

## Supplementary Material for

# Interplay between transition metal K-edge XMCD and magnetism in Prussian blue analogs.

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### **Electronic Supplementary Information**

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S8. Powder X-ray diffraction of CuFe PBAs containing an increasing amount of Cs<sup>+</sup> cations inserted in interstitial sites.

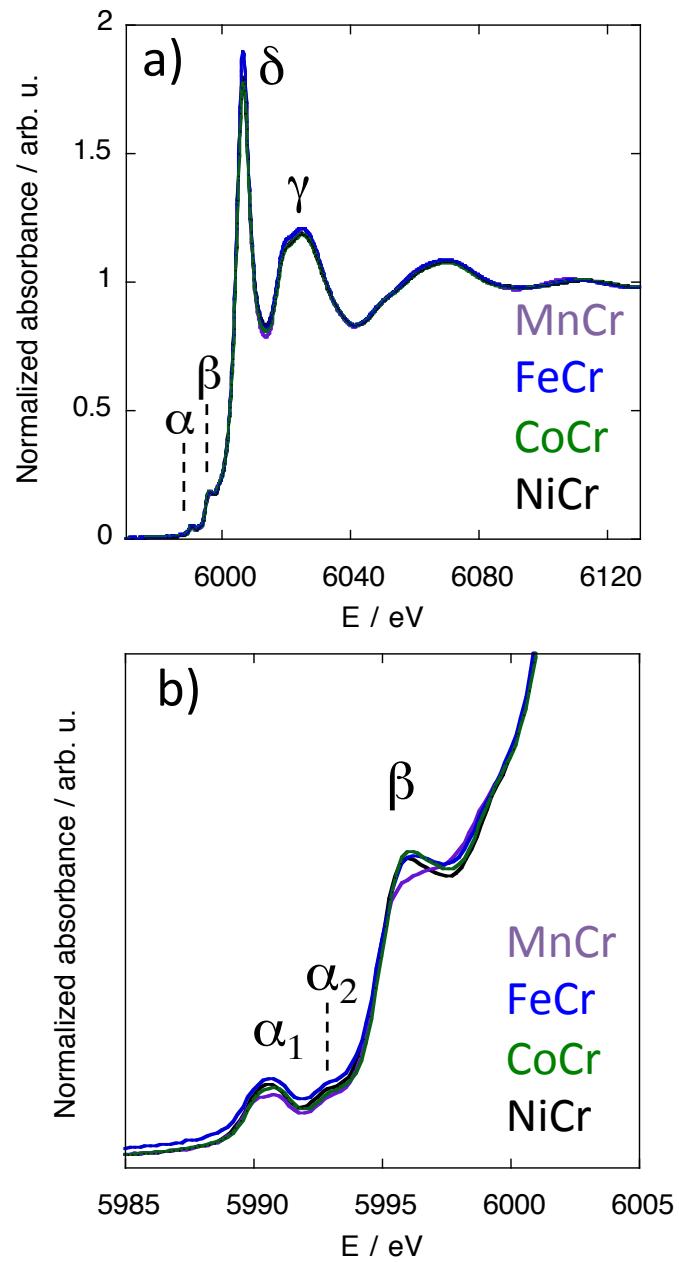
S9. Ni K-edge XANES spectra of **NiFe** and **NiCr**.

Table S1. Normalized absolute value of the intensity of the pre-edge XMCD feature at the A and B K-edges of **MnFe**, **CoFe**, **NiFe**, **CuFe**, **MnCr**, **FeCr**, **CoCr** and **NiCr**.

Sample	A K-edge		B K-edge	
	I pre-edge	I pre-edge / I edge %	I pre-edge $a_1/a_2$ (Fe)	I pre-edge / I edge %
<b>MnFe</b>	0.0001	2.5	0.0013/0	35/0
<b>CoFe</b>	0.0008	7	0.0004/0	20/0
<b>NiFe</b>	0.0005	9	0.003/0	35/0
<b>CuFe</b>	-*	-*	0.002/0	25/0
<b>MnCr</b>	0.00045	4	0	0
<b>FeCr</b>	0.0005	2.5	0	0
<b>CoCr</b>	0.0001	8	0	0
<b>NiCr</b>	0.00085	15	0	0

