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Supplementary Material for

Interplays between Transition-Metal K-edge XMCD, slight structural distortion and magnetism in a series of trimetallic $Co_xNi_{(1-x)}Fe$ Prussian Blue Analogs.

Adama N'Diaye,[b] Amélie Bordage,[a] Lucie Nataf,[b] François Baudelet,[b] Eric Rivière[a] and Anne Bleuzen*[a]

[a] Institut de Chimie Moléculaire et des Matériaux d'Orsay, Université Paris-Saclay, CNRS, 91405 Orsay, France
[b] Synchrotron SOLEIL, L'Orme des Merisiers, St Aubin, BP 48, 91192 Gif sur Yvette, France.

Electronic Supplementary Information

S1. X-ray diffraction patterns, cell parameters and EDS analyses of **NiFe**, **$Ni_{75}Co_{25}Fe$**, **$Ni_{50}Co_{50}Fe$** and **$Ni_{25}Co_{75}Fe$**.

S2. Field Cooled (FC) and Zero Field Cooled (ZFC) magnetization curves for **NiFe**, **$Ni_{75}Co_{25}Fe$**, **$Ni_{50}Co_{50}Fe$** and **$Ni_{25}Co_{75}Fe$**.

S3. Temperature dependence of the inverse of the magnetic molar susceptibilities for **NiFe**, **$Ni_{75}Co_{25}Fe$**, **$Ni_{50}Co_{50}Fe$** and **$Ni_{25}Co_{75}Fe$**.

S4. Variation of the lattice constant value with the Co% along the **$Co_xNi_{(1-x)}Fe$** series.

S5. Variation of the Curie-Weiss temperature (θ) with x (the mole fraction of Co ions at the A site).

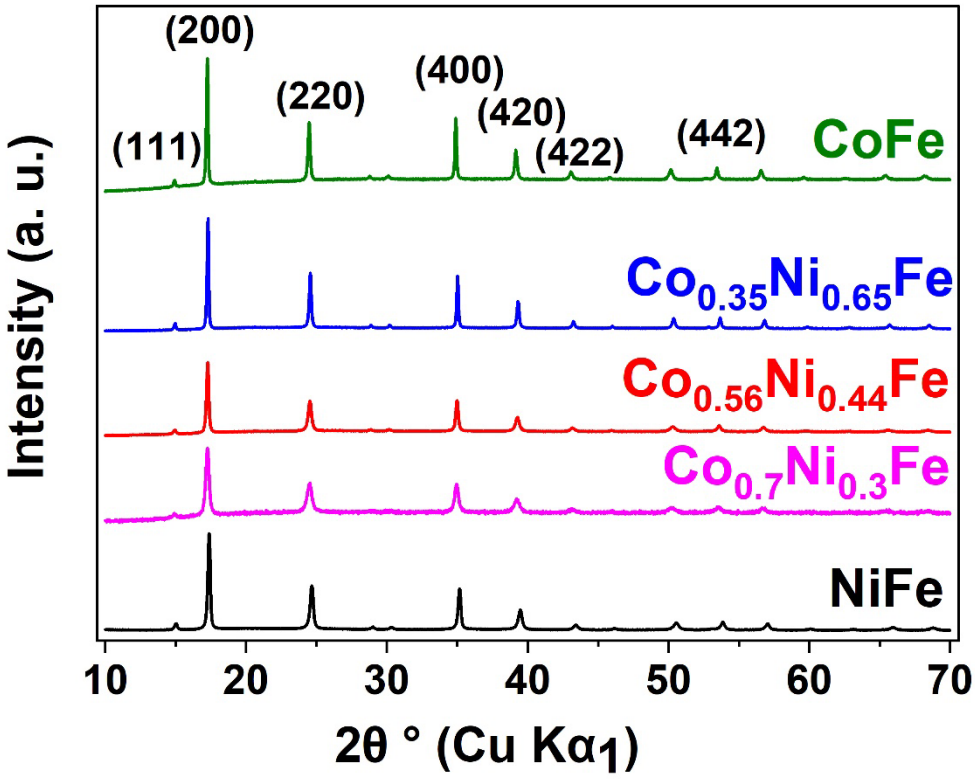
S6. X-ray diffraction pattern of **$NiCr_{0.50}Fe_{0.50}$**.

