Supplementary Materials: Magnetic transitions in the Co-modified Mn_2Sb system

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Table S1: Nominal vs. actual compositions for $Mn_{1.9}Co_{0.1}Sb$. The errors are standard deviations multiplied by a factor of 3.

Element	Nominal composition [wt. %]	Actual composition [wt.%]
Mn	45.0	46.1(1.5)
Co	2.5	2.6(0.6)
Sb	52.5	52.1(4.8)

Table S2: Nominal vs. actual compositions for $Mn_{1.85}Co_{0.15}Sb$. The errors are standard deviations multiplied by a factor of 3.

Element	Nominal composition [wt. %]	Actual composition [wt.%]
Mn	43.8	42.9(1.8)
Co	3.8	3.9(1.8)
Sb	52.4	54.0(0.6)

Table S3: Nominal vs. actual compositions for $Mn_{1.8}Co_{0.2}Sb$. The errors are standard deviations multiplied by a factor of 3.

Element	Nominal composition [wt.%]	Actual composition [wt.%]
Mn	42.5	42.2(5.7)
Co	5.1	5.1(0.6)
Sb	52.4	51.4(4.5)

Table S4: Lattice parameters, unit-cell volumes, and *a*/*c* ratios obtained from the x-ray powder diffractograms at RT. The errors are standard deviations.

x	*a* [Å]	*c* [Å]	*V* [Å^3]	*a*/*c* ratio
0.1	4.0796(5)	6.4898(5)	108.011(16)	0.6286
0.15	4.0794(4)	6.4630(3)	107.551(10)	0.6312
0.2	4.0798(4)	6.4403(3)	107.198(09)	0.6335
0.1 [6]				0.6338
0.15 [7]				0.6358
0.2 [7]				0.6366
0.18 [8]				0.6351
0.2 [12]				0.6346

Table S5: *a* lattice parameters for each composition at different temperatures from the neutron data. The errors are standard deviations.

x	Temperature [K]	Shubnikov group	*a* lattice parameter [Å]
0.1	550	X	4.140(1)
	350	P4/nm'm'	4.113(2)
	297	P4/nm'm'	4.106(1)
	200	Pmm'n'	4.095(2)
	50	P[c]mcn + Pmm'n'	4.078(1)
0.2	550	X	4.138(2)
	350	P4/nm'm'	4.114(1)
	297	Pmm'n'	4.107(1)
	200	P[c]mcn + Pmm'n'	4.094(1)
	50	P[c]mcn	4.094(1)

Table S6: *c* lattice parameters for each composition at different temperatures from the neutron data. The errors are the standard deviations.

x	Temperature [K]	Shubnikov group	c lattice parameter [Å]
0.1	550	X	6.603(4)
	350	P4/nm'm'	6.541(5)
	297	P4/nm'm'	6.533(4)
	200	Pmm'n'	6.526(4)
	50	P[c]mcn	13.036(7)
		Pmm'n'	6.518
0.2	550	X	6.553(7)
	350	P4/nm'm'	6.490(2)
	297	Pmm'n'	6.485(3)
	200	P[c]mcn	12.951(6)
		Pmm'n'	6.476(3)
	50	P[c]mcn	12.862(5)

Table S7: Unit cell volumes for each composition at different temperatures from the neutron data. The errors are the standard deviations.

x	Temperature [K]	Shubnikov group	Unit cell volume [Å^3]
0.1	550	X	113.2(1)
	350	P4/nm'm'	110.7(1)
	297	P4/nm'm'	110.2(1)
	200	Pmm'n'	109.4(1)
	50	P[c]mcn	216.8(2)
		Pmm'n'	108.4(1)
0.2	550	X	112.2(1)
	350	P4/nm'm'	109.8(5)
	297	Pmm'n'	109.4(1)
	200	P[c]mcn	217.1(1)
		Pmm'n'	108.6(1)
	50	P[c]mcn	215.6(1)