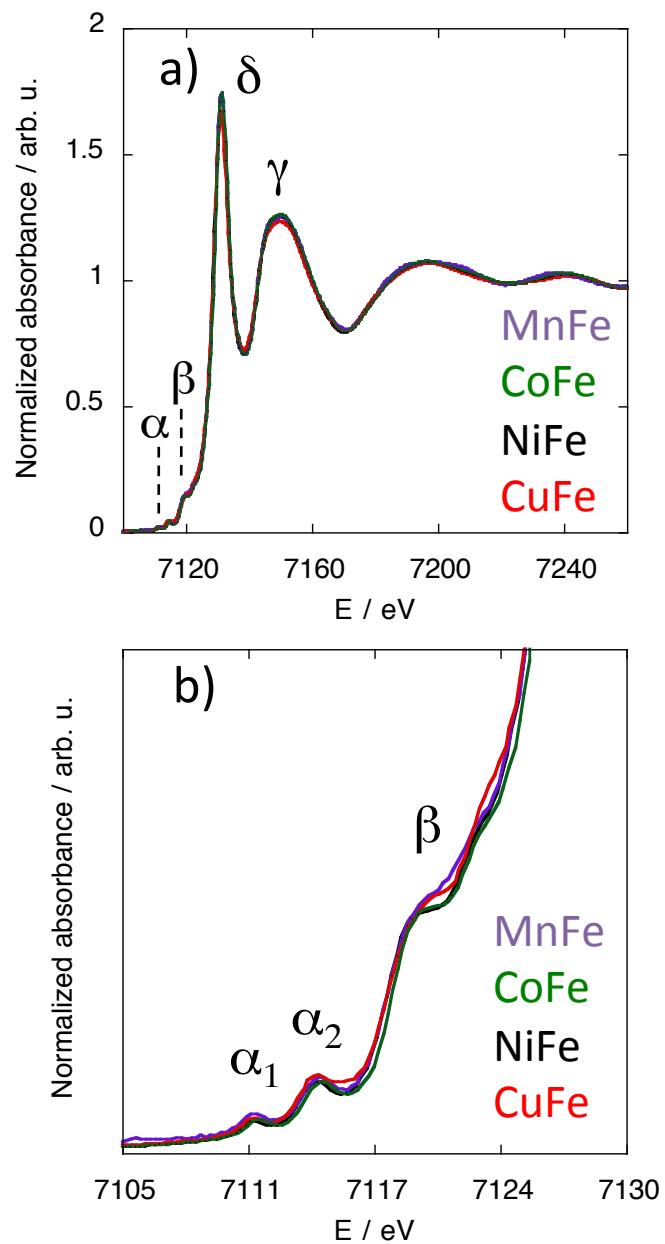
S1. Cr K-edge XANES spectra of the ACr series.

The XANES spectra are displayed in figure a) and figure b) presents an enlargement of the spectra over the pre-edge region.

