated lattice parameters are slightly different with SXRD results, which may be caused by the test error.

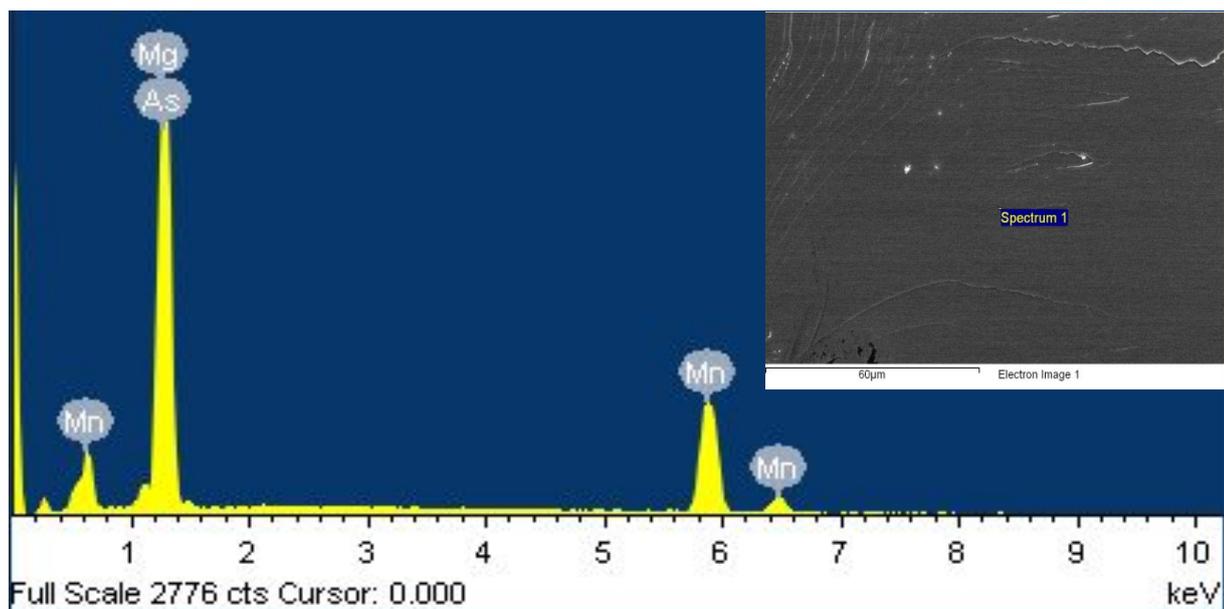


Figure S2. EDS analysis on the composition of single crystals for $\text{Mg}_{0.83(2)}\text{Mn}_{2.17}\text{As}_2$.

Sample 1

Element	Weight%	Atomic%
Mg L	9.15	21.01
Mn L	41.55	42.23
As L	49.31	36.76

Sample 2

Element	Weight%	Atomic%
Mg L	9.28	21.24
Mn L	41.92	42.49
As L	48.80	36.27

Sample 3

Element	Weight%	Atomic%
Mg L	9.04	20.92
Mn L	39.56	40.50
As L	51.40	38.59

The content of Mn calculated from EDS: 41.7%

The content of Mn calculated from SXRD: 43.4%

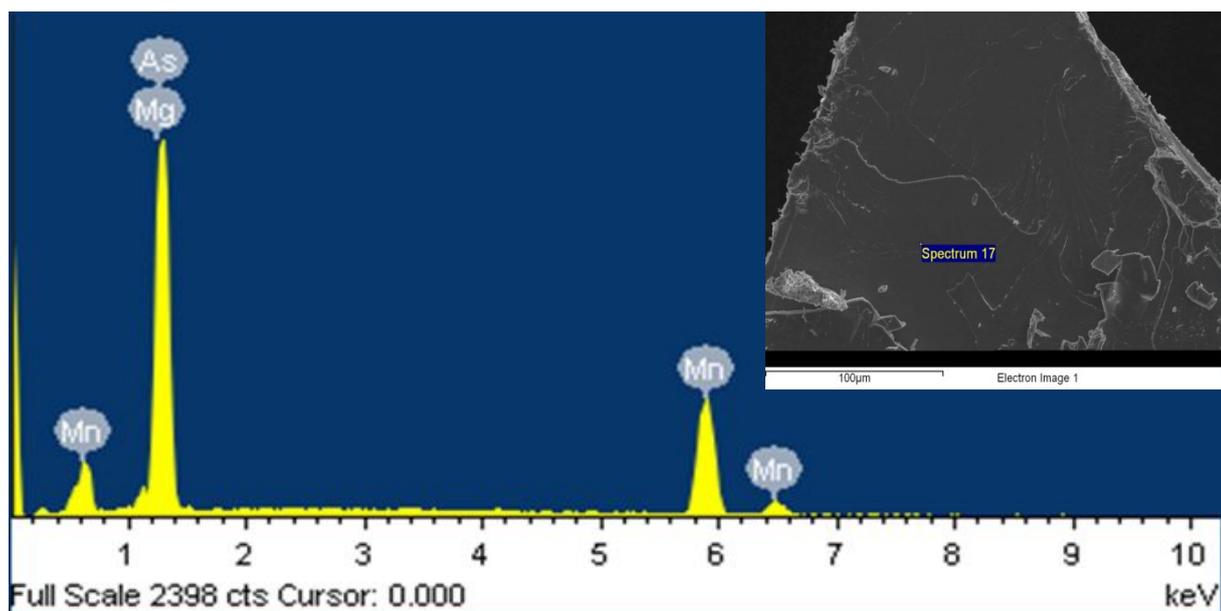


Figure S3. EDX analysis on the composition of single crystals for $\text{Mg}_{0.52(2)}\text{Mn}_{2.48}\text{As}_2$.