Table S8: z-coordinates from Rietveld refinements of the neutron powder diffractograms at five temperatures (550K, 350 K, RT, 200 K, 50 K) for x = 0.2 and x = 0.1. The errors are the standard deviations.

x	Temperature [K]	Shubnikov group	$z_{Mn\ II}$	$z_{Sb\ I}$
0.1	550	X	0.204(2)	0.781(1)
	350	P4/nm'm'	0.199(3)	0.772(2)
	RT	P4/nm'm'	0.198(3)	0.769(2)
	200	Pmm'n'	0.186(4)	0.756(5)
	50	P[c]mcn	0.095(1)	0.382(1)
		Pmm'n'	0.191	0.763
0.2	550	X	0.208(4)	0.781(1)
	350	P4/nm'm'	0.204(2)	0.771(1)
	RT	Pmm'n'	0.197(2)	0.765(2)
	200	P[c]mcn	0.097(1)	0.381(1)
		Pmm'n'	0.194	0.762
	50	P[c]mcn	0.145(1)	0.363(1)

Table S9: Isotropic thermal displacement parameters U_{iso} from neutron powder diffraction at five temperatures (550K, 350 K, RT, 200 K, 50 K) for x = 0.2 and x = 0.1. The errors are the standard deviations.

x	Temperature [K]	Shubnikov group	$U_{iso\ Mn\ I}$	$U_{iso\ Mn\ II}$	$U_{iso\ Sb\ I}$
0.1	550	X	0.037(9)	0.034(7)	0.024(5)
	350	P4/nm'm'	0.020(5)	0.020(5)	0.020(5)
	RT	P4/nm'm'	0.021(4)	0.021(4)	0.021(4)
	200	Pmm'n'	0.033(4)	0.033(4)	0.033(4)
	50	P[c]cmn + Pmm'n'	0.023	0.023	0.023
0.2	550	X	0.04(2)	0.027(9)	0.053(9)
	350	P4/nm'm'	0.018(2)	0.018(2)	0.018(2)
	RT	Pmm'n'	0.022(3)	0.022(3)	0.022(3)
	200	P[c]cmn + Pmm'n'	0.023	0.023	0.023
	50	P[c]cmn	0.016(3)	0.016(3)	0.016(3)

Table S10: Co occupancy on the Mn1 site at 550 K – stoichiometric vs. refined values.

x	Stoichiometric Co1 occupancy	Refined Co1 occupancy
0.1	0.012	0.015(4)
0.15	0.019	0.019(3)
0.2	0.025	0.025(5)

Table S11. Final agreement factors of the magnetic structure refinements for the $Mn_{1.9}Co_{0.1}Sb$ (350K, RT, 200 K, 50 K) neutron powder diffractograms. The chosen final models are printed in bold. For the refinement at 50 K two phases were considered. Phase I corresponds to the magnetic symmetry given in the table, while phase II corresponds to the FRI-II state with the magnetic symmetry Pmm'n'. The volume fractions from the two-phase refinement are mentioned in the table.

350 K	**R_{obs} (%)**	**R_{wp} (%)**	**R_p (%)**
P4/nm'm'	**6.16**	**7.35**	**6.00**
Cm'me'	9.05	8.79	6.67
Pmm'n'	9.17	8.82	6.66

RT	**R_{obs} (%)**	**R_{wp} (%)**	**R_p (%)**
P4/nm'm'	**5.99**	**7.59**	**5.40**
Cm'me'	9.28	9.36	6.14
Pmm'n'	9.84	9.38	6.10

200 K	**R_{obs} (%)**	**R_{wp} (%)**	**R_p (%)**
P4/nm'm'	13.34	13.98	9.44
Cm'me'	11.27	13.33	8.77
Pmm'n'	**6.96**	**9.73**	**7.54**