S7. Field Cooled (FC) and Zero Field Cooled (ZFC) magnetization curves for **$NiCr_{0.50}Fe_{0.50}$**.

S8. Magnetic field dependance of the magnetization at 4 K of **$NiCr_{0.50}Fe_{0.50}$**.

S1. X-ray diffraction patterns, cell parameters and EDS analyses of **CoFe**, **$Co_{0.7}Ni_{0.3}Fe$**, **$Co_{0.56}Ni_{0.44}Fe$**, **$Co_{0.35}Ni_{0.65}Fe$** and **NiFe**.

X-ray diffraction patterns were recorded using a Phillips PAN X-Pert PRO MPD X-ray diffractometer (Cu $K\alpha_1$; λ = 1.5418 Å). An aluminum sample holder was used.



All X-ray diffraction patterns exhibit the peaks of the well-known fcc structure of the PBAs. The cell parameters for the four PBAs are :

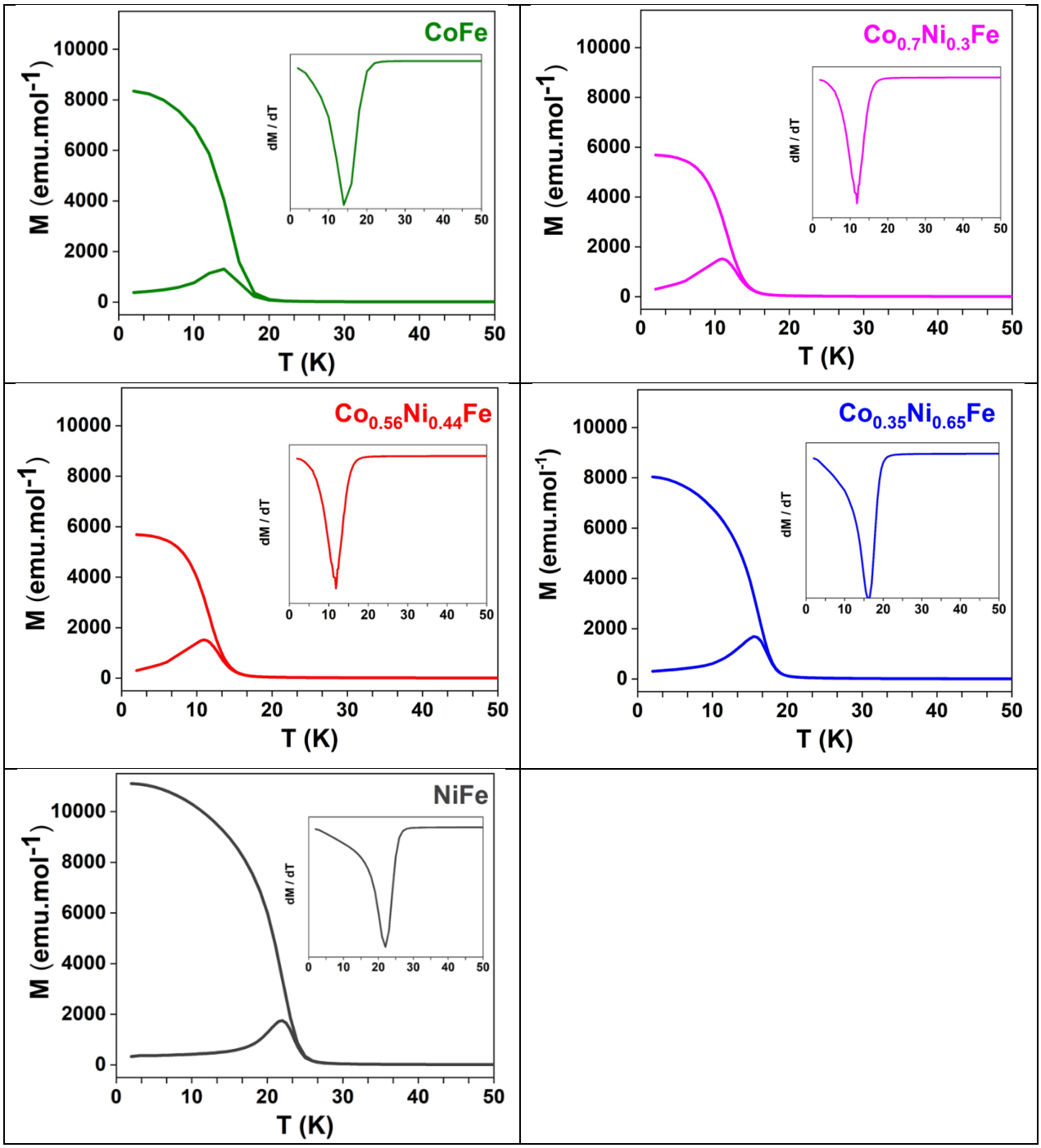
NiFe : a = 10.204±0.001 Å
$Co_{0.35}Ni_{0.65}Fe$: a = 10.25685±0.0004 Å
$Co_{0.56}Ni_{0.44}Fe$: a = 10.2607±0.0004 Å
$Co_{0.7}Ni_{0.3}Fe$: a = 10.2701±0.0004 Å
CoFe : a = 10.286±0.001 Å

