Supplementary Material to the Paper Titled:

Unraveling the Magnetic Ground State and Local lattice Distortions in Z_2XY-Type Full Heusler Compounds: An EXAFS Study

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S1: Elemental analysis using Energy Dispersive X–Ray (EDX) Spectroscopy

The elemental ratio represents an average value obtained after recording several EDX spectra from different spatial locations on the flat piece of the sample. A standard deviation of about 3% in the mean value was obtained for all compositions.

Table S1: Elemental analysis using EDX.

Composition	Ga	Al	Mn	Co	Pd
Ga_2MnCo	51.4	–	25	23.7	–
Ga_2MnPd	49.49	–	25.75	–	24.76
Al_2MnCo	–	49.85	25.32	24.83	–
Al_2MnPd	–	51.4	24.17	–	24.43

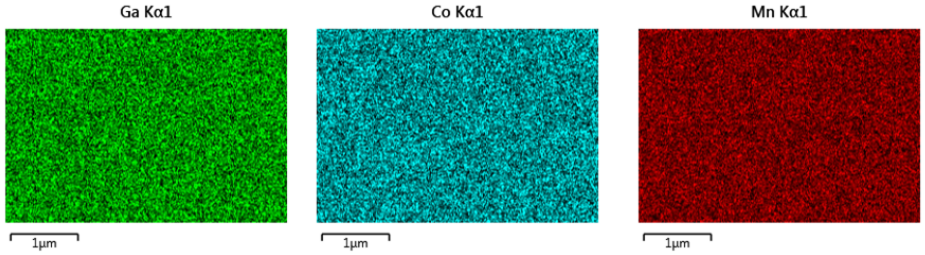

Figure S1: Elemental mapping of Ga_2MnCo.

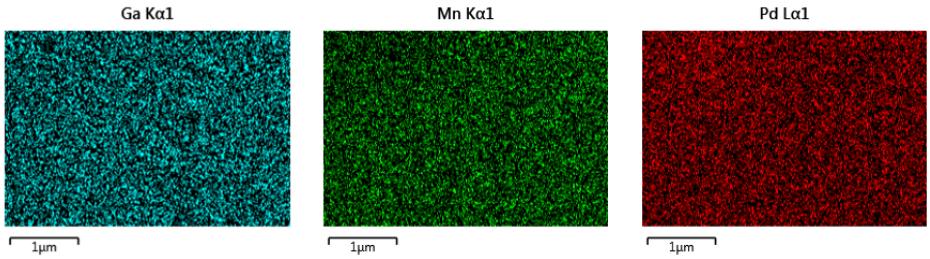

Figure S2: Elemental mapping of Ga_2MnPd.

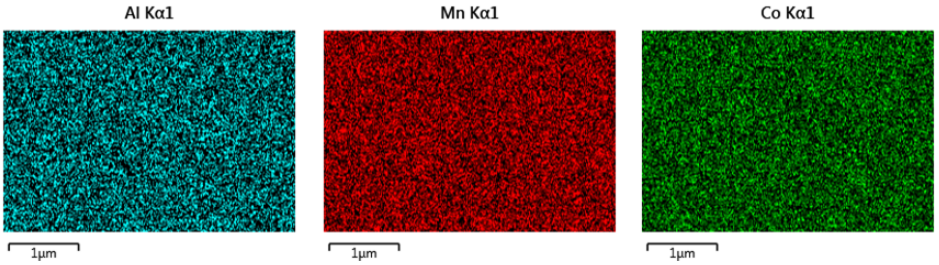

Figure S3: Elemental mapping of Al_2MnCo.

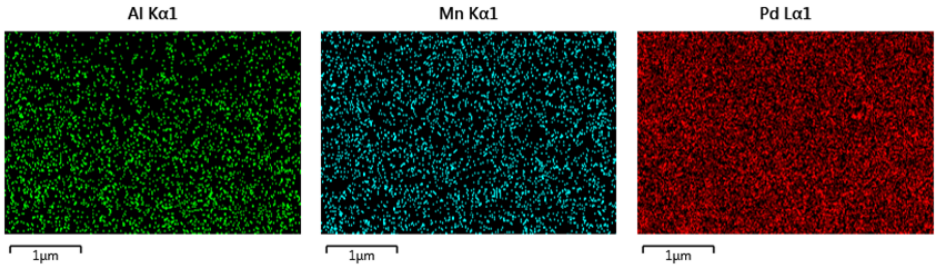

Figure S4: Elemental mapping of Al_2MnPd.

S3: Rietveld refinement

The crystal structure was investigated by recording the powder x-ray diffraction (XRD) profile using high resolution synchrotron source. These measurements were carried out in capillary mode and the capillary was rotated at 150 rpm to reduce the orientation effects. Desired wave length for AD-XRD diffraction experiments was selected using a Si(111) channel cut monochromator. The monochromatic beam was focused on to the sample with a Kirkpatrick-Baez mirror. Data was recorded using an image plate area detector. The sample to detector distance and the beam wavelength were calibrated using NIST standards

LaB_6. The data was analyzed by Rietveld method. The parameters of the refinement can be found in Table S2. The XRD profiles along with the fitting are shown in Fig. S5 and S6.