50 K	**R_{obs} (%) phase I/II**	**R_{wp} (%)**	**R_p (%)**	**Volume fractions phase I/II**
P[c]mcn	3.00/3.13	9.36	6.67	0.21(5)/0.79(5)
R factors from magnetic reflections	**R_{obs} (%)**	**wR_{obs}**	**R_{all} (%)**	**wR_{all}**
P[c]mcn	9.71	10.48	3.91	7.30

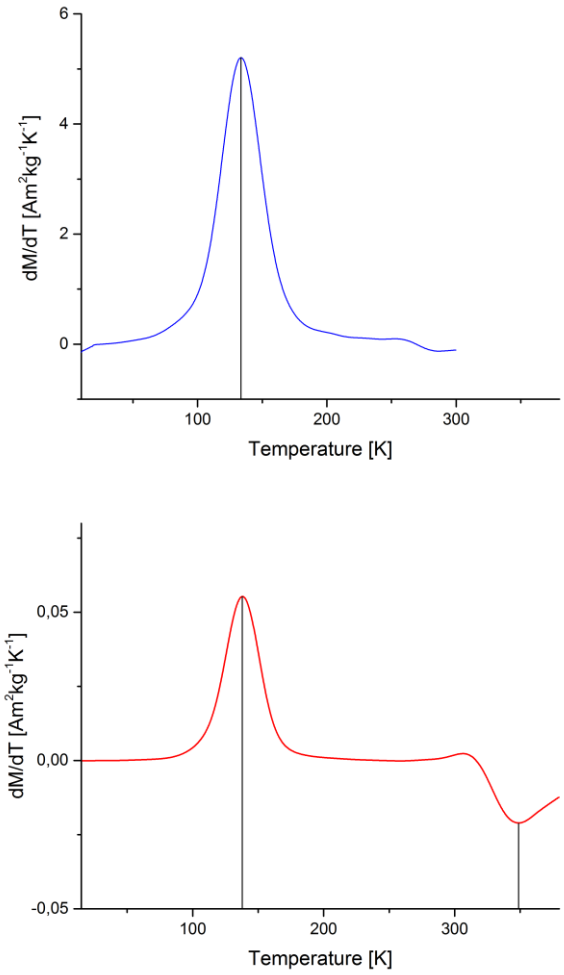

Figure S1: d*M*/d*T* curves for the cooling (top) and heating (bottom) cycles on $Mn_{1.8}Co_{0.2}Sb$. The lines indicate the transition temperatures.

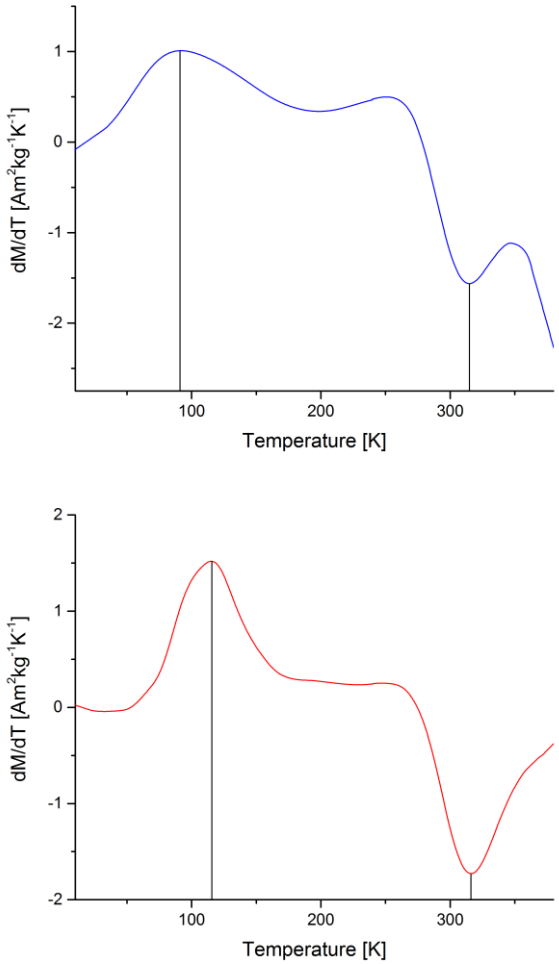

Figure S2: dM/dT curves for the cooling (top) and heating (bottom) cycles on $Mn_{1.85}Co_{0.15}Sb$. The lines indicate the transition temperatures.