The EDS analyses were used to determine the chemical formula of the trimetallic compounds. The results are given in the following table. The results for **NiFe** ($Ni_4[Fe(CN)_6]_{2.7}\bullet 17H_2O$) and **CoFe** ($Co_4[Fe(CN)_6]_{2.7}\bullet 17H_2O$) were given in ref. 37.

Sample	**$Co_{0.35}Ni_{0.65}Fe$**	**$Co_{0.56}Ni_{0.44}Fe$**	**$Co_{0.7}Ni_{0.3}Fe$**
Co:Ni:Fe Atomic%	21.67 : 39.57 : 38.76	33.53 : 26.88 : 39.59	39.31 : 14.47 : 46.22
Proposed chemical formula	$(Ni_{0.65}Co_{0.35})_4[Fe(CN)_6]_{2.5}\bullet 17H_2O$	$(Ni_{0.44}Co_{0.56})_4[Fe(CN)_6]_{2.6}\bullet 17H_2O$	$(Ni_{0.3}Co_{0.70})_4[Fe(CN)_6]_{2.73}\bullet 17H_2O$

S2. Field Cooled (FC) and Zero Field Cooled (ZFC) magnetization curves for **CoFe**, **$Co_{0.7}Ni_{0.3}Fe$**, **$Co_{0.56}Ni_{0.44}Fe$**, **$Co_{0.35}Ni_{0.65}Fe$** and **NiFe**.

The inset of each figure presents the derivative of the magnetization versus temperature, used for the determination of the magnetic ordering temperature $T_{C/N}$ (which is the minimum of each curve).