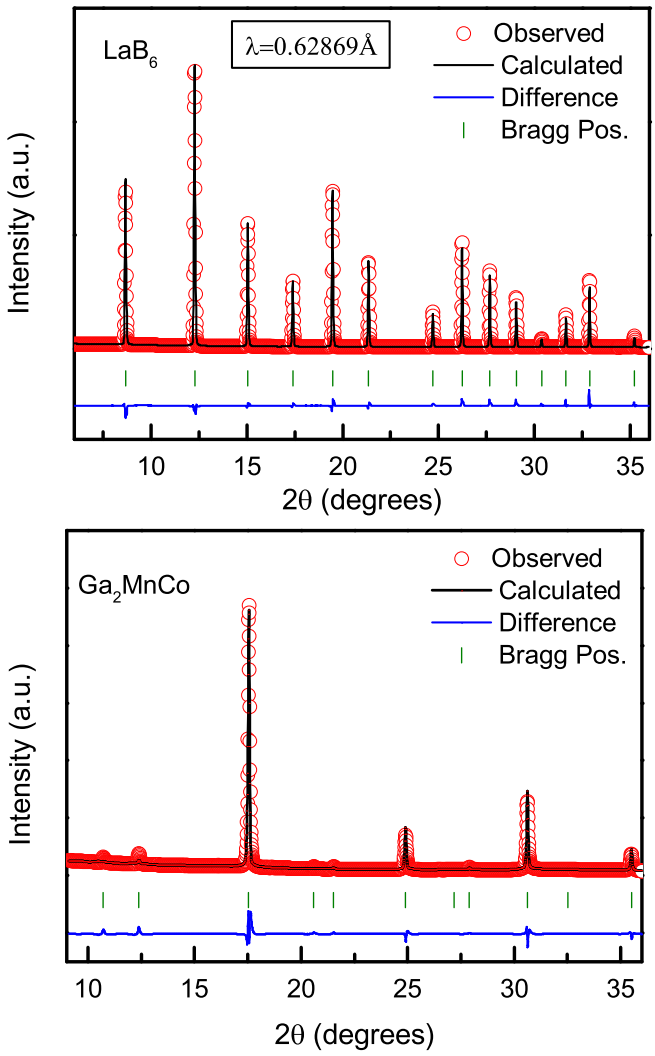

Figure S5: The Rietveld refinement of the standard sample and Ga_2MnCo composition.

Table S2: Parameters from the Rietveld refinement of the high–resolution synchrotron XRD patterns of the standard LaB_6 and all four Z_2XY compositions .

	Space group	Wyckoff positions	Lattice constant (a)	R–factors
LaB_6 (standard)	$Pm\bar{3}m$	La (1a) B (6f)	4.156 Å	R_{wp} = 13.0 R_{exp} = 3.07 R_{Bragg} = 4.2
Ga_2MnCo ($L2_1$+B2 disorder)	$Fm\bar{3}m$	Ga (8c) Mn (4a) Co (4b) Mn (4b) Co (4a)	5.8315(2) Å	R_{wp} = 10.1 R_{exp} = 7.2 R_{Bragg} = 5.09
Ga_2MnPd ($L2_1$)	$Fm\bar{3}m$	Ga (8c) Mn (4a) Pd (4b)	6.0631(3) Å	R_{wp} = 4.54 R_{exp} = 3.58 R_{Bragg} = 2.82
Al_2MnCo (B2)	$Pm\bar{3}m$	Al (1a) Mn (1b) Co (1b)	2.9109(9) Å	R_{wp} = 5.03 R_{exp} = 2.96 R_{Bragg} = 2.87
Al_2MnPd (B2; main phase $\sim$ 76%)	$Pm\bar{3}m$	Al (1a) Mn (1b) Pd (1b)	2.9789(7) Å	R_{wp} = 8.01 R_{exp} = 3.11 R_{Bragg} = 1.85
Al_3Pd_2 (secondary phase $\sim$ 24%)	$P\bar{3}m1$	Al (1a) Al (2d) Pd (2d)	4.1961(2) Å 5.1209(5) Å	R_{wp} = 8.01 R_{exp} = 3.11 R_{Bragg} = 8.88

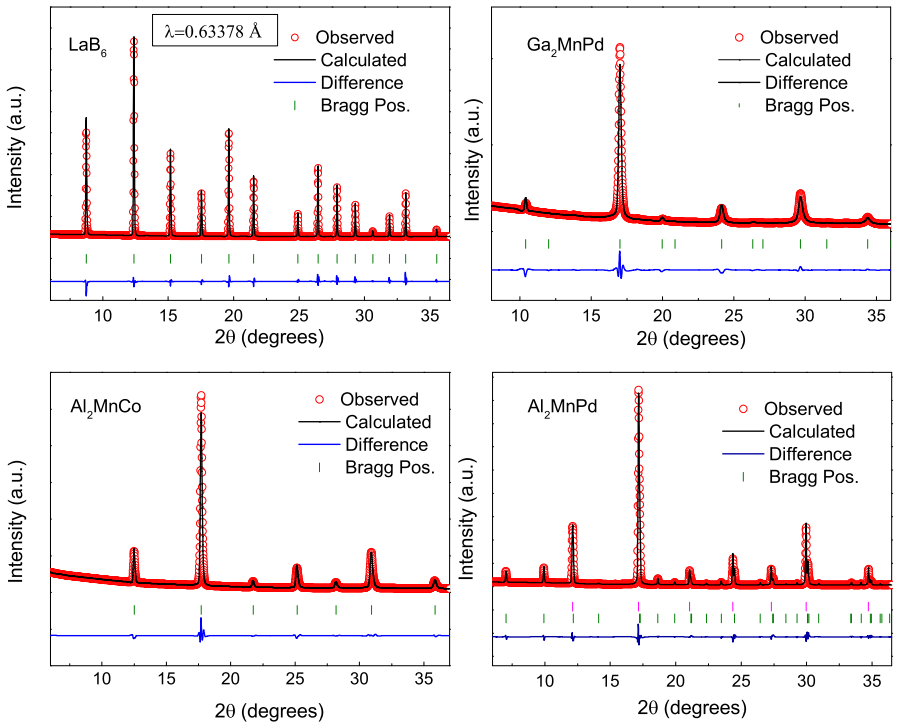

Figure S6: The Rietveld refinement of the standard sample and Ga_2MnPd, Al_2MnCo, and Al_2MnPd compositions. The refinement of Al_2MnPd is done considering the secondary Al_3Pd_2 binary phase.

