Supplementary Information (SI) for Journal of Materials Chemistry C.

Disorder stabilisation of high pressure $Mn_{2-x}Co_xScSbO_6$ double perovskites with complex magnetic properties

K. Ji,*[a,b] F. Denis Romero,[a] C. Ritter,[c] J. Paul Attfield,[b] Y. Shimakawa[a]

a. Institute for Chemical Research, Kyoto University, Uji, Kyoto 611-0011, Japan.

b.Centre for Science at Extreme Conditions (CSEC) and School of Chemistry, University of Edinburgh, Edinburgh EH9 3FD, UK

c. Institute Laue-Langevin, 38042 Grenoble, France

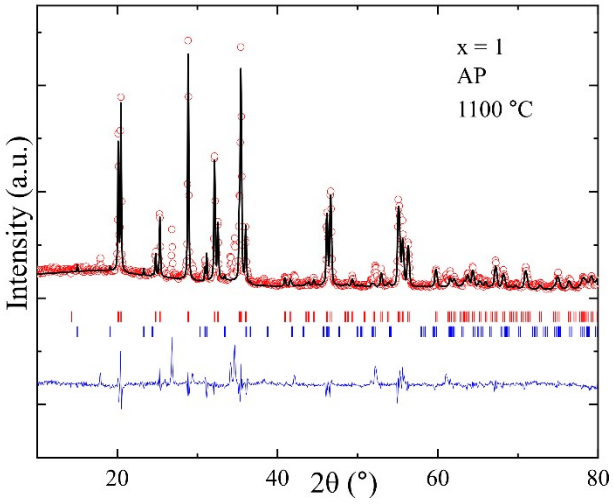

Fig. S1 Rietveld fits of room temperature XRD data for x = 1 $MnCoScSbO_6$ synthesised at 1100 ˚C under AP. Data are fitted using a distorted Mg_3TeO_6-type structural model (*R*-3, red tick marks) with refined lattice parameters a = 8.8201(1) Å and c = 10.5507(6) Å. Blue tick marks refer to $Sc_{5.5}Sb_{1.5}O_{12}$. There are several unidentified impurity peaks.

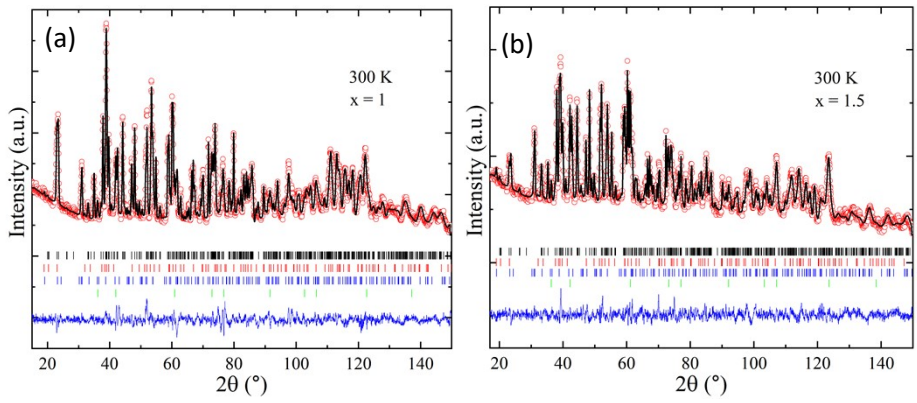

Fig. S2 Rietveld fits of 300 K high resolution (λ = 1.54 Å) NPD data for (a) x = 1 (DPv: 77(1)%, NTO phase: 10(1)%, $Sc_{5.5}Sb_{1.5}O_{12}$: 9(1)% and (Mn/Co)O: 4(1)%) and (b) x = 1.5 (DPv: 72(1) %, NTO phase: 11(1) %, $Sc_{5.5}Sb_{1.5}O_{12}$:12(1)% and (Mn/Co)O: 5(1)%) are denoted by black, red, blue, and green tick marks, respectively.