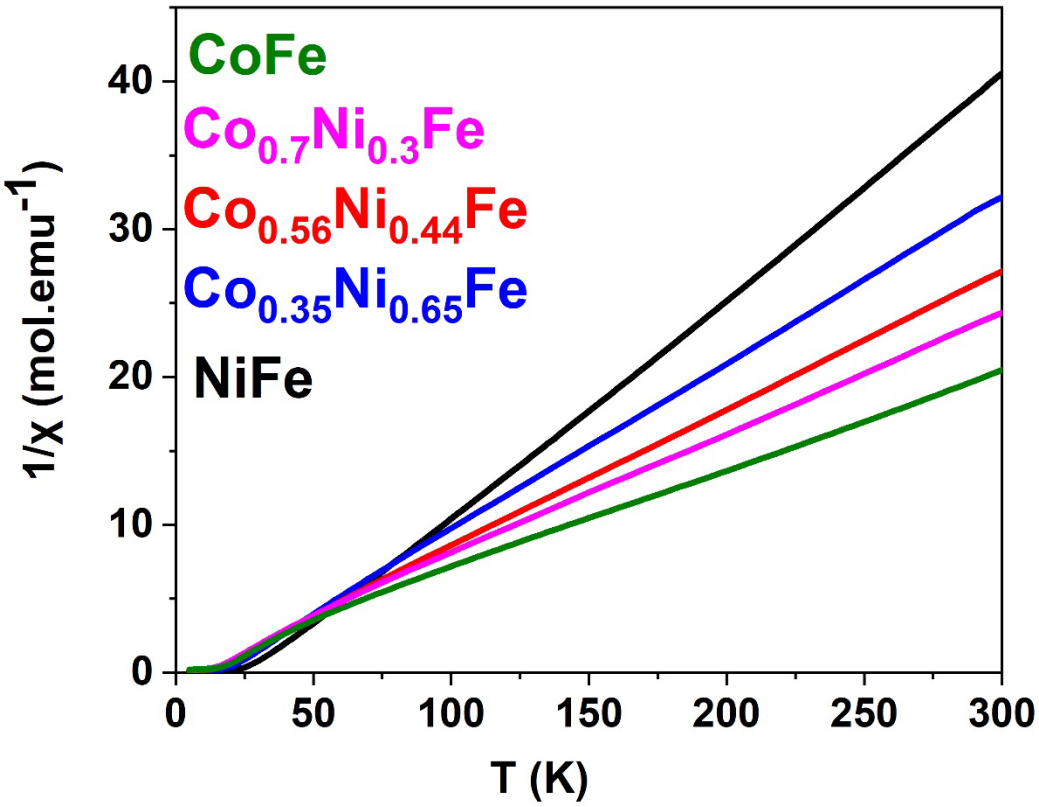


S3. Temperature dependence of the inverse of the magnetic molar susceptibilities for **CoFe**, **$Co_{0.7}Ni_{0.3}Fe$**, **$Co_{0.56}Ni_{0.44}Fe$**, **$Co_{0.35}Ni_{0.65}Fe$** and **NiFe**.

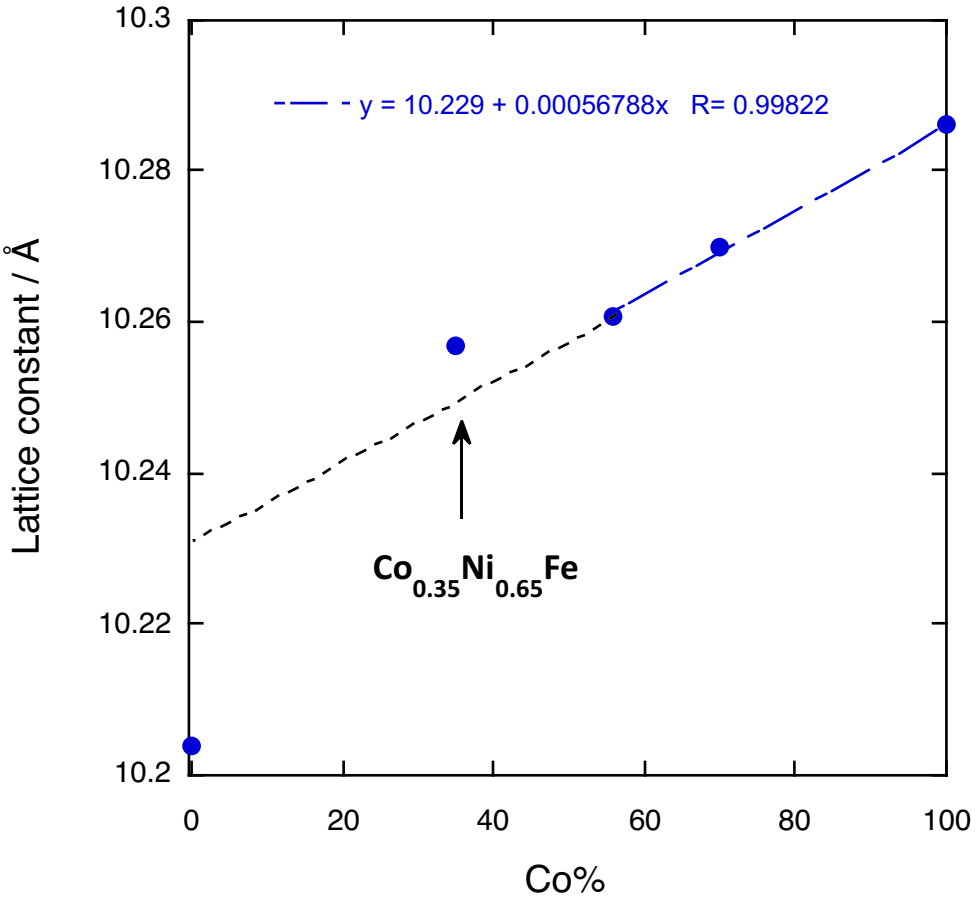
The curves were reproduced on the linear part in the appropriate high temperature range by a Curie-Weiss law :