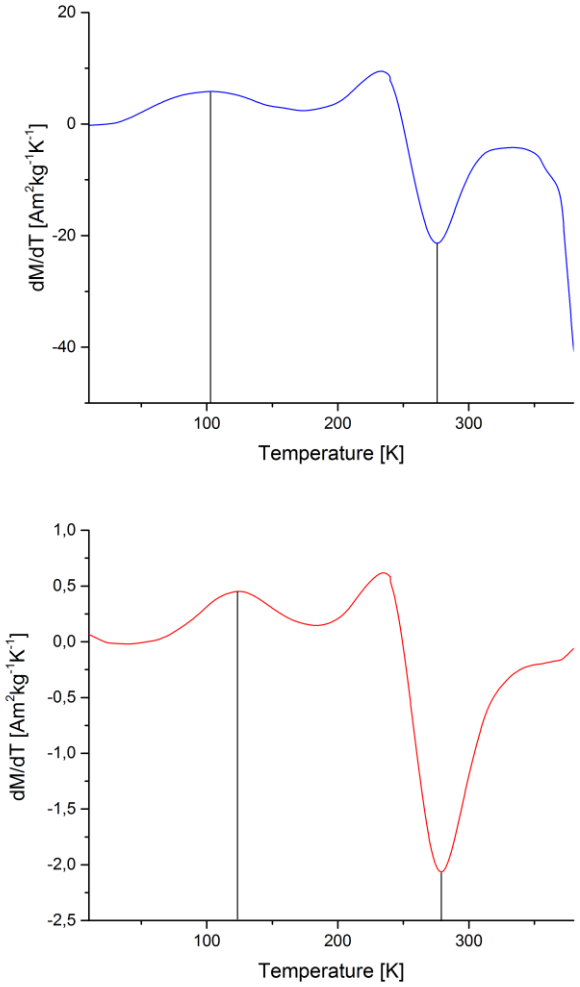

Figure S3: d*M*/d*T* curves for the cooling (top) and heating (bottom) cycles on $Mn_{1.9}Co_{0.1}Sb$. The lines indicate the transition temperatures.