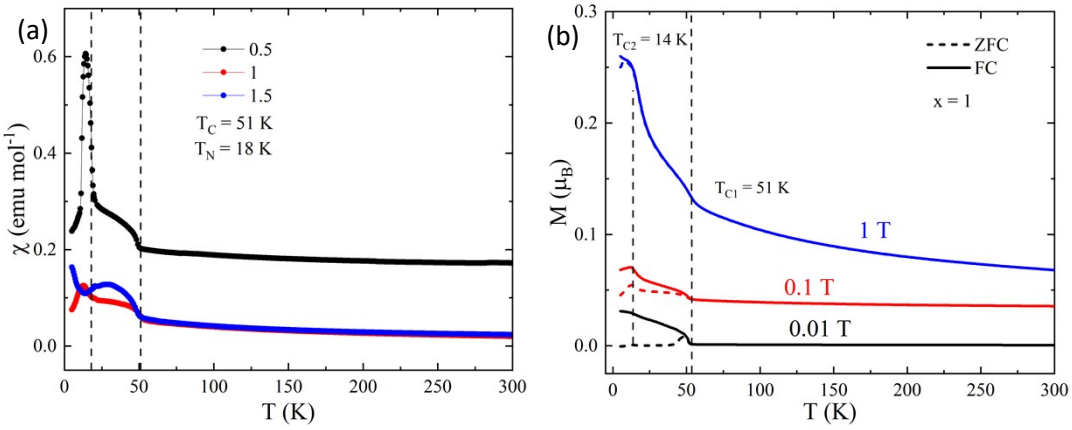

Fig. S3 (a) ZFC-FC magnetic susceptibility of DPv *x* = 0.5, 1 and 1.5 collected under 0.1 T. Magnetic transitions at 51 K and 18 K are observed. (c) Magnetic susceptibility measured under different applied magnetic fields for *x* = 1.

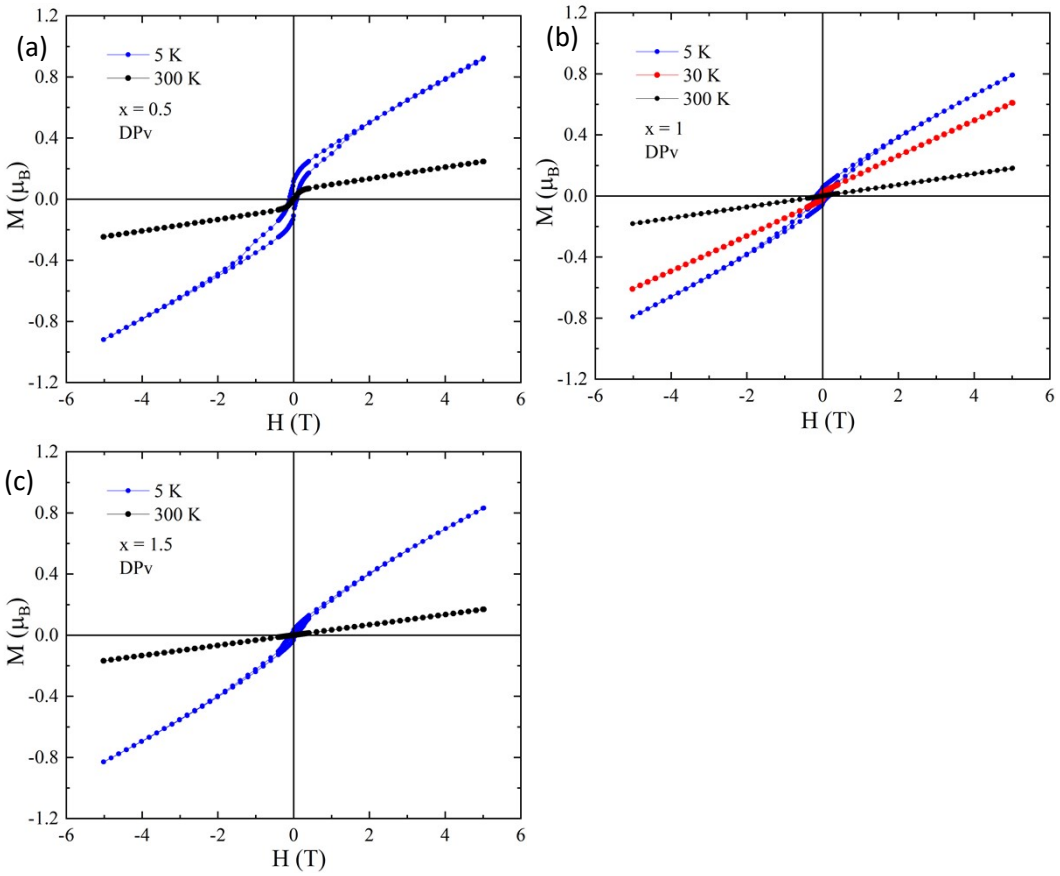

Fig. S4 Magnetic hysteresis loops of (a) *x* = 0.5, (b) *x* = 1 and (c) *x* = 1.5 samples. Hysteresis loops for *x* = 0.5 show a weak hysteretic component at 5 K, whereas *x* = 1 and 1.5 exhibit largely linear *M-H* behaviour consistent with glassy magnetic behaviour associated with spin freezing. The 300 K *x* = 0.5 *M-H* loop displays a weak ferromagnetic impurity contribution, whereas *x* = 1 and 1.5 300 K loops show paramagnetic linear *M-H* correlation.

