

Al-driven peculiarities of local coordination and magnetic properties in single-phase $\text{Al}_x\text{-CrFeCoNi}$ high-entropy alloys

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Section S1. Powder diffraction patterns

The phase purity of studied fcc- and bcc-HEA samples was confirmed by PXRD patterns recorded at RT with X-rays of $\lambda = 1.54 \text{ \AA}$ and $\lambda = 0.50 \text{ \AA}$ wavelengths, respectively. The relative intensities of the peaks and corresponding crystallographic indexes are presented in Fig. S1. The (1 0 0) superstructure peak is characteristic to the B2 structure. There are a lot of debates in the literature what would be the most accurate notation of the formed crystallographic structure: B2 (ordered bcc), A2 (disordered bcc) or a mixture of B2+A2 phases in the $\text{Al}_x\text{-CrFeCoNi}$ alloys depending on the stoichiometry and external conditions. According to results of our studies we tend to think that the most accurate description of the crystallographic ordering in the studied $\text{Al}_3\text{-CrFeCoNi}$ alloy is a partially ordered BCC structure.

Section S2. EXAFS data recorded at the Al K edge

EXAFS spectra recorded at the Al K absorption edge with linearly polarized X-rays at RT for both HEA samples (Fig. S2) exhibit a well pronounced Si K edge white line; thus, it is not possible to extract EXAFS oscillations up to $12 \text{ \AA}^{-1}$ with the desired accuracy. Spectra are not normalized and illustrate a different amount of Al absorbers in the sub-surface volume of Al-deficient (fcc-HEA) and Al-rich (bcc-HEA) samples.

Section S3. Partial distribution functions of aluminum atoms

Partial distribution functions (PDFs) of Al atoms obtained with reasonable accuracy from structural models retrieved by RMC simulations even without fitting the experimental EXAFS spectra taken at the Al K absorption edge are shown in Fig. S3. Despite increased noise level, these PDFs can be effectively used for the calculation of Al-Al nearest-neighbours. The individual element PDFs might be interpreted as pointing to preferential occupation of the first coordination shell around Al absorbers by Co and Ni atoms while Cr and Fe occupy the sites in the second coordination shell; as well as that the farer shells are occupied more homogeneously and, hence, confirming the presence of only partial ordering in the existing bcc structure. However, such an interpretation should be used with a great care.

Section S4. Temperature dependence of inverse magnetisation and a magnetization derivative

The temperature dependence of the inverse magnetisation ($1/M$) for the fcc- and bcc-HEAs is presented in Fig. S4, where clear deviations from the linear behaviour are visible. Considering the results of the element-specific XANES presented in the main manuscript, the non-linearity of the temperature dependence could partially be explained by the presence of surface oxidation that results in a mixture of several magnetic phases that influence the magnetometry data. The inset shows the derivative $\partial M/\partial T$ (T) of the fcc-HEA sample with a clear minimum at around $T \sim 50 \text{ K}$.

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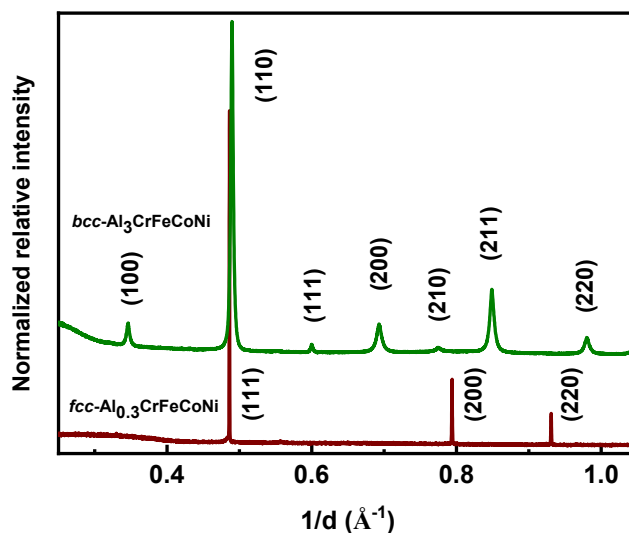


Figure S1 PXRD patterns recorded for the fcc- and bcc- HEAs under the study at RT with $\lambda = 1.54 \text{ \AA}$ and $\lambda = 0.50 \text{ \AA}$, respectively. The parameter $1/d$ takes into account the different wavelengths used ($1/d = 2\sin\theta/\lambda$). The diffraction patterns point to a phase purity for both samples.