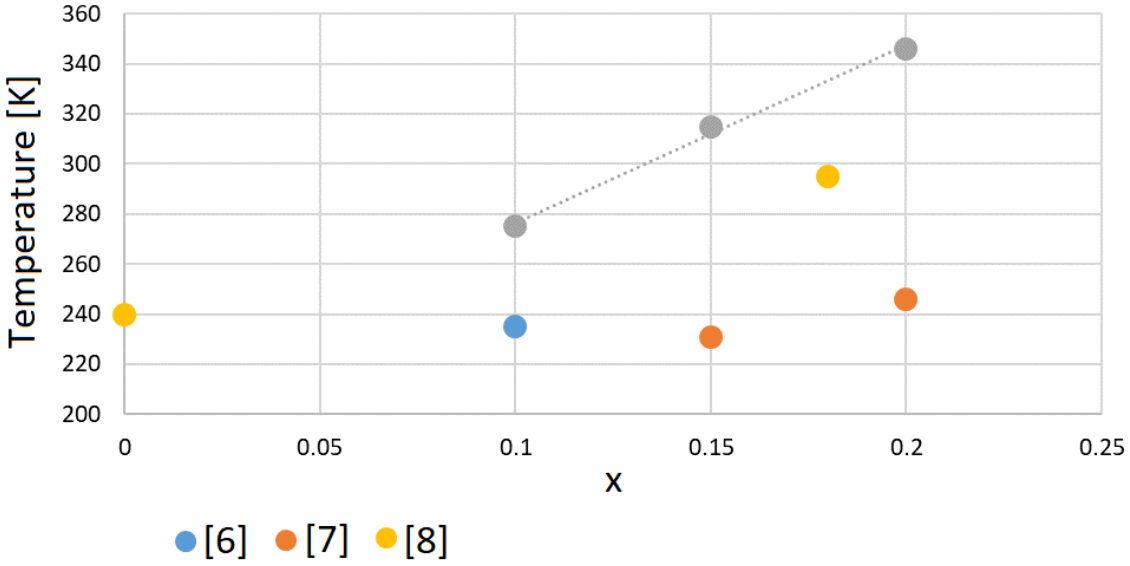

Figure S4: Spin flip transition temperatures T_{SF} in $Mn_{2-x}Co_xSb$ compounds as a function of the Co content. The gray symbols stand for the powder data of this study. All other symbols represent the data from the literature.

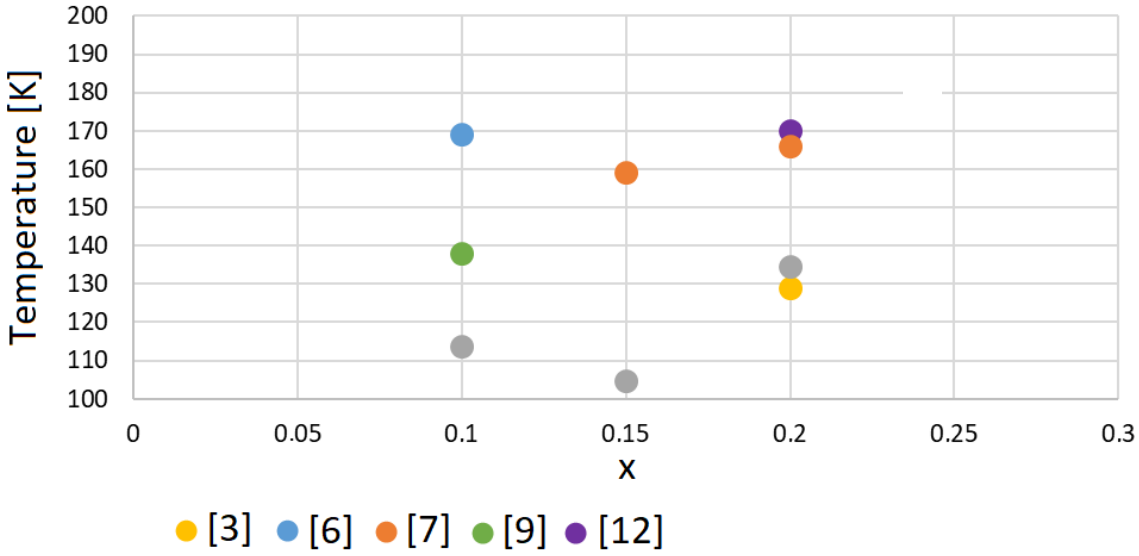

Figure S5: Magnetic phase transition temperatures T_t in $Mn_{2-x}Co_xSb$ compounds as a function of the Co content. The gray symbols stand for the powder data of this study. All other symbols represent the data from the literature.