Sample 1

Element	Weight%	Atomic%
Mg L	7.53	17.65
Mn L	43.40	45.02
As L	49.07	37.33

Sample 2

Element	Weight%	Atomic%
Mg L	7.82	18.36
Mn L	41.06	42.68
As L	51.13	38.97

Sample 3

Element	Weight%	Atomic%
Mg L	6.16	14.80
Mn L	42.55	45.22
As L	51.29	39.97

The content of Mn calculated from EDS: 44.3%

The content of Mn calculated from SXRD: 49.6%

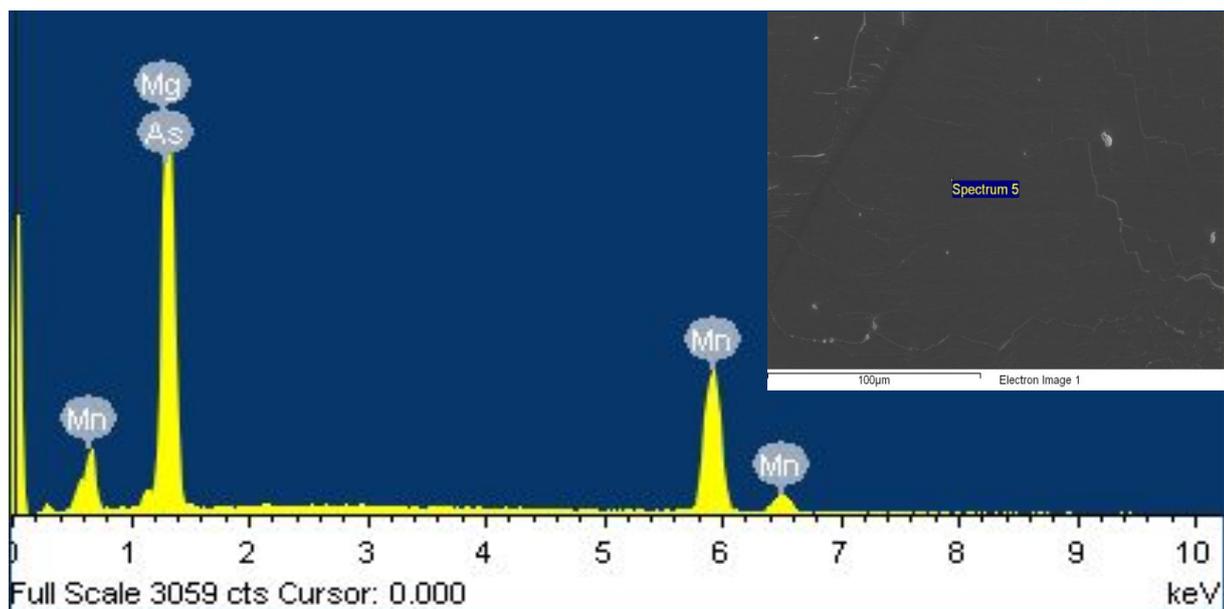


Figure S4. EDX analysis on the composition of single crystals for $\text{Mg}_{0.31(3)}\text{Mn}_{2.69}\text{As}_2$.

Sample 1

Element	Weight%	Atomic%
Mg L	5.83	13.96
Mn L	45.65	48.35
As L	48.52	37.69

Sample 2

Element	Weight%	Atomic%
Mg L	5.11	12.32
Mn L	47.19	50.35
As L	47.70	37.32

Sample 3

Element	Weight%	Atomic%
Mg L	5.82	13.91
Mn L	46.32	48.98
As L	47.86	37.11

The content of Mn calculated from EDS: 49.2%

The content of Mn calculated from SXRD: 53.8%

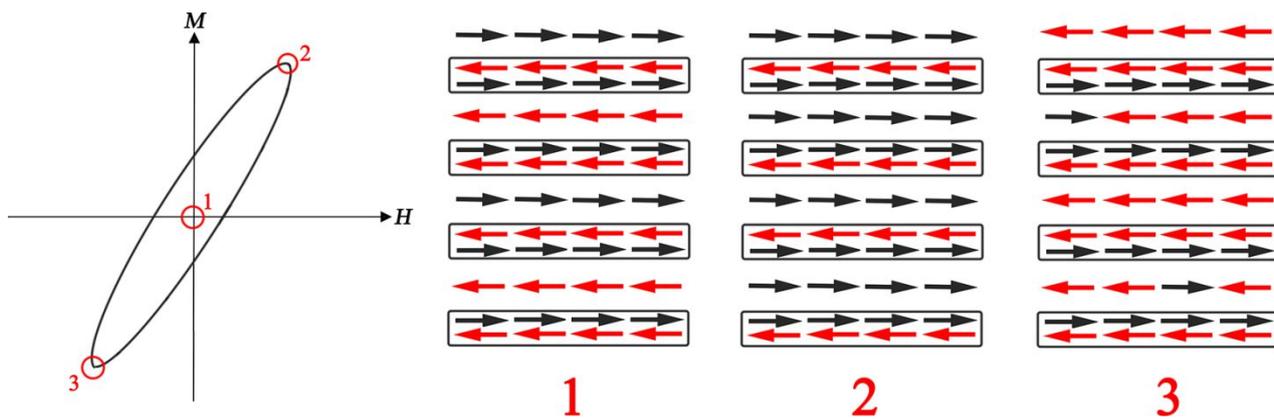


Figure S5. Schematic diagram of the spin configuration for MH curve at 5 K.