S4: EXAFS spectra of the compositions

We have examined the local structure of the present compositions by means of EXAFS spectroscopy recorded at multiple K-edges and at varying temperatures. Bond distances (R) and Debye–Waller factors (σ^2) have been extracted by the analysis of the EXAFS oscillations.

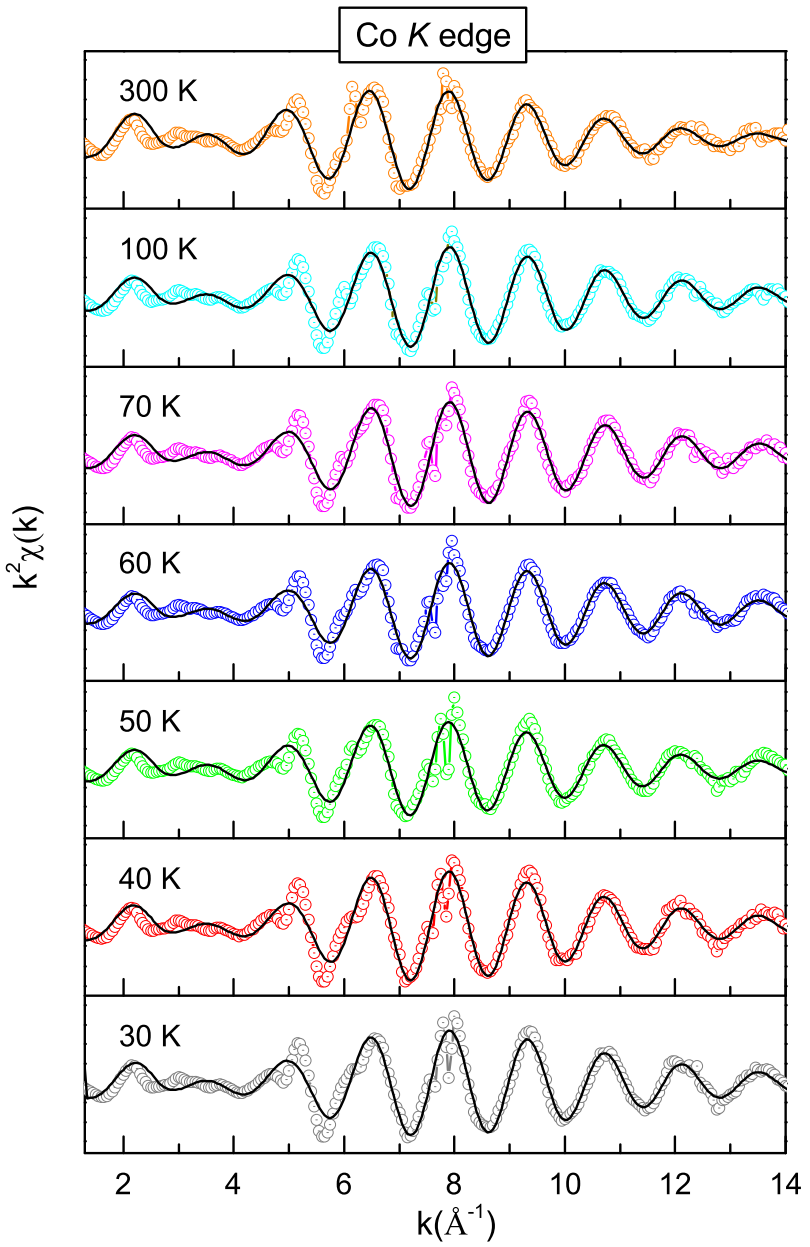

Figure S7: The k^2 weighted $\chi(k)$ spectra of Ga_2MnCo for Co K-edge in the temperature range 30 K - 300 K. Black solid lines denote the fit to the data.