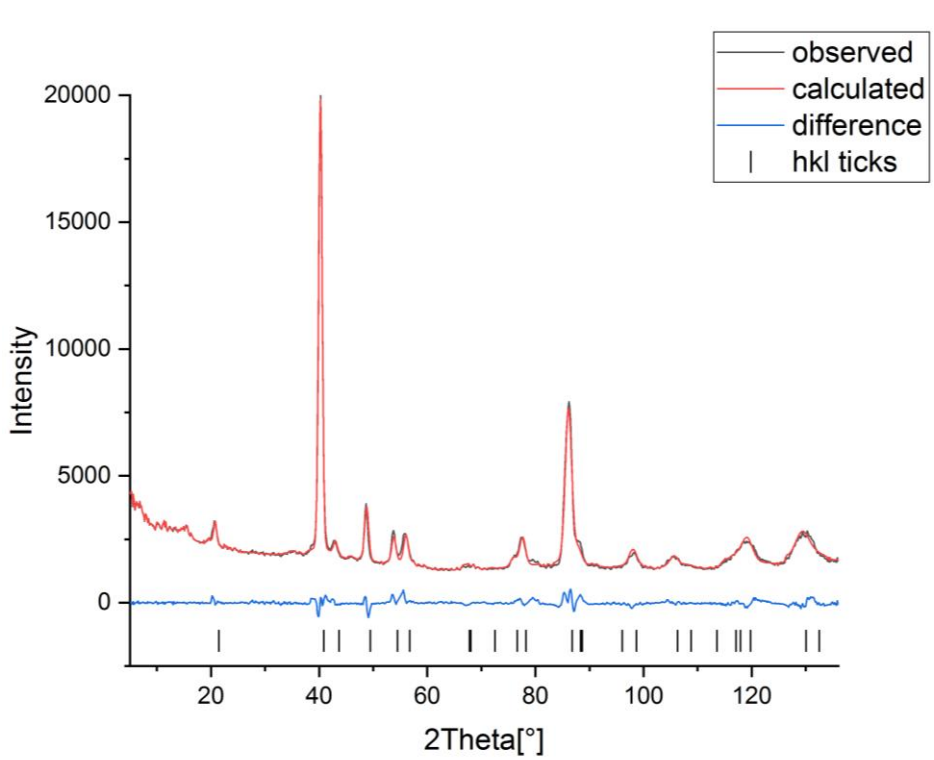

Figure S6: Rietveld refinement of the neutron powder diffractogram of $Mn_{1.85}Co_{0.15}Sb$ at 550 K (R_{obs} = 3.92%, R_{wp} = 4.18%, R_p = 2.89%).

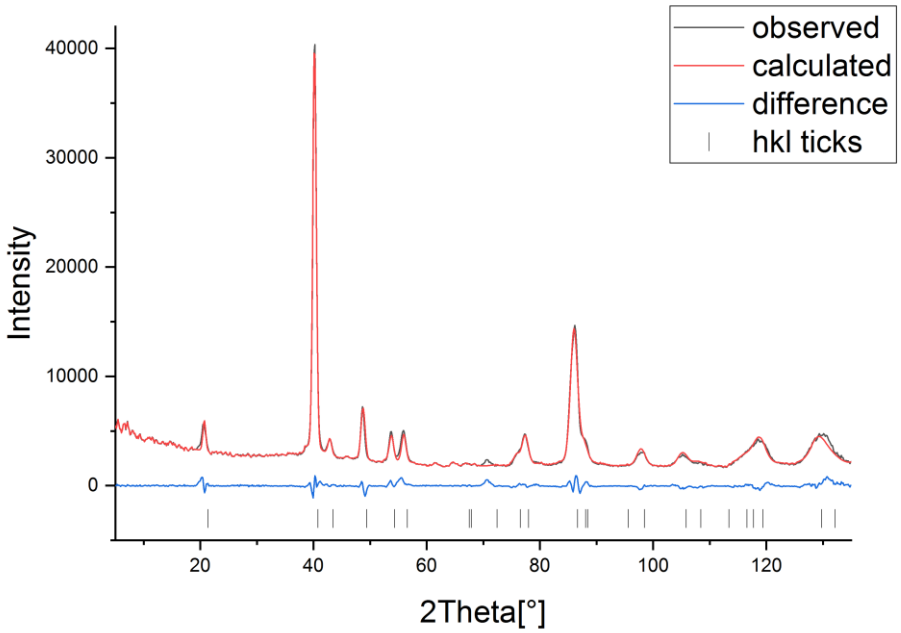

Figure S7: Rietveld refinement of the neutron powder diffractogram of $Mn_{1.9}Co_{0.1}Sb$ at 550 K (R_{obs} = 3.84%, R_{wp} = 5.09%, R_p = 3.18%).

