Electronic Supplementary Information for;

Competing antiferromagnetic orders in the double perovskite Mn2MnReO6 (Mn3ReO6), A. M. Arevalo-Lopez, F. Stegemann, and J. P Attfield.

Experimental details

PXRD was performed with a Bruker AXS D2 Phaser diffractometer equipped with Cu-K_α radiation and is shown along with the Rietveld refinement on Figure S1. Single crystal data were collected on a SuperNova X-ray Diffraction System (Oxford) and the structure was solved using the Superflip package in Jana2006. Further details are in Table S1. Magnetic properties were measured using a Quantum Design MPMS XL commercial device.

Four high pressure products were combined to give a ~70 mg sample for neutron powder diffraction. Powder neutron diffraction profiles were recorded between 10 < T < 200 K at a wavelength of 2.41 Å in high flux mode on the D20 diffractometer at ILL, Grenoble (Figure S3). Rietveld refinement was performed with the FULLPROF program. Refinement of the data at 200 K was performed using the single crystal structure as starting model. MnO phase was observed as secondary phase (< 3% wt.) and taken into account in the structural and magnetic refinements. Lattice parameter variations are in Fig. S2.

Magnetic symmetry analysis was performed with BasIrreps within the Fullprof suite and is summarized in Table S2.Magnetic moments were constrained in the ab-plane, as attempts to refine magnetic components along the *c*-axis (ψ_3 or ψ_6, see Table S2) gave unstable refinements or zero magnitude within error. The best agreement with the experimental data required a combination of Γ_1 and Γ_3 Irreps. Constraints were needed in view of the large number of independent spin components, and the small magnitude of Re^{6+} compared to Mn^{2+} spins. The small monoclinic angle of Mn_2MnReO_6 also precludes unambiguous determination of the moment directions in the *ab*-plane from powder data. The magnitude for the A site was constrained to be $\sqrt{2}\mu_x = \mu_y$, almost parallel to the [1T0] direction, and those for the B/B' sites to be perpendicular along [110] with $2\mu_x = \mu_y$.

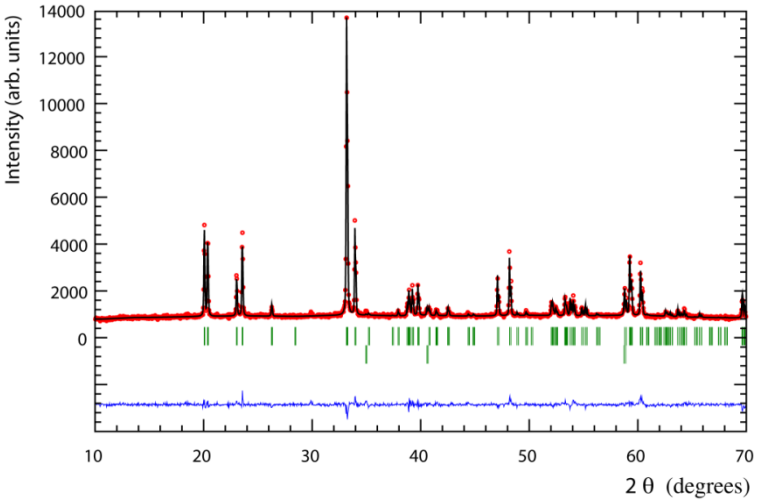

Figure S1: Laboratory powder X-ray diffraction for Mn_2MnReO_6 at 300 K. Lower marks correspond to MnO (<3 % wt.).

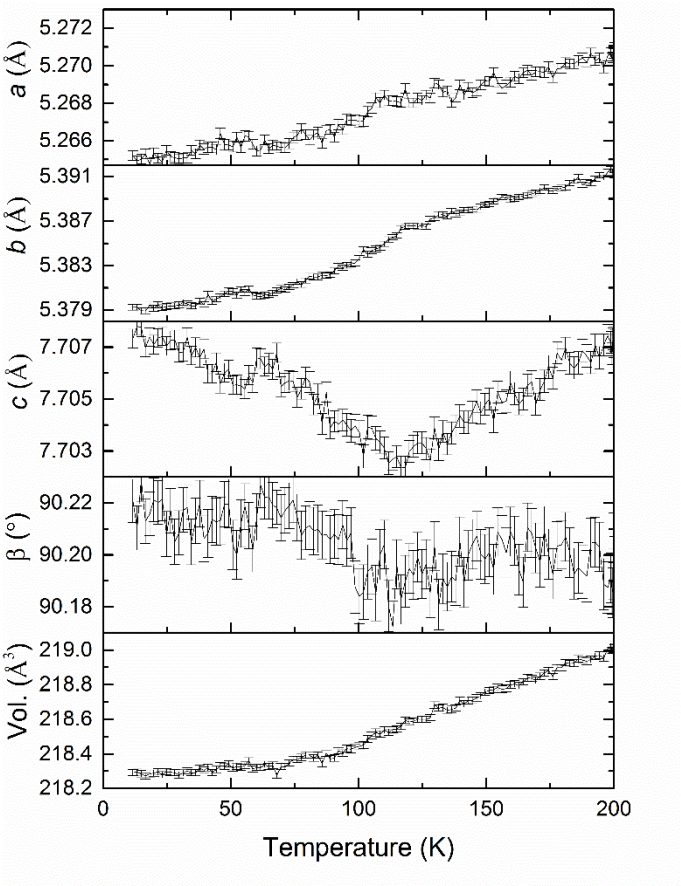

Figure S2. Plot of the lattice parameters from neutron diffraction against temperature for Mn_2MnReO_6, showing anomalies around the 99 and 109 K transitions.