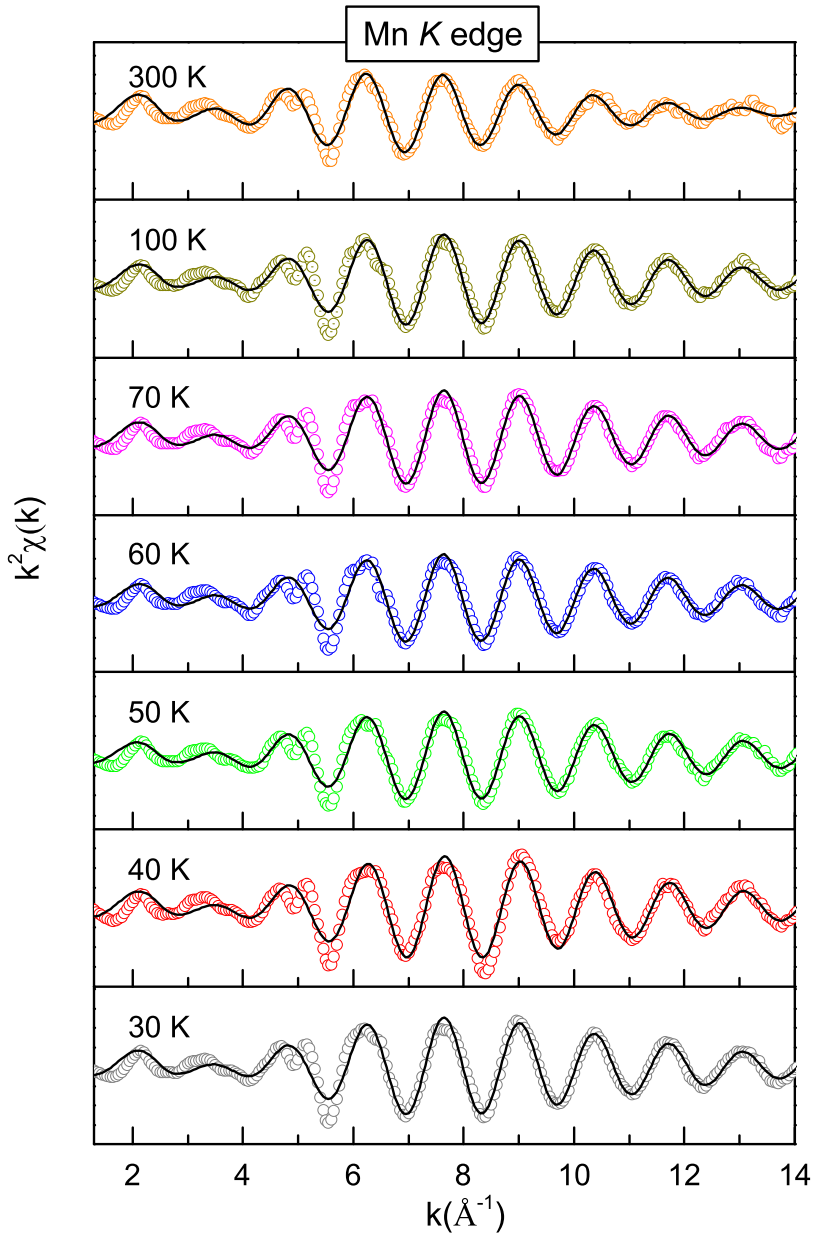

Figure S8: The k^2 weighted $\chi(k)$ spectra of Ga_2MnCo for Mn K-edge in the temperature range 30 K - 300 K. Black solid lines denote the fit to the data.

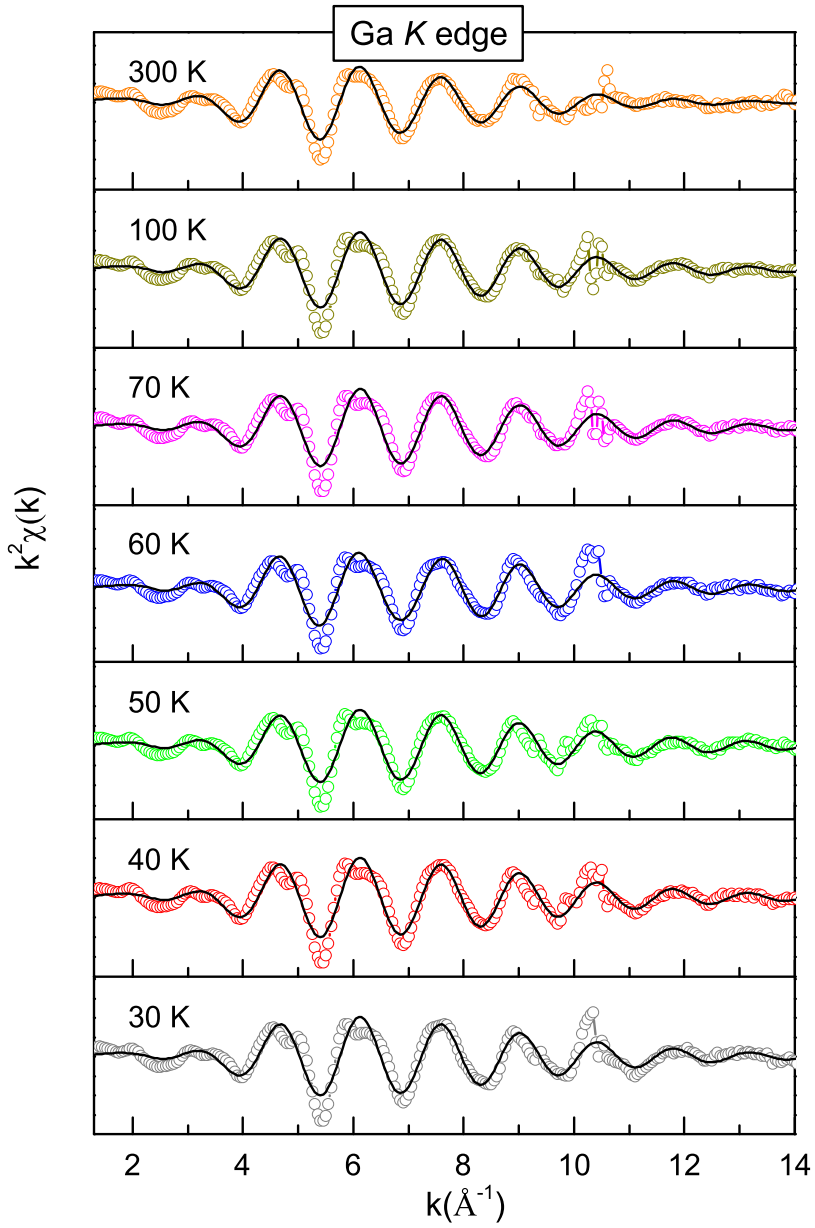

Figure S9: The k^2 weighted $\chi(k)$ spectra of Ga_2MnCo for Ga K-edge in the temperature range 30 K - 300 K. Black solid lines denote the fit to the data.

