Supplementary Figures and Tables for:

Colossal Magnetoresistance in the Mn^{2+} oxypnictides $NdMnAsO_{1-x}F_x$

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Experimental

Polycrystalline samples of $Nd_{0.97}MnAsO_{1-x}F_x$ with nominal x = 0.05, 0.065, 0.08 were prepared *via* a two step solid-state reaction. Pieces of rare earth Nd (Aldrich 99.9%) and As (Alfa Aesar 99.999%) were reacted at 900°C in a quartz tube sealed under vacuum. The precursor was then reacted with stoichiometric amounts of MnO_2, Mn and MnF_2 (Aldrich 99.99%). The powders were ground and pressed into pellets of 10 mm diameter and placed in a Ta crucible. The pellets were then sintered at 1150°C for 48 hours in an evacuated quartz tube.

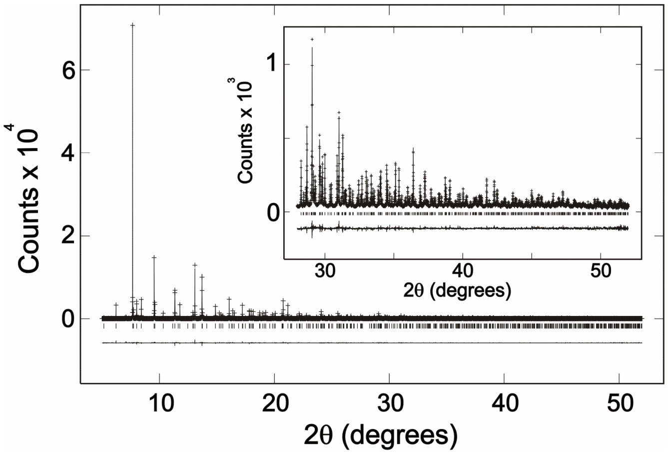
High resolution synchrotron X-ray powder diffraction patterns were recorded on the ID31 beamline for $NdMnAsO_{0.95}F_{0.05}$ at the ESRF, Grenoble, France at 290 K with a wavelength of 0.3999 Å. The powder sample was inserted into a 0.5mm diameter borosilicate glass capillary and spun at ~1Hz. The patterns were collected between $2^o < 2\vartheta < 53^o$.

Powder neutron diffraction data were recorded for $NdMnAsO_{0.95}F_{0.05}$ using the D20 high intensity diffractometer at the Institute Laue Langevin (ILL, Grenoble, France). Neutrons of wavelength 2.4188 Å were incident on an 8 mm vanadium can contained in a vertical cryomagnet. Zero field data were recorded between 4-370 K and variable field data were recorded at 4 K between 0 – 5 T and again at 0 T with a collection time of 20 minutes per field/temperature. The temperature and field dependency of the magnetic structure of the compound was obtained by Rietveld refinement of the neutron data.

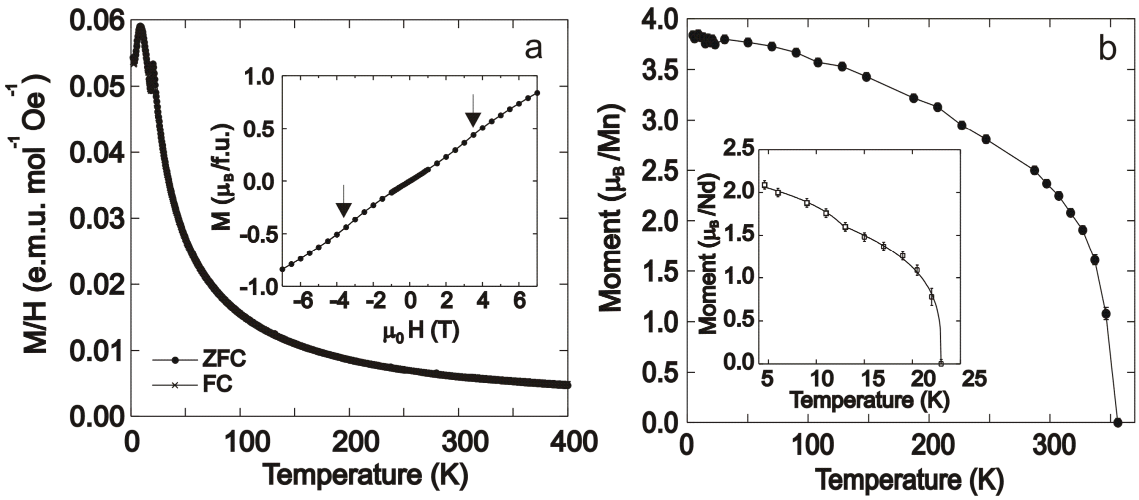
The temperature and field dependence of the electrical resistance were recorded using a Quantum Design physical property measurement system (PPMS) between 4 and 400 K and in magnetic fields up to 9 T.