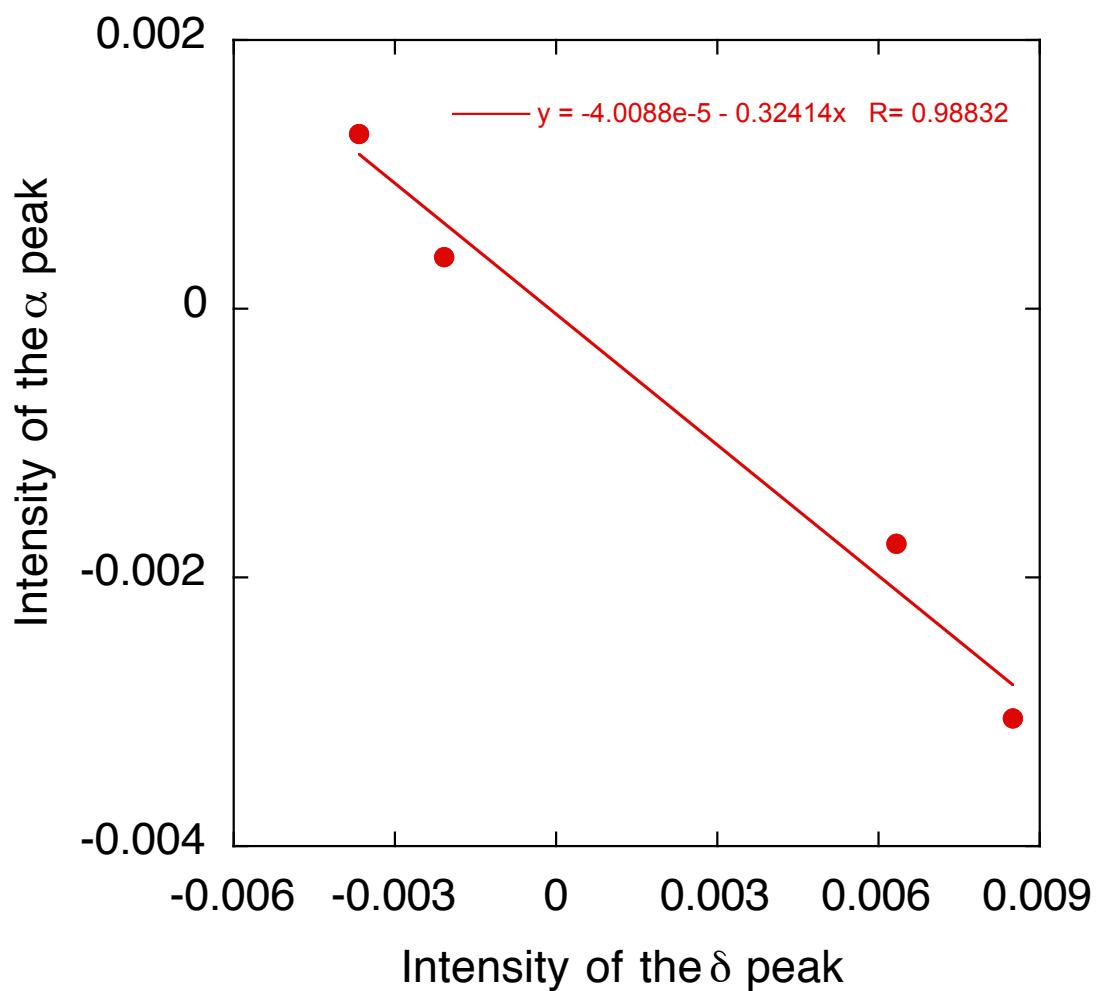


## S2. Fe K-edge XANES spectra of the AFe series.

The XANES spectra are displayed in figure a) and figure b) presents an enlargement of the spectra over the pre-edge region.

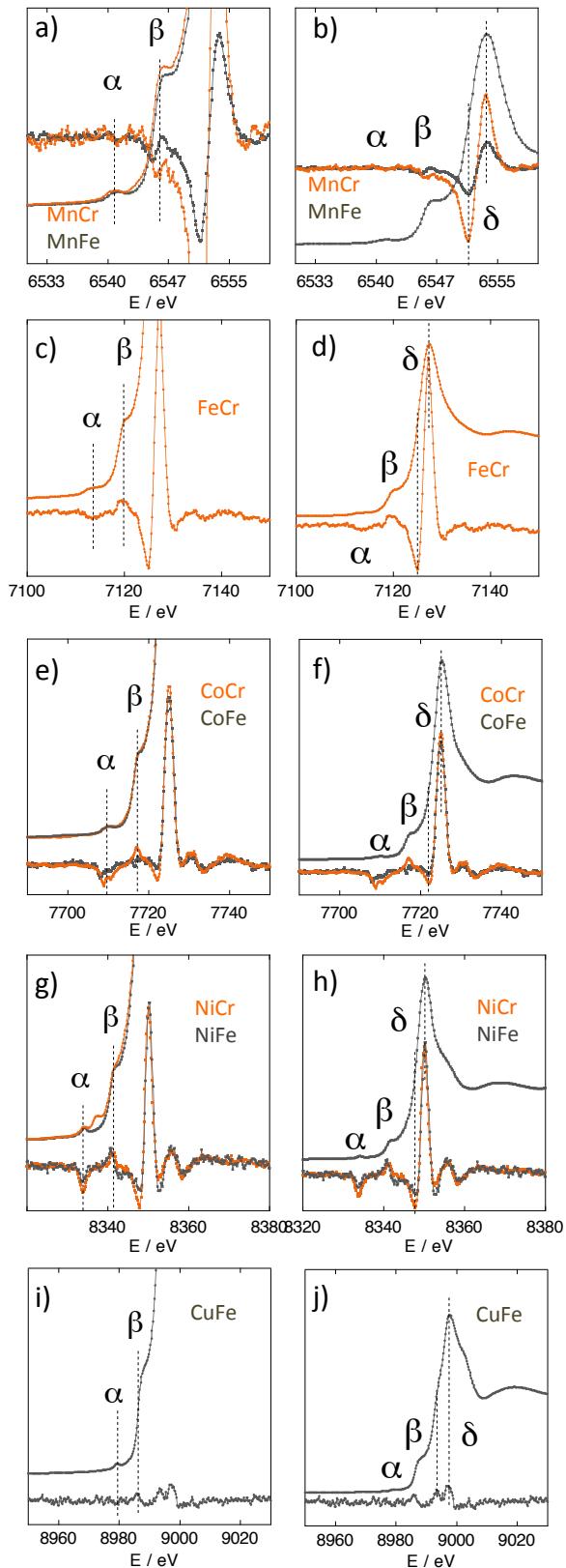


S3. Intensities of the Fe K-edge XMCD  $\alpha$  pea as a function of the  $\delta$  peak.