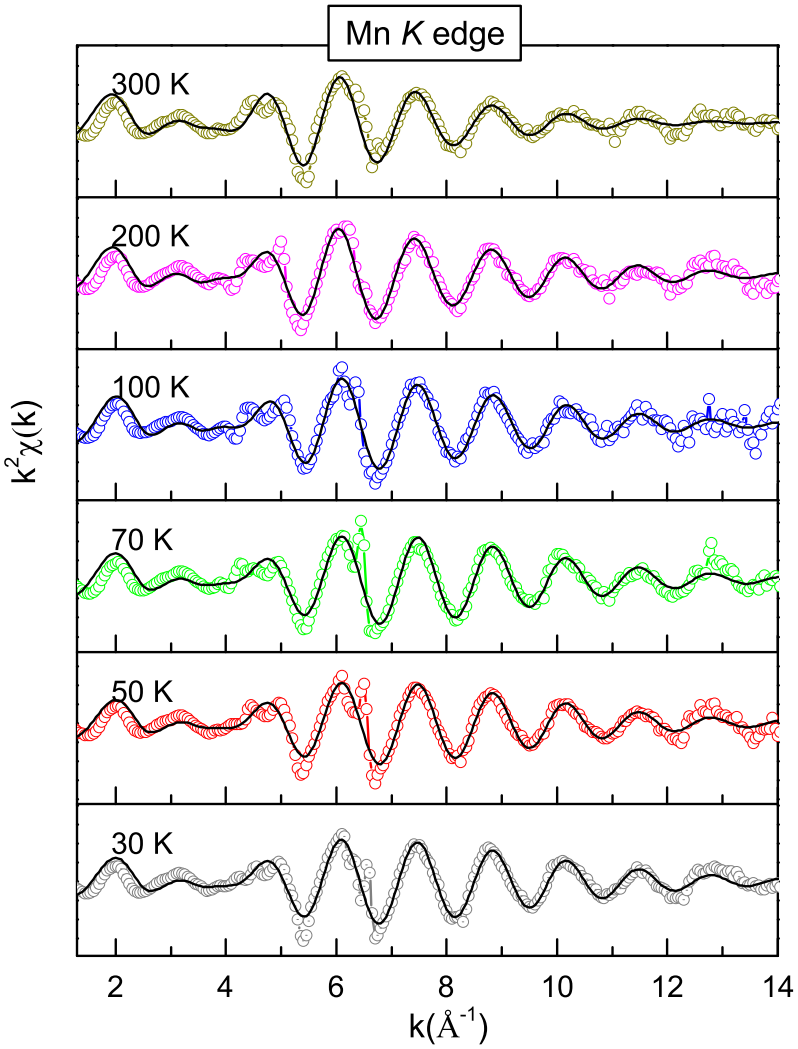

Figure S10: The k^2 weighted $\chi(k)$ spectra of Ga_2MnPd for Mn K-edge in the temperature range 30 K - 300 K. Solid lines denote the fit to the data.

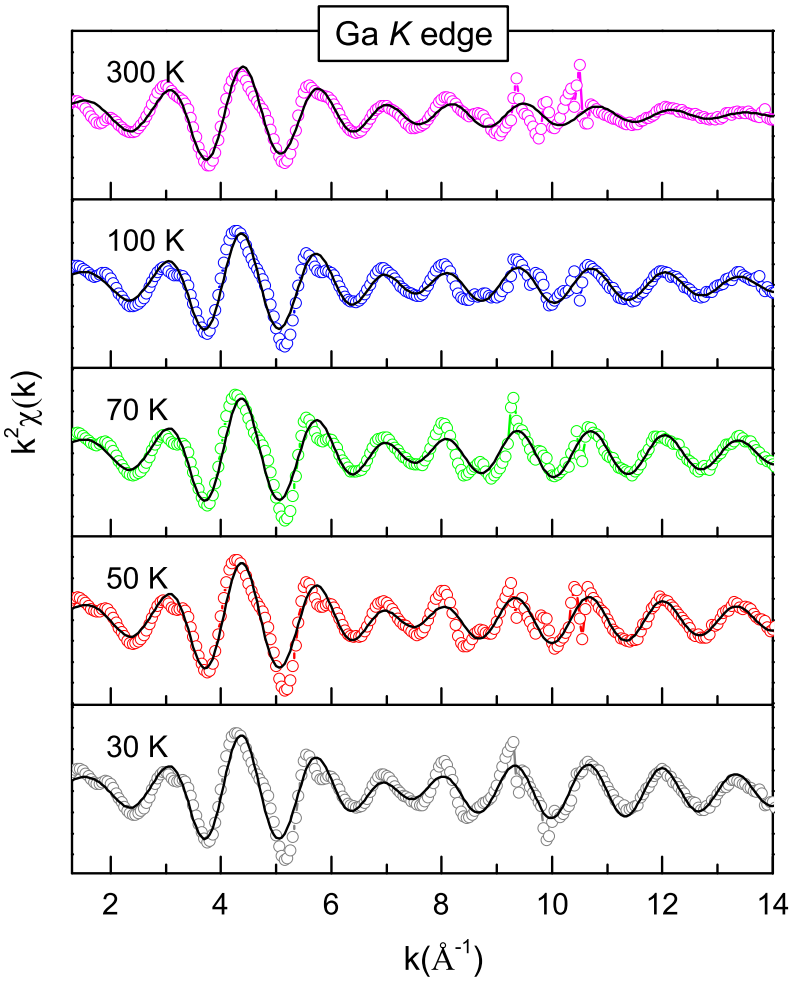

Figure S11: The k^2 weighted $\chi(k)$ spectra of Ga_2MnPd for Ga K-edge in the temperature range 30 K - 300 K. Solid lines denote the fit to the data.