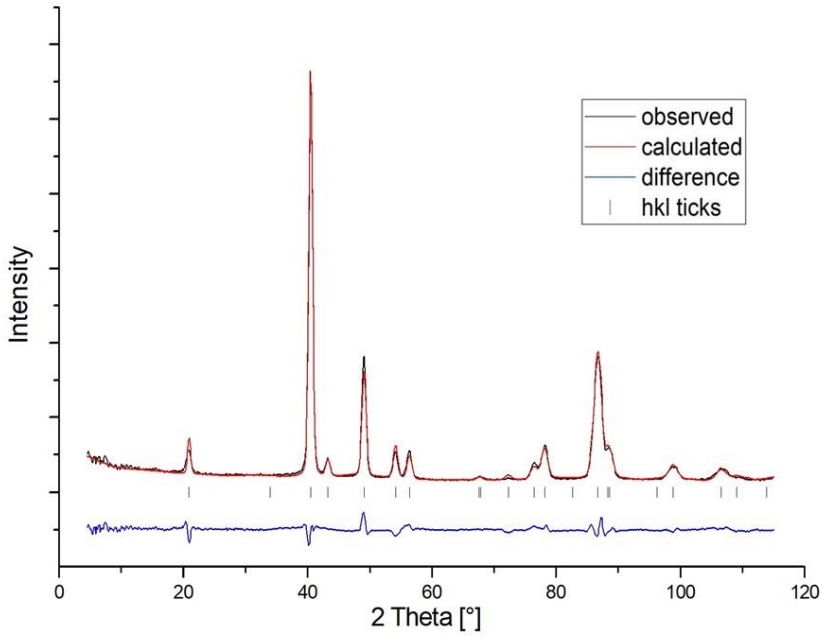

Figure S8: Magnetic refinement of the neutron powder diffractogram of $Mn_{1.9}Co_{0.1}Sb$ at 350 K (P4/nm′m′ Shubnikov group).

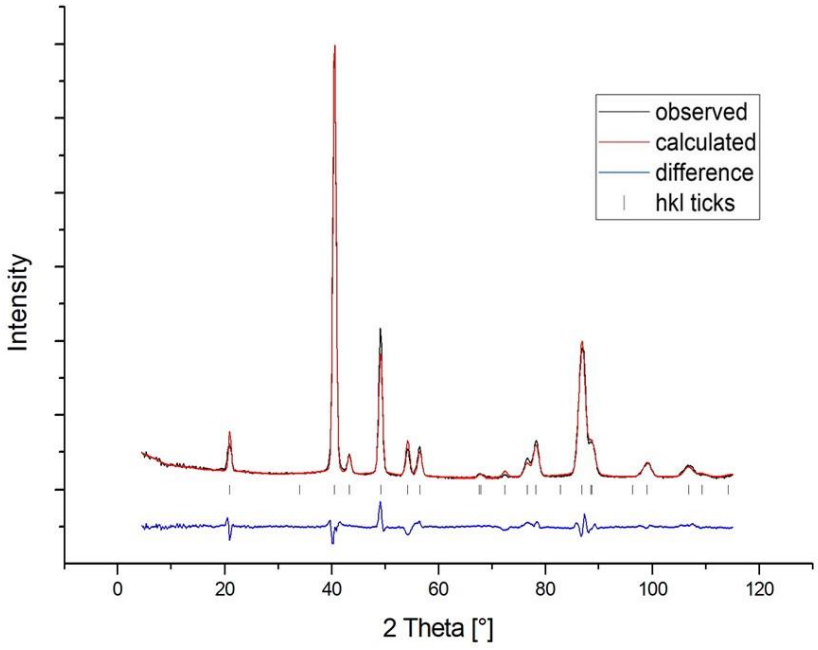

Figure S9: Magnetic refinement of the neutron powder diffractogram of $Mn_{1.9}Co_{0.1}Sb$ at room temperature (P4/nm′m′ Shubnikov group).