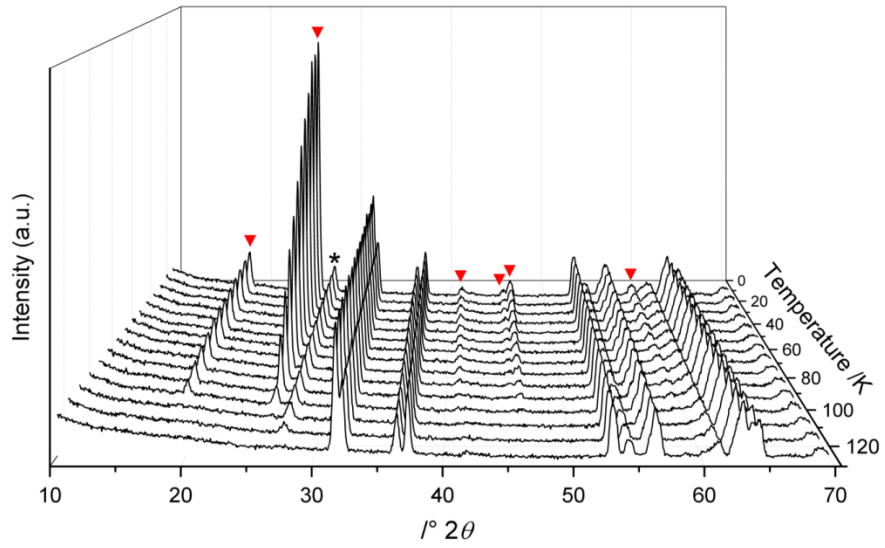

Figure S3. Thermal evolution of the NPD patterns of Mn2MnReO6 between 10 and 130 K, red triangles mark the reflections corresponding to the perovskite, asterisk marks the magnetic MnO reflections.

Table S1: Crystal data and structure refinement information for Mn_2MnReO_6 at T = 120 K, space group $P2_1/n$.

Molar mass (g mol^{-1})	447.02
Unit cell dimensions	a = 5.2708(3) Å b =5.3869(4) Å c = 7.7100(5) Å β = 90.097(5) °
Unit cell volume	218.91 Å^3
Calculated density (g cm^3)	6.7617
Crystal size (μm^3)	60 x 50 x 30
Wavelength	Mo $K\alpha$ (71.073 pm)
Absorption coefficient (mm^{-1})	35.708
F(000)	395
Θ range /°	4.62 to 29.49
hkl range	$-7 \le h \le 7$, $-7 \le k \le 7$, $-7 \le l \le 7$
total no. reflections	3766
independent reflections	585
data/parameters	585/52
Goodness of fit on F^2	0.96
R1/wR2 ($I \ge 3\sigma(I)$)	1.34/1.56
R1/wR2 (all data)	1.96/1.69
Largest diff. peak and hole (e$^-$ A^3)	0.60/-0.50

Table S2. Symmetry analysis for Mn_2MnReO_6 in $P2_1/n$ setting with k = (0.5 -0.5 0) propagation vector. Linear combinations of the basis vectors (BV) along with the constraints responsible of the magnetic structure are also given. The coordinates are: MnA1 = (x, y, z), MnA2 = (-x+3/2, y-½, -z+3/2), MnA3 = (-x+1, -y+1, -z+1), MnA4 = (x+½, -y+3/2, z-½), MnB1 = (0.5, 0, 0), MnB2 = (0, 0.5, 0.5), Re1 = (0.5, 0, 0.5) and Re2 = (0, 0.5, 0). (These coordinates are shifted by (0, 0.5, 0) relative to those shown in Table 1, but this does not affect the refinement results.)

Irreps	Γ_1			Γ_3		
BV	ψ_1	ψ_2	ψ_3	ψ_4	ψ_5	ψ_6
	μ_x	μ_y	μ_z	μ_x	μ_y	μ_z
MnA1	1	1	1	–	–	–
MnA2	–	–	–	-1	-1	-1
MnA3	-1	-1	-1	–	–	–
MnA4	–	–	–	-1	1	-1
MnB1	1	1	1	–	–	–
MnB2	–	–	–	1	-1	1
Re1	1	1	1	–	–	–
Re2	–	–	–	1	-1	1
MnA	$\psi_1+\psi_2+\psi_4+\psi_5$			$\lvert\psi_1\rvert=3.6(1)\ \mu_B$	$\lvert\psi_5\rvert=2.5\mu_B$	
MnB / ReB	$\psi_2+\psi_5$ / $\psi_1+\psi_4$			$\lvert\psi_{1Mn}\rvert=1.5(2)\ \mu_B$	$\lvert\psi_{1Re}\rvert=0.7(2)\mu_B$	
			constraints			
A site	$\lvert\psi_1\rvert = \lvert\psi_4\rvert$		$\lvert\psi_2\rvert = -\lvert\psi_5\rvert$		$\lvert\psi_5\rvert = \sqrt{2}\lvert\psi_4\rvert$	
B site	$\lvert\psi_1\rvert = -\lvert\psi_4\rvert$		$\lvert\psi_1\rvert = \lvert\psi_2\rvert = \lvert\psi_5\rvert$			