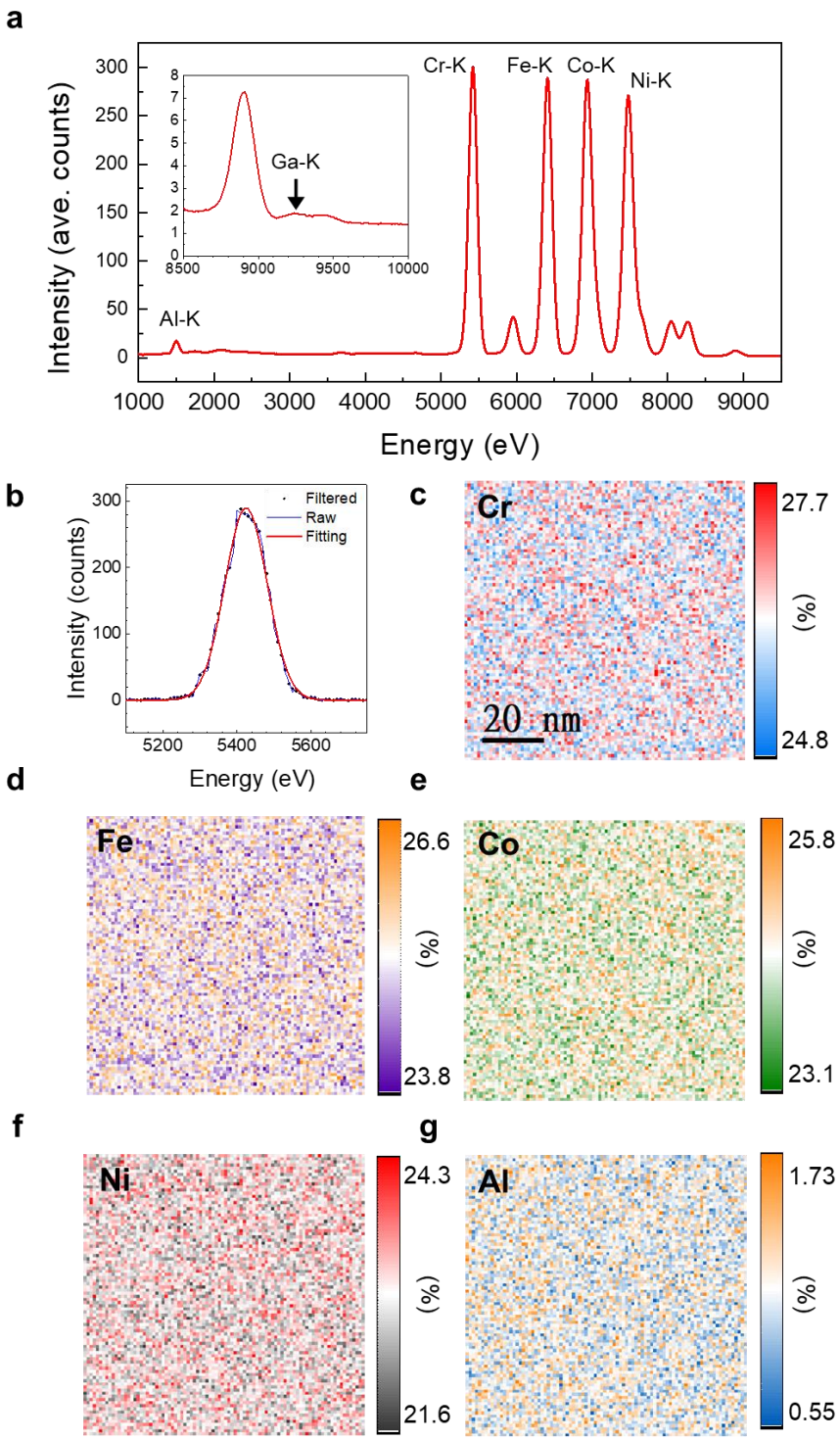
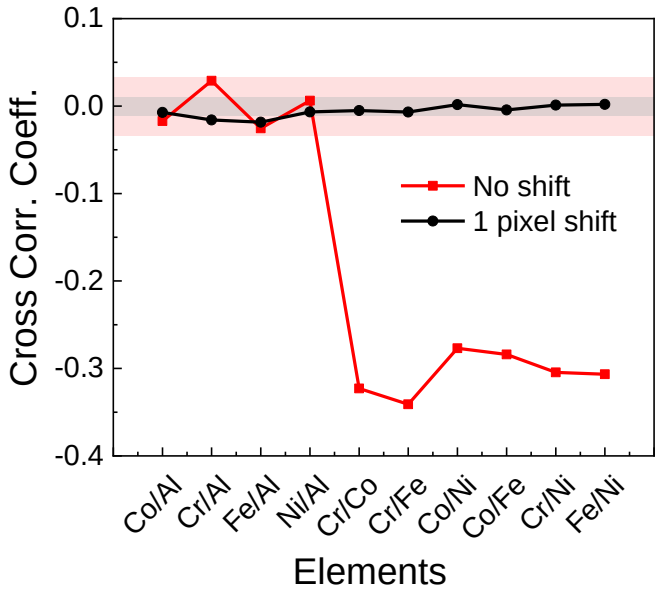
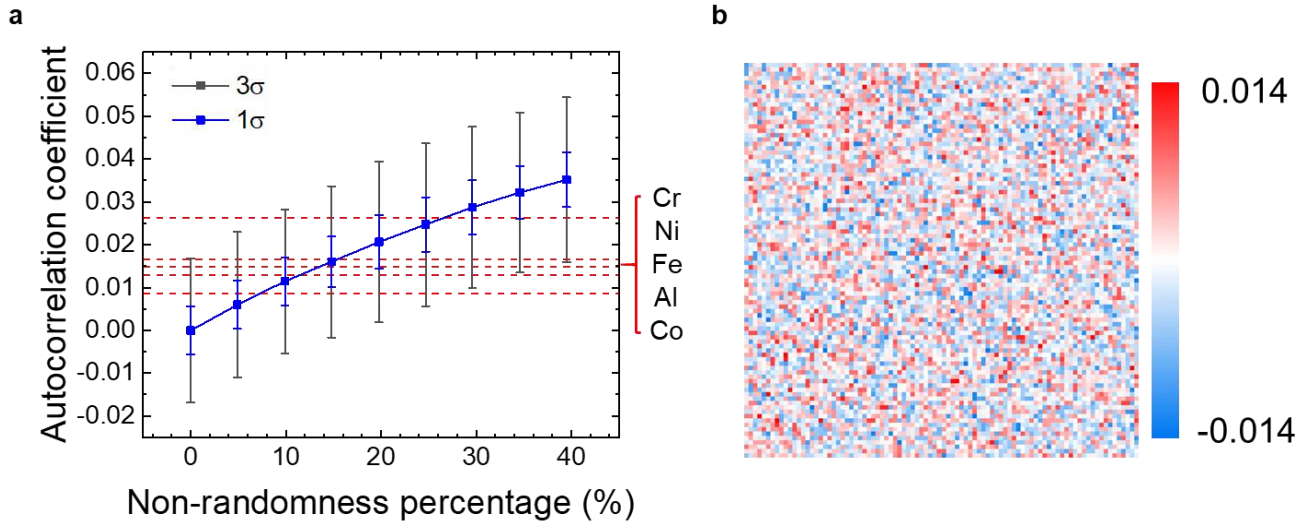