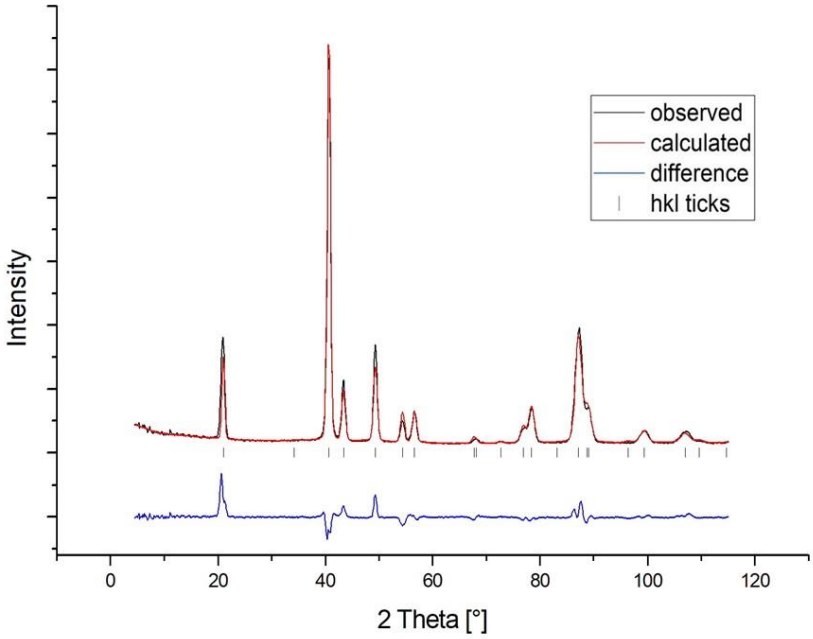

Figure S10: Magnetic refinement of the neutron powder diffractogram of $Mn_{1.9}Co_{0.1}Sb$ at 200K (Pmm'n' Shubnikov group).

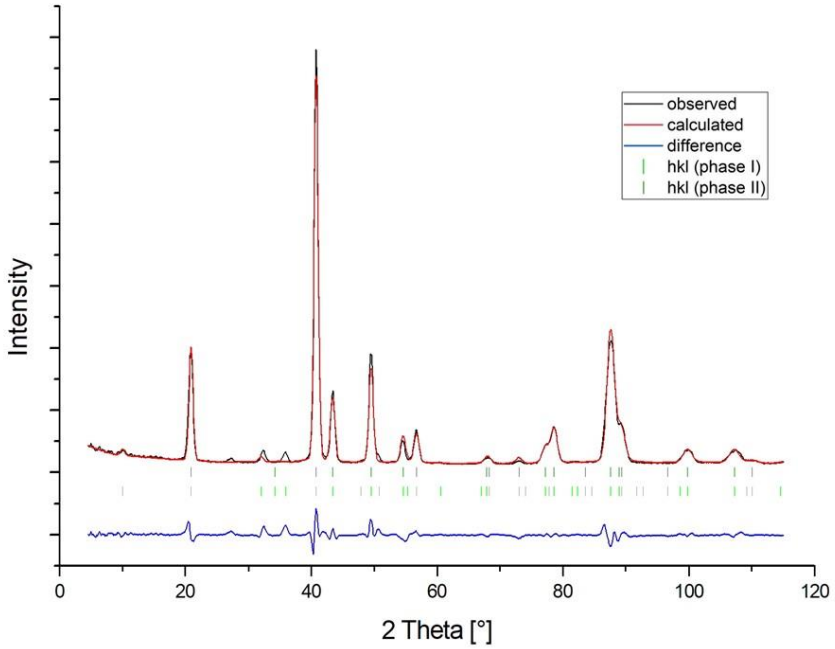

Figure S11: Magnetic refinement of the neutron powder diffractogram of $Mn_{1.9}Co_{0.1}Sb$ at 50K – phase I: P[c]mcn Shubnikov group; phase II: Pmm'n' Shubnikov group.