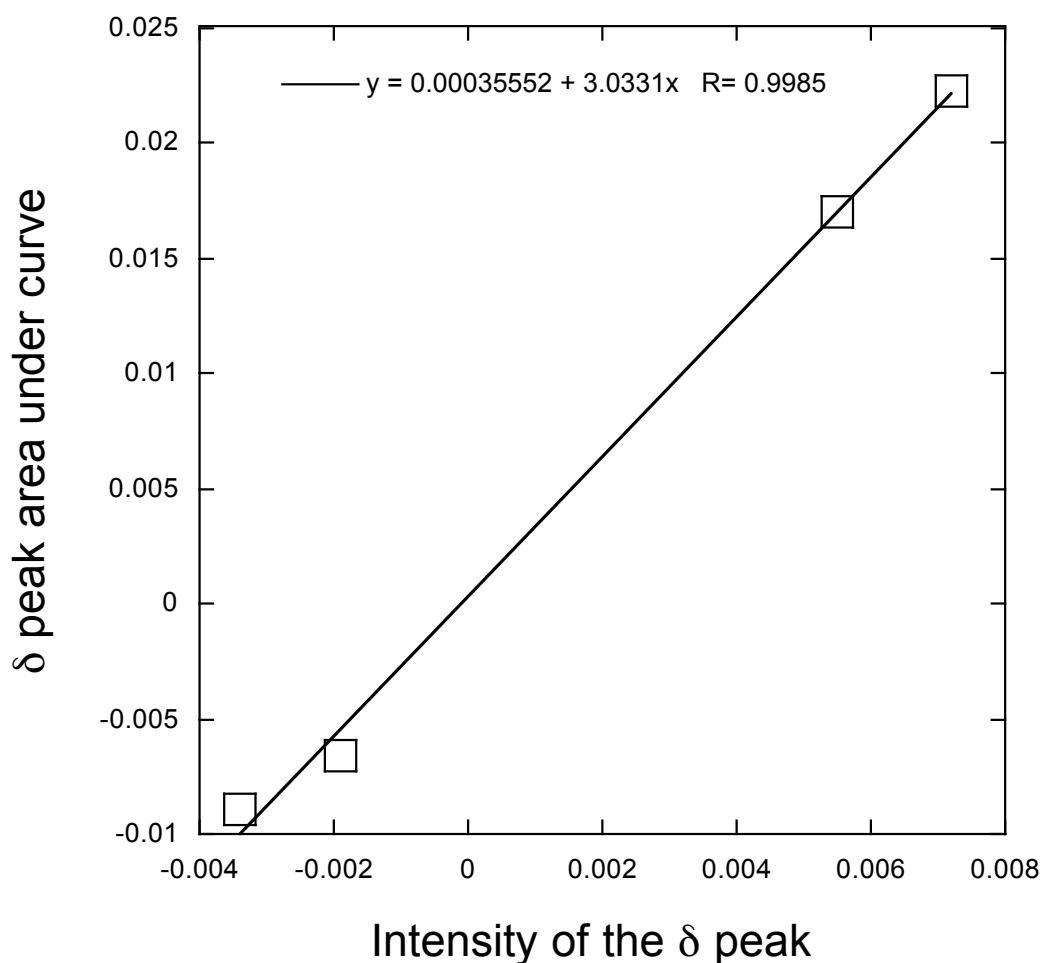


S4. A K-edge XMCD signal compared to the adapted enlargement of the XANES spectrum over the pre-edge/rising edge and edge regions.

a, b) **MnCr** and **MnFe** at the Mn K-edge ; c, d) **FeCr** at the Fe K-edge ; e, f) **CoCr** and **CoFe** at the Co K-edge ; g, h) **NiCr** and **NiFe** at the Ni K-edge ; i, j) **CuFe** at the Cu K-edge.