The magnetic susceptibility was measured with a Quantum Design superconducting quantum interference device magnetometer (SQUID). Zero field cooled (ZFC) and field cooled (FC) measurements were recorded between 2-400 K in a field of 1000 Oe. Variable field data were recorded at 2 K between ±7 T.

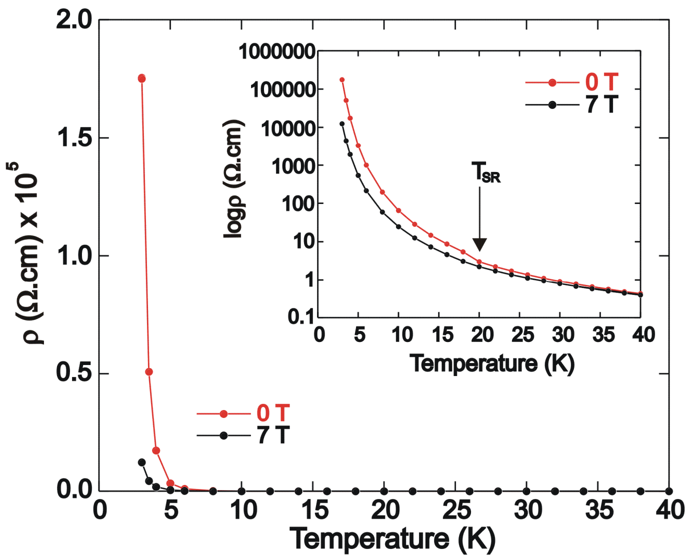
Supplementary Figures



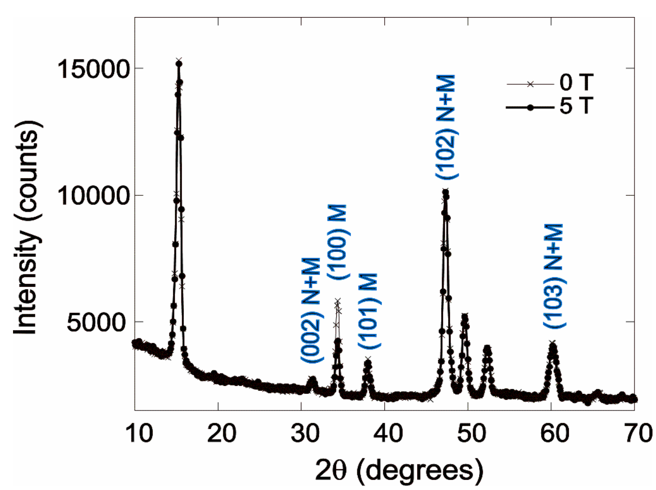
Supplementary Figure 1. Rietveld refinement fit to the 4 K synchrotron X-ray powder pattern of $NdMnAsO_{0.95}F_{0.05}$ in which an excellent fit is obtained with the tetragonal space group *P4/nmm*.



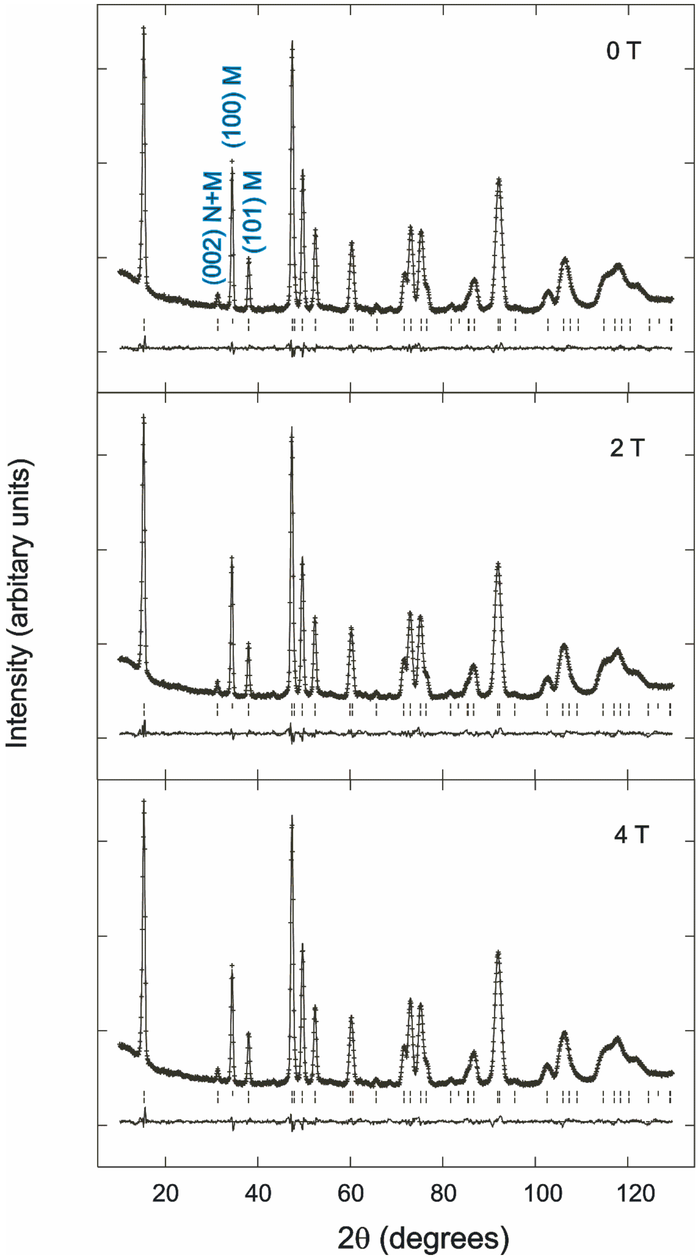
Supplementary Figure 2. a, Zero field cooled (ZFC) and field cooled (FC) variable temperature susceptibility data recorded for $NdMnAsO_{0.95}F_{0.05}$. The inset shows the field variation of the magnetisation; a change of slope is observed above 3.5 T as indicated by the arrow. b**,** The thermal variation of the Mn^{2+} magnetic moment refined from neutron data with T_N (Mn) = 356 K. The inset shows the variation of the Nd^{3+} moment with temperature evidencing an antiferromagnetic transition at 23 K.