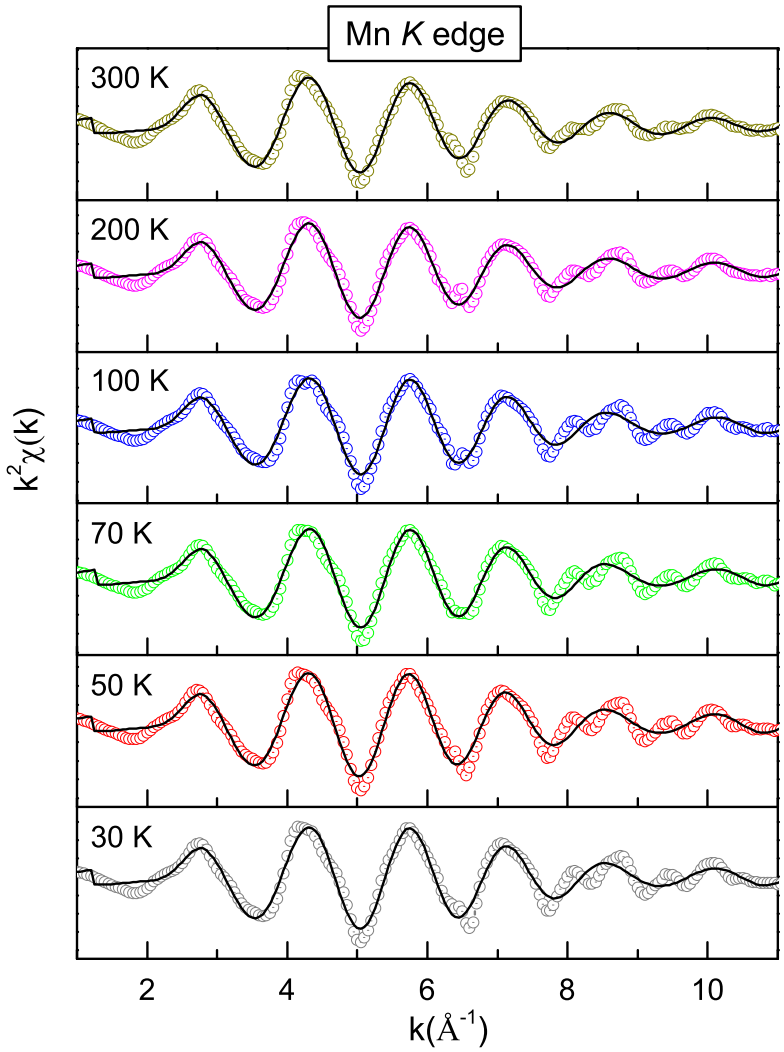

Figure S12: The k^2 weighted $\chi(k)$ spectra of Al_2MnCo for Mn K-edge in the temperature range 30 K - 300 K. Black lines denote the fit to the data.

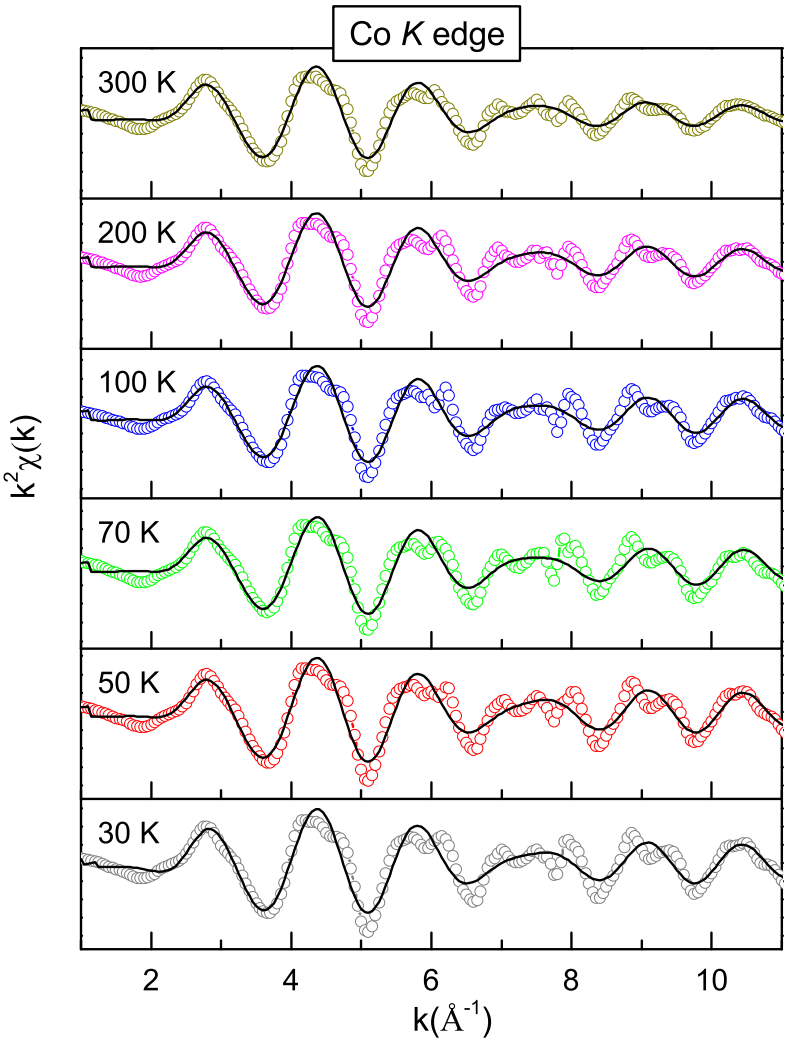

Figure S13: The k^2 weighted $\chi(k)$ spectra of Al_2MnCo for Co K-edge in the temperature range 30 K - 300 K. Black lines denote the fit to the data.