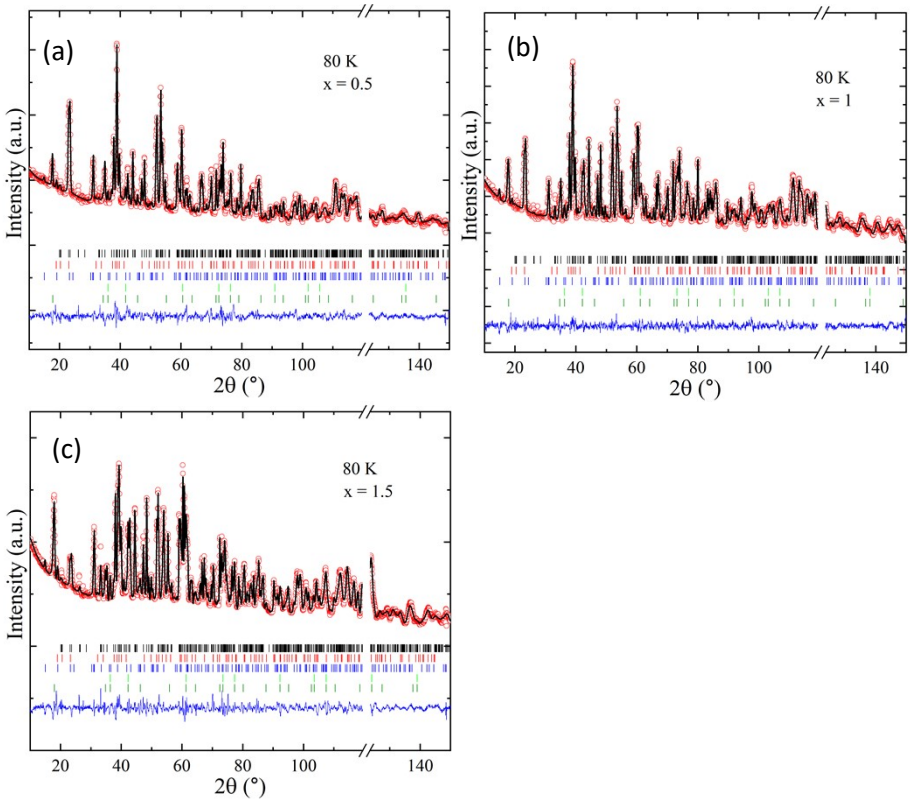

Fig. S5 Rietveld fits of 80 K high resolution (λ = 1.54 Å) NPD data for (a) x = 0.5, (b) x = 1 and (c) x =1.5. DPv, NTO phase, $Sc_{5.5}Sb_{1.5}O_{12}$ and (Mn/Co)O nuclear and magnetic are denoted by black, red, blue and green, dark green tick marks, respectively. The excluded region at $2\vartheta \sim 120^{\circ}$ is due to an uncalibrated detector cell.

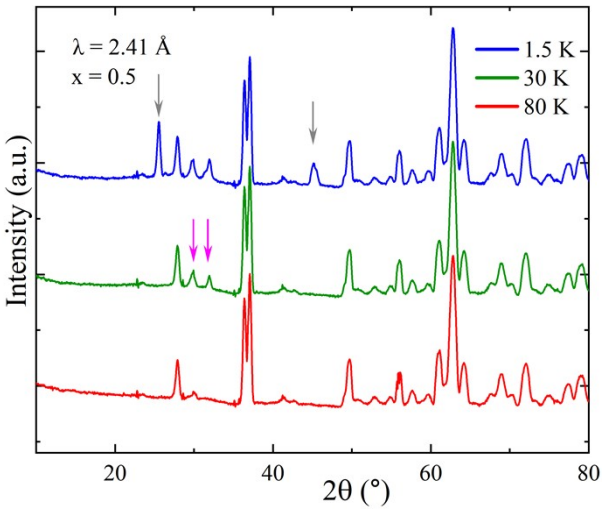

Fig. S6 High flux NPD data (λ = 2.41 Å) for x = 0.5 collected at 1.5, 30 and 80 K. Magnetic intensities from DPv and NTO-type phases are denoted by grey and pink arrows.