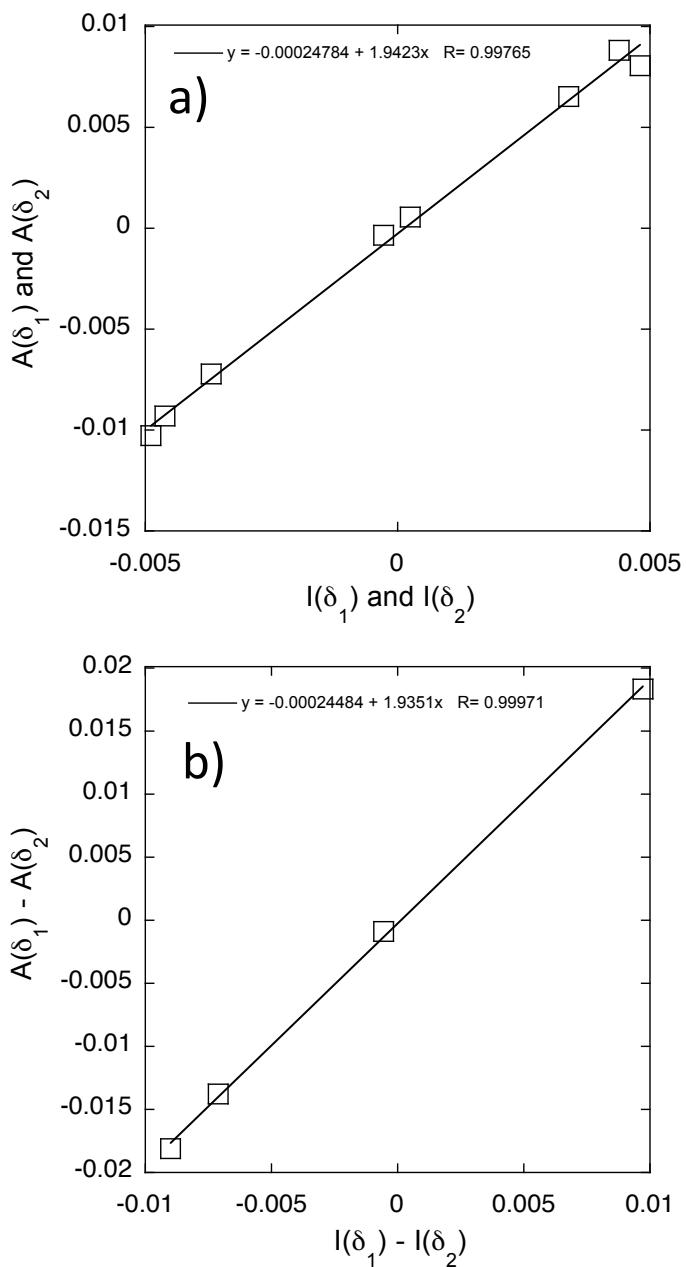


S5. Area under curve for the main  $\delta$  peak of the XMCD signal at the Fe K-edge as a function of its intensity for the **AFe** series.



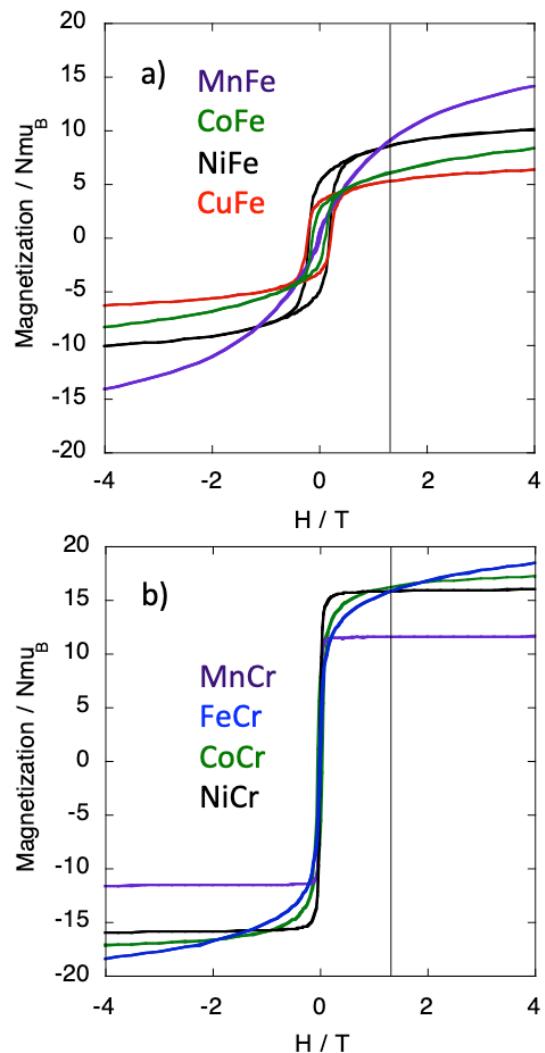
S6. Area under curve for the main  $\delta$  contribution to the XMCD signal at the Cr K-edge as a function of its intensity for the **ACr** series.