$$\chi = \frac{C_{PBA}}{T - \theta}$$

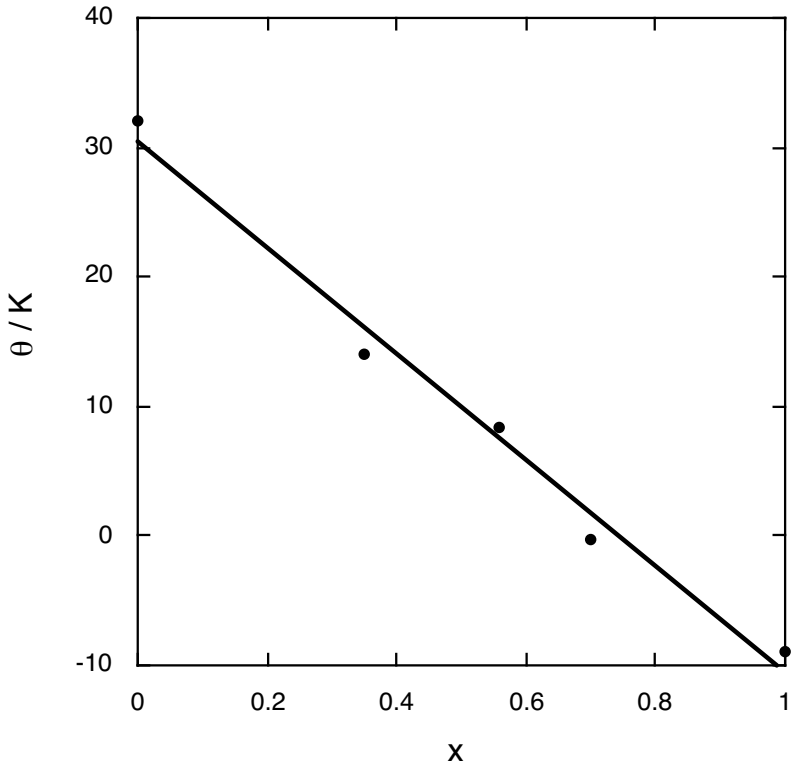
where χ is the molar magnetic susceptibility, C_{PBA} the Curie constant, T the temperature and θ the Weiss constant.



S4. Variation of the lattice constant value with the Co% along the **$Co_xNi_{(1-x)}Fe$** series.



S5. Variation of the Curie-Weiss temperature (θ) with x (the mole fraction of Co ions at the A site).