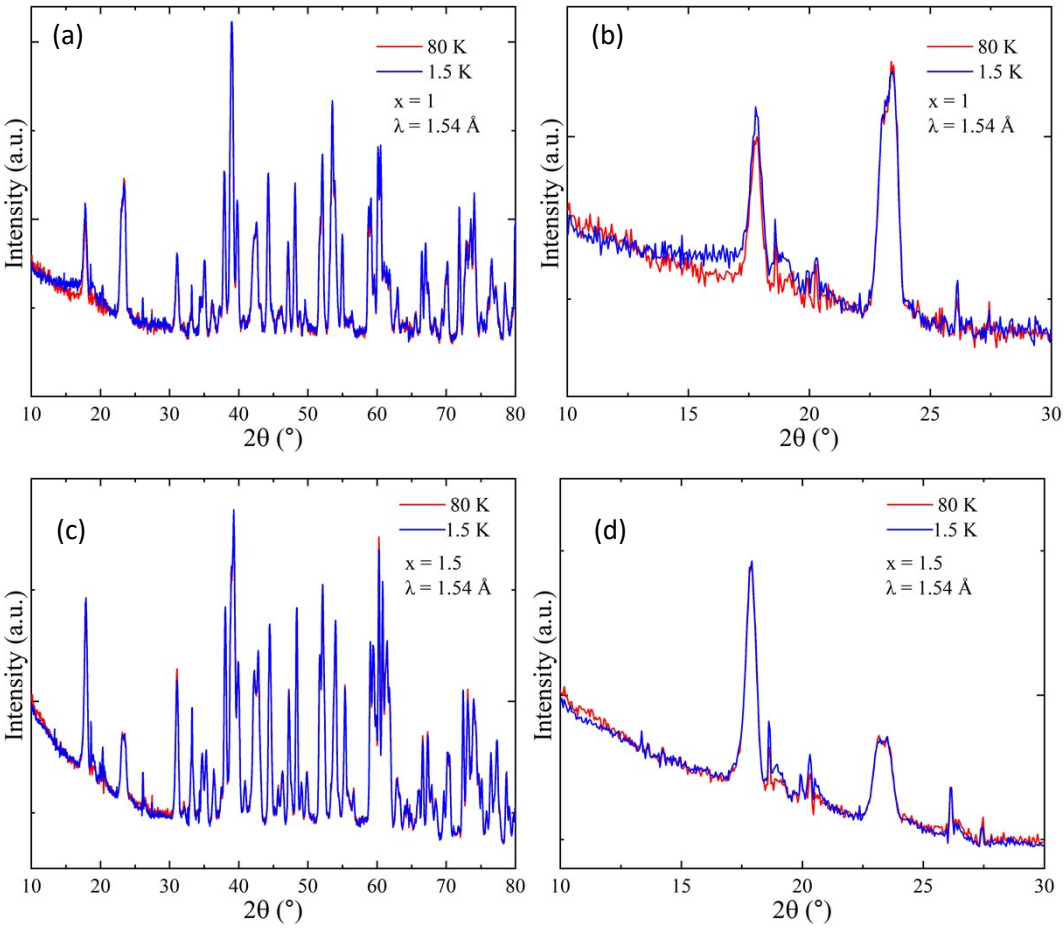

Fig. S7 Comparison of 80 K and 1.5 K high resolution (λ = 1.54 Å) NPD data of DPv (a) x = 1, (b) zoomed comparison of neutron diffraction data of x = 1, (c) x = 1.5 and (d) zoomed comparison of neutron diffraction data of x = 1.5. A broad low-angle enhancement centred at around 2ϑ = 18˚ is clearly observed in the zoomed x = 1 data, consistent with short-range magnetic correlations.

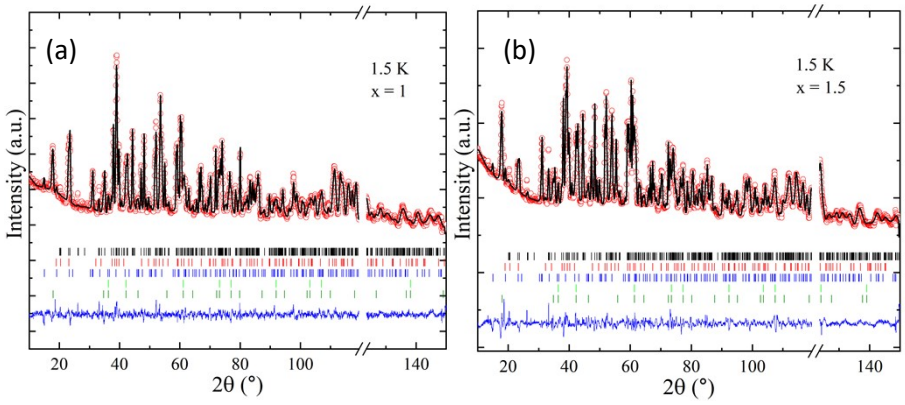

Fig. S8 Rietveld fits of 1.5 K high resolution (λ = 1.54 Å) NPD data for (a) x = 1 and (b) x = 1.5. DPv, NTO phase are denoted by black and red tick marks. Secondary phases are denoted same as in Fig. **S5**.