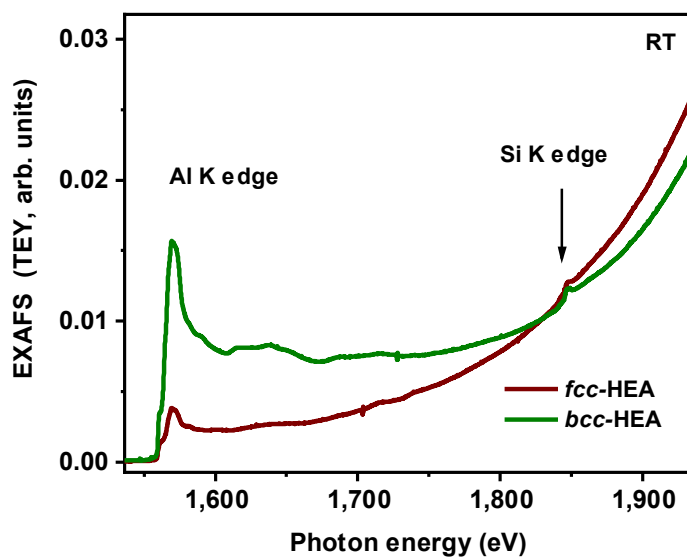


Figure S2 Raw EXAFS spectra recorded at the Al K absorption edge from the fcc- and bcc-HEA samples at RT with horizontally polarized X-rays by TEY. The presence of the Si K absorption edge is visible.

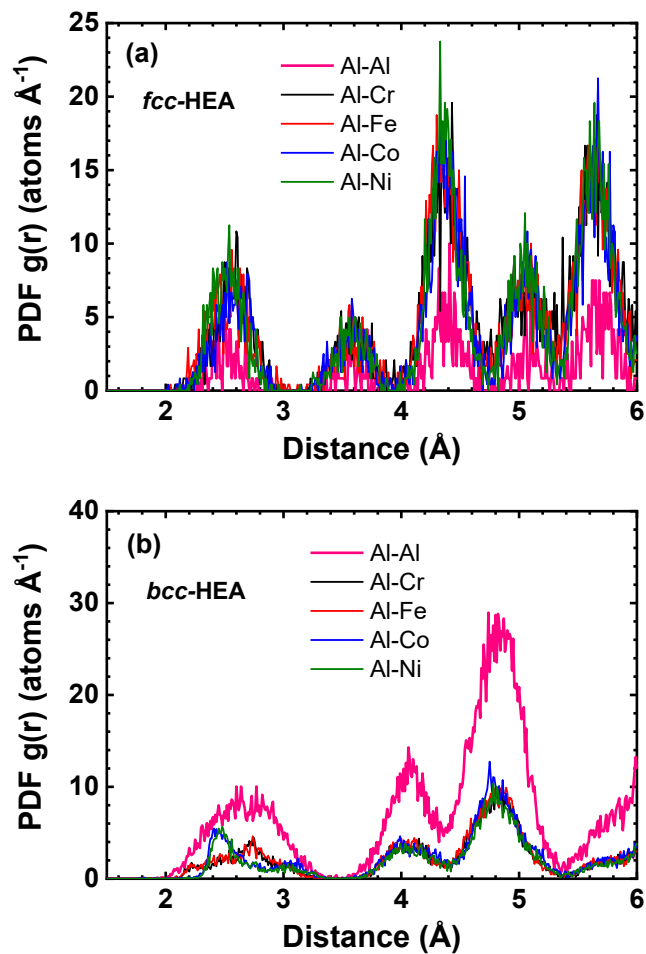


Figure S3 Pair distribution functions $g(r)$ related to Al atoms in the fcc- (a) and bcc- (b) HEAs extracted from K-edge EXAFS spectra of Cr, Fe, Co and Ni using the RMC method.

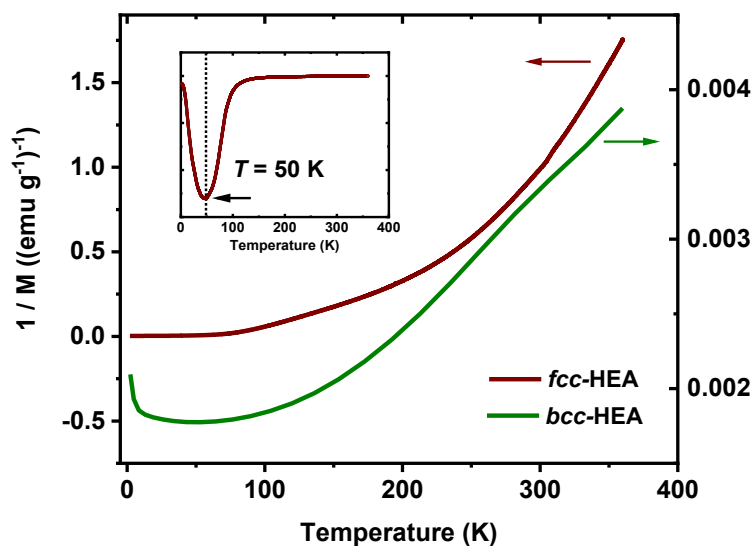


Figure S4 The temperature dependence of inverse magnetization ($1/M$) for both HEA samples (with oxidized surfaces); the inset shows the derivative $\partial M / \partial T(T)$ of the fcc-HEA sample (smoothed using a Loess function).