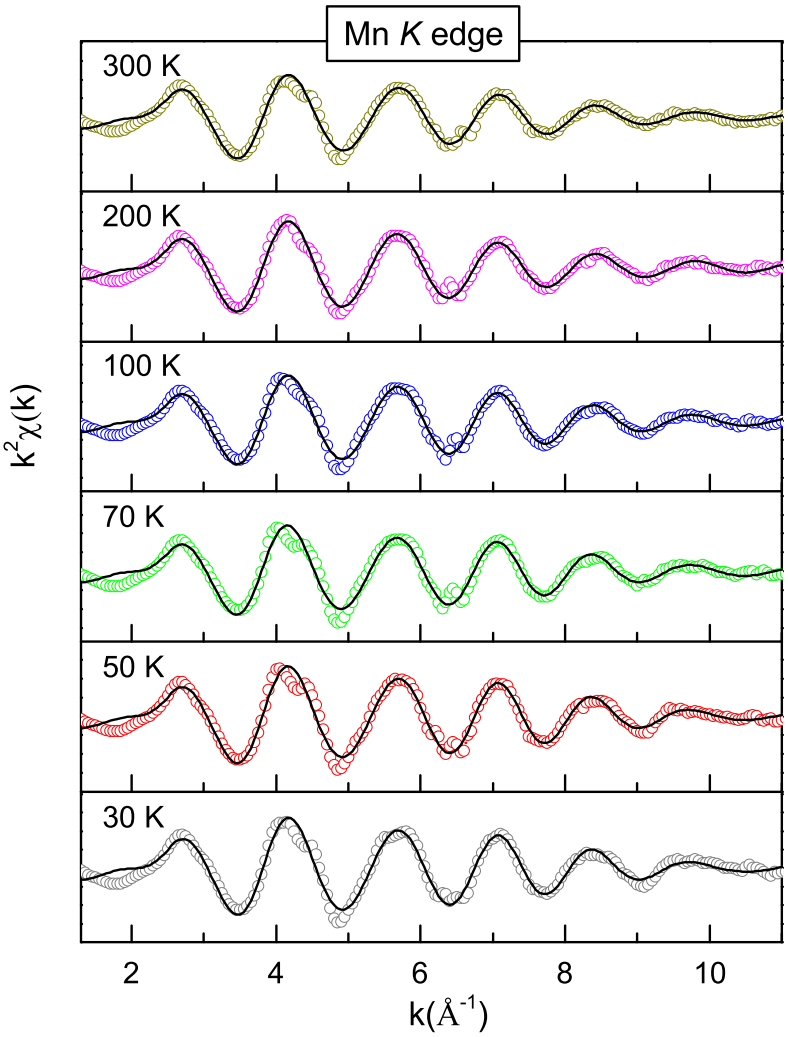

Figure S14: The k^2 weighted $\chi(k)$ spectra of Al_2MnPd for Mn K-edge in the temperature range 30 K - 300 K. Solid lines denote fit to the data.

Table S3: Local structural parameters obtained from EXAFS analysis of Ga_2MnCo. R denotes the bond distances obtained from EXAFS analysis, and σ^2 the mean-square disorder in R obtained from EXAFS analysis. The analysis were carried out in the k range 3-14 $Å^{-1}$ and R range 1-3 Å . Values in parenthesis represent the uncertainty in last digit.

	300 K		100 K		70 K		60 K		50 K	
Bonds	R (Å)	σ^2(Å^2)	R(Å)	σ^2(Å^2)	R(Å)	σ^2(Å^2)	R(Å)	σ^2(Å^2)	R(Å)	σ^2(Å^2)
Mn-Ga	2.588(3)	0.0088(2)	2.583(1)	0.0054(2)	2.583(1)	0.0051(1)	2.585(1)	0.0051(1)	2.577(1)	0.0043(1)
Mn-Mn	2.988(4)	0.05(22)	2.983(1)	0.039(14)	2.982(1)	0.047(17)	2.984(2)	0.057(13)	2.976(2)	0.053(22)
Mn-Co	2.719(1)	0.024(3)	2.738(2)	0.023(4)	2.747(1)	0.025(3)	2.749(2)	0.025(4)	2.679(2)	0.026(4)
Co-Ga	2.496(4)	0.0089(5)	2.491(6)	0.0064(2)	2.489(3)	0.0060(4)	2.492(4)	0.0058(3)	2.494(4)	0.0068(4)
Co-Co	2.882(5)	0.034(21)	2.876(6)	0.024(9)	2.874(4)	0.026(18)	2.877(4)	0.027(21)	2.879(4)	0.019(9)
Ga-Ga	3.14(6)	0.029(11)	3.15(7)	0.026(12)	3.15(12)	0.029(20)	3.19(6)	0.018(9)	3.18(6)	0.026(9)
	40 K		30 K							
Bonds	R (Å)	σ^2(Å^2)	R(Å)	σ^2(Å^2)						
Mn-Ga	2.578(1)	0.0047(1)	2.581(3)	0.0050(1)						
Mn-Mn	2.977(1)	0.038(22)	2.981(3)	0.035(8)						
Mn-Co	2.756(1)	0.028(9)	2.743(3)	0.020(3)						
Co-Ga	2.493(2)	0.0065(6)	2.492(4)	0.0060(4)						
Co-Co	2.878(3)	0.021(19)	2.878(5)	0.024(17)						
Ga-Ga	3.16(14)	0.026(23)	3.15(11)	0.027(19)						