Supplementary Figure 3. Temperature variation of the low temperature resistivity with H = 0 T and H = 7 T. The inset shows the variation of log(resistivity) for the same temperatures and fields. The spin reorientation transition is labelled.



Supplementary Figure 4. A portion of the 4 K neutron diffraction pattern showing a reduction in intensity of the (100) magnetic diffraction peak with increasing magnetic field (μ_0H) from 0 – 5 T, where N and M represent nuclear and magnetic diffraction respectively. There is no evidence of an increasing ferromagnetic component upon increasing μ_0H from 0 – 5 T.



Supplementary Figure 5. Rietveld fits to 4 K neutron diffraction patterns of $NdMnAsO_{0.95}F_{0.05}$ at selected magnetic fields.

Supplementary Table 1: Refined cell parameters, agreement factors, atomic parameters and selected bond lengths and angles for $NdMnAsO_{0.95}F_{0.05}$ from Rietveld fits against ID31 synchrotron X-ray diffraction data at 4 K and 290 K. Nd and As are at 2c (¼, ¼, z), Mn at 2b (¾, ¼, ½) and O/F at 2a (¼, ¾, 0).

Atom	Occupancy		Temperature (K)	
			4	290
Nd	1.00	z	0.13125(3)	0.13090(3)
		U_{iso} (Å^2)	0.0022(1)	0.0090(5)
Mn	1.00	U_{iso} (Å^2)	0.0020(2)	0.0103(1)
As	1.00	z	0.67431(6)	0. 67406(5)
		U_{iso} (Å^2)	0.0014(1)	0.0094(1)
O/F	1.00	U_{iso} (Å^2)	0.0057(7)	0.0129(6)
		a (Å)	4.04002(2)	4.04870(2)
		c (Å)	8.86238(6)	8.89654(6)
		χ^2 (%)	4,17	4.23
		R_{WP} (%)	6.87	7.21
		R_P (%)	5.50	6.80
		Nd-O (Å)	2.3317(4)	2.3354(1)
		Mn-As (Å)	2.5498(7)	2.5487(3)
		α_2 Nd-O-Nd (^{o})	120.13(1)	120.18(2)
		α_1 Nd-O-Nd (^{o})	104.420(6)	104.397(5)
		α_1 As-Mn-As (^{o})	111.66(1)	111.664(8)
		α_2 As-Mn-As (^{o})	105.19(2)	105.17(2)

Supplementary Table 2: The variation of cell parameters with x for the $NdMnAsO_{1-x}F_x$ samples as determined from laboratory x-ray diffraction data recorded on a Bruker D8 advance. The cell parameters for NdMnAsO are shown for comparative purposes.

			x	
	0.000	**0.050**	**0.065**	**0.080**
***a* (Å)**	4.0503(1)	4.0500(1)	4.0497(1)	4.0490(1)
***c* (Å)**	8.9150	8.9040(4)	8.8988(4)	8.8956(4)

Supplementary Table 3: –MR can be fit to the equation $-MR = A + BH + CH^2$ (A, B and C are constants) between 0.5 – 3.5 T for 3 – 16 K and between 1.5 - 9 T for 30 K. Refined values of A, B and C are given below.

		Temperature	**(K)**	
	3	**4**	**16**	**30**
A	5.0(1)	5.0(1)	2.0(1)	2.1(1)
B	-19.0(1)	-18.4(1)	-4.1(1)	-2.5(1)
C	-1.0(3)	-0.7(1)	-1.11(2)	0.00(2)