- a) Area under curve for each lobe ( $\delta_1$  and  $\delta_2$ ) of the XMCD signal as a function of its intensity.
- b) Difference between the area under curve of the  $\delta_2$  and  $\delta_1$  lobes as a function of the difference between their intensity.



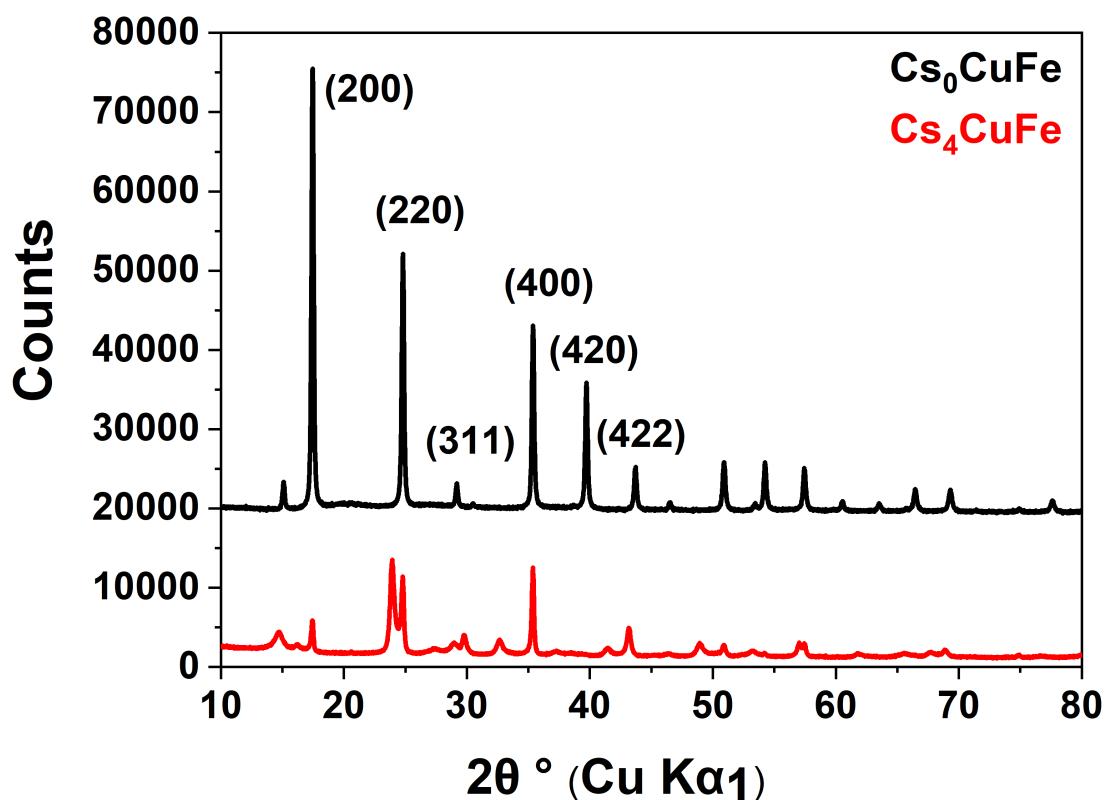
S7. Magnetic field dependence of the magnetization at 4K for a) the **AFe** series and b) the **ACr** series.

The vertical line indicates the magnetic field value used for XMCD measurements (1.3 T).



S8. Powder X-ray diffraction of CuFe PBAs containing 0 (**CuFe**, black line) and 4 ( $\text{Cs}_4\text{CuFe}$ , red line)  $\text{Cs}^+$  cations inserted in interstitial sites.

The X-ray diffraction pattern of  $\text{Cs}_4\text{CuFe}$  is characteristic of the tetragonal structure, revealing a cooperative Jahn-Teller distortion with all elongated axes of the  $\text{Cu}^{2+}$  complexes along the same direction, while **CuFe** exhibits the well-known fcc structure of PBAs.



### S9. Ni K-edge XANES spectra of **NiFe** and **NiCr**.

The XANES spectra are presented in figure a and figure b displays an enlargement of the spectra over the pre-edge region.