Table S4: Local structural parameters obtained from EXAFS analysis of Ga_2MnPd. R denotes the bond distances, and σ^2 the mean-square disorder in R obtained from EXAFS analysis. The analysis were carried out in the k range 3-14 $Å^{-1}$ and R range 1-3 Å . Values in parenthesis represent uncertainty in the last digit.

T	Mn-Ga		Mn-Mn		Mn-Pd		Ga-Ga		Ga-Pd	
	R (Å)	σ^2 (Å^2)	R(Å)	σ^2(Å^2)	R (Å)	σ^2 (Å^2)	R(Å)	σ^2(Å^2)	R (Å)	σ^2(Å^2)
300	2.638(3)	0.0117(4)	2.849(4)	0.013(2)	3.046(4)	0.017(3)	3.004(4)	0.05(1)	2.609(4)	0.0072(8)
200	2.637(6)	0.0106(7)	2.847(7)	0.017(3)	3.044(5)	0.016(5)	–	–	–	–
100	2.633(3)	0.0100(4)	2.847(3)	0.023(6)	3.040(3)	0.018(5)	3.002(2)	0.04(1)	2.600(2)	0.0065(2)
70	2.632(4)	0.0093(4)	2.872(4)	0.021(5)	3.039(4)	0.024(9)	3.003(3)	0.03(1)	2.600(3)	0.0054(3)
50	2.630(2)	0.0090(3)	2.847(3)	0.022(5)	3.037(3)	0.025(8)	3.010(4)	0.04(2)	2.606(3)	0.0054(4)
30	2.631(2)	0.0087(3)	2.845(3)	0.019(2)	3.038(2)	0.021(5)	3.011(4)	0.04(2)	2.608(4)	0.0046(4)

Table S5: Local structural parameters obtained from EXAFS analysis of Al_2MnCo. R denotes the bond distances, and σ^2 the mean-square disorder in R obtained from EXAFS analysis. The analysis were carried out in the k range 2-10 $Å^{-1}$ and R range 1-3 Å . Values in parenthesis represent uncertainty in the digit.

	Mn-Al		Mn-Mn		Mn-Co		Co-Al		Co-Co	
T(K)	R (Å)	σ^2 (Å^2)	R(Å)	σ^2 (Å^2)	R (Å)	σ^2 (Å^2)	R(Å)	σ^2 (Å^2)	R (Å)	σ^2(Å^2)
300	2.574(1)	0.0070(2)	2.973(1)	0.044(13)	2.870(1)	0.0137(8)	2.476(1)	0.0070(3)	2.859(2)	0.0119(12)
200	2.571(2)	0.0057(3)	2.969(2)	0.032(6)	2.868(1)	0.0099(6)	2.473(2)	0.0055(4)	2.856(3)	0.0116(17)
100	2.568(2)	0.0045(4)	2.965(3)	0.033(12)	2.860(3)	0.0073(11)	2.471(4)	0.0043(6)	2.853(5)	0.0119(35)
70	2.570(1)	0.0044(3)	2.968(2)	0.033(9)	2.861(2)	0.0068(5)	2.471(5)	0.0040(7)	2.849(6)	0.0109(35)
50	2.575(2)	0.0042(3)	2.973(3)	0.032(13)	2.870(2)	0.0069(10)	2.469(3)	0.0041(4)	2.851(4)	0.0115(22)
30	2.572(2)	0.0044(3)	2.969(3)	0.033(9)	2.862(3)	0.0072(5)	2.479(3)	0.0042(3)	2.863(4)	0.0102(15)
20	2.569(2)	0.0045(4)	2.967(3)	0.030(8)	2.865(3)	0.0068(7)	2.472(4)	0.0043(5)	2.849(5)	0.0115(31)

Table S6: Local structural parameters obtained from EXAFS analysis of Mn K edge spectra of Al_2MnPd. R denotes the bond distances, and σ^2 the mean-square disorder in R obtained from EXAFS analysis. The analysis were carried out in the k range 2-10 $Å^{-1}$ and R range 1-3 Å . Values in parenthesis represent uncertainty in the last digit.

	Mn-Al		Mn-Mn		Mn-Pd	
T(K)	R (Å)	σ^2 ($Å^2$)	R(Å)	σ^2($Å^2$)	R (Å)	σ^2($Å^2$)
300	2.620(20)	0.0101(6)	2.824(23)	0.029(6)	3.026(23)	0.019(5)
200	2.621(20)	0.0085(3)	2.875(23)	0.027(4)	3.026(24)	0.020(4)
100	2.622(21)	0.0078(4)	2.860(24)	0.021(3)	3.027(25)	0.014(3)
70	2.626(25)	0.0073(4)	2.858(30)	0.023(3)	3.033(30)	0.012(2)
50	2.622(21)	0.0076(4)	2.848(24)	0.019(2)	3.027(24)	0.011(2)
30	2.618(17)	0.0075(4)	2.866(20)	0.018(2)	3.022(19)	0.011(2)