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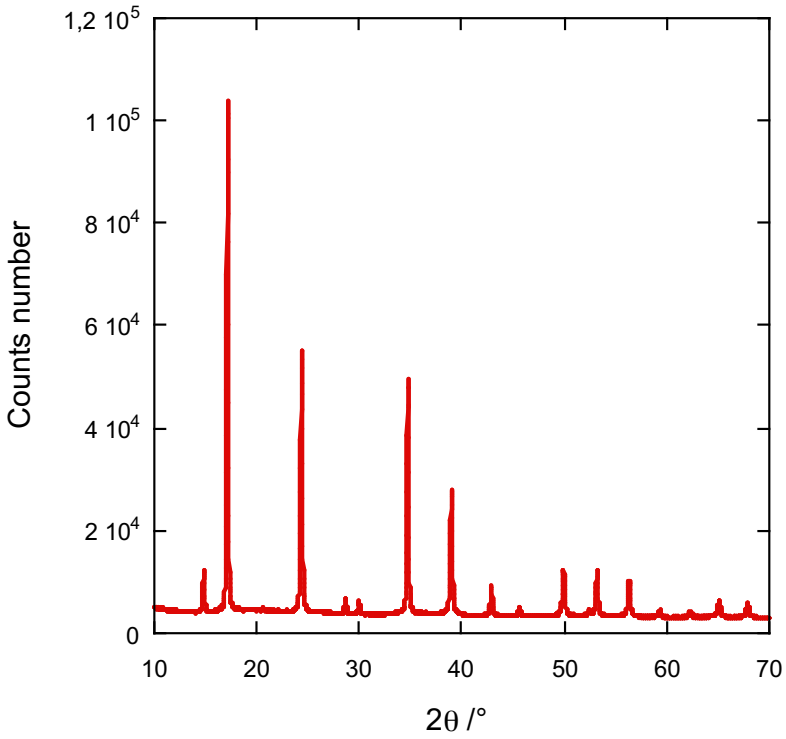
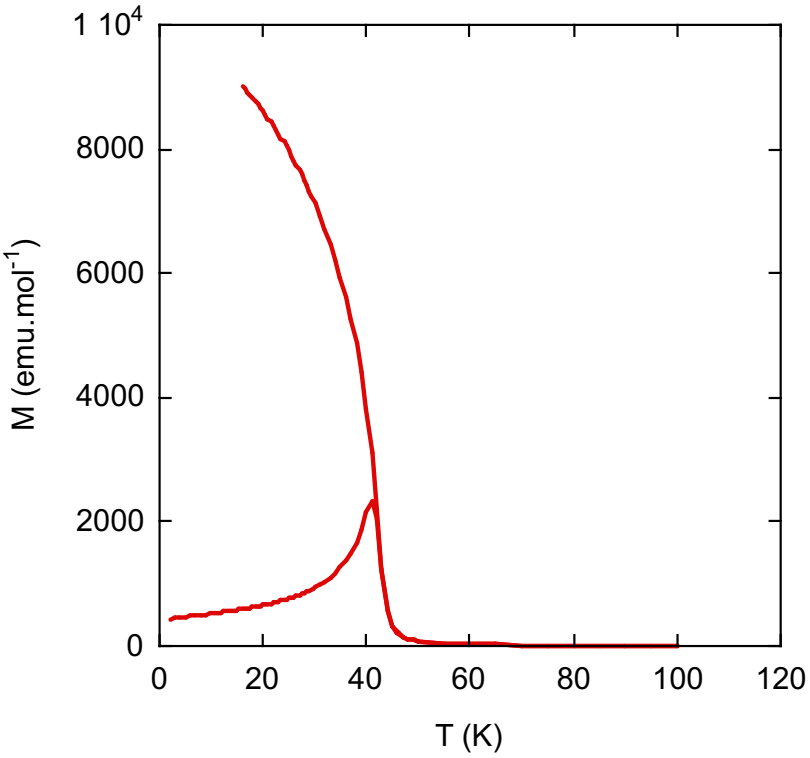


S7. Field Cooled (FC) and Zero Field Cooled (ZFC) magnetization curves for **$NiCr_{0.50}Fe_{0.50}$**.



S8. Magnetic field dependance of the magnetization at 4 K of **$NiCr_{0.50}Fe_{0.50}$**.