Table S1 Structural information for DPv $Mn_{1.5}Co_{0.5}ScSbO_6$ from the 300 K NPD Rietveld fit in monoclinic space group $P2_1/n$. Lattice parameters: a = 5.2803(3), b = 5.4443(3), c = 7.7222(4) Å and β = 90.285(4)˚.

Atom/site	x	y	z	Biso (Å^2)	Occupancy
Mn/Sc	0.014(3)	0.115(2)	0.724(2)	0.45(1)	0.748(3)/0.252(3)
Sc/Co	0.50	0.00	0.00	0.45(1)	0.540(7)/0.460(7)
Sb	0.00	0.50	0.00	0.45(1)	1.0
O1	0.314(1)	0.328(1)	0.940(1)	1.1(1)	1.0
O2	0.329(1)	0.303(1)	0.578(1)	1.1(1)	1.0
O3	0.875(7)	0.432(7)	0.760(6)	1.1(1)	1.0
A-O1-A	103.9(8)	A-O2-A	104.7(5)	A-O3-A	136.2(7)
B-O1-B′	139.0(2)	B-O2-B′	135.3(2)	B-O3-B′	136.4(2)

Residuals are R_p = 1.65%, R_{wp} = 2.17%, R_{Bragg} = 7.29%, R_F = 4.15%, and χ^2 = 4.30

Table S2 Structural information for DPv $MnCoScSbO_6$ from the 300 K NPD Rietveld fit in monoclinic space group $P2_1/n$. Lattice parameters: a = 5.2653(3), b = 5.4155(2), c = 7.7026(4) Å and β = 90.410(3)˚.

Atom/site	x	y	z	Biso (Å^2)	Occupancy
Mn/Co/Sc	0.017(2)	0.085(2)	0.730(2)	0.9(1)	0.51(3)/0.214(7)/0.286(7)
Sc/Co	0.50	0.00	0.00	0. 9(1)	0.428(7)/0.572(7)
Sb	0.00	0.50	0.00	0.9(1)	1.0
O1	0.311(1)	0.318(1)	0.936(1)	0.3 (1)	1.0
O2	0.323(1)	0.308(1)	0.571(1)	0.3 (1)	1.0
O3	0.870(1)	0.421(1)	0.760(1)	0.3 (1)	1.0
A-O1-A	102.1(5)	A-O2-A	103.3(4)	A-O3-A	137.0(5)
B-O1-B′	139.8(4)	B-O2-B′	136.5(3)	B-O3-B′	135.1(3)

Residuals are R_p = 2.05%, R_{wp} = 2.73%, R_{Bragg} = 8.61%, R_F = 5.49%, and χ^2 = 6.31

Table S3 Structural information for DPv $Mn_{0.5}Co_{1.5}ScSbO_6$ from the 300 K NPD Rietveld fit in monoclinic space group $P2_1/n$. Lattice parameters: a = 5.2521(2), b = 5.3808(2), c = 7.6857(3) Å and β = 90.571(3)˚.

Atom/site	x	y	z	Biso (Å^2)	Occupancy
Mn/Co/Sc	0.020(1)	0.065(1)	0.738(1)	1.0(1)	0.26(2)/0.435(4)/0.315(4)
Sc/Co	0.50	0.00	0.00	1.0(1)	0.370(4)/0.630(4)
Sb	0.00	0.50	0.00	1.0(1)	1.0
O1	0.314(1)	0.325(1)	0.938(1)	1.0(1)	1.0
O2	0.330(1)	0.308(1)	0.577(1)	1.0(1)	1.0
O3	0.869(1)	0.418(1)	0.761(1)	1.0(1)	1.0
A-O1-A	105.3(4)	A-O2-A	104.3(3)	A-O3-A	133.6(3)
B-O1-B′	138.6(3)	B-O2-B′	135.1(3)	B-O3-B′	133.9(3)

Residuals are R_p = 1.86%, R_{wp} = 2.37%, R_{Bragg} = 7.43%, R_F = 4.40%, and χ^2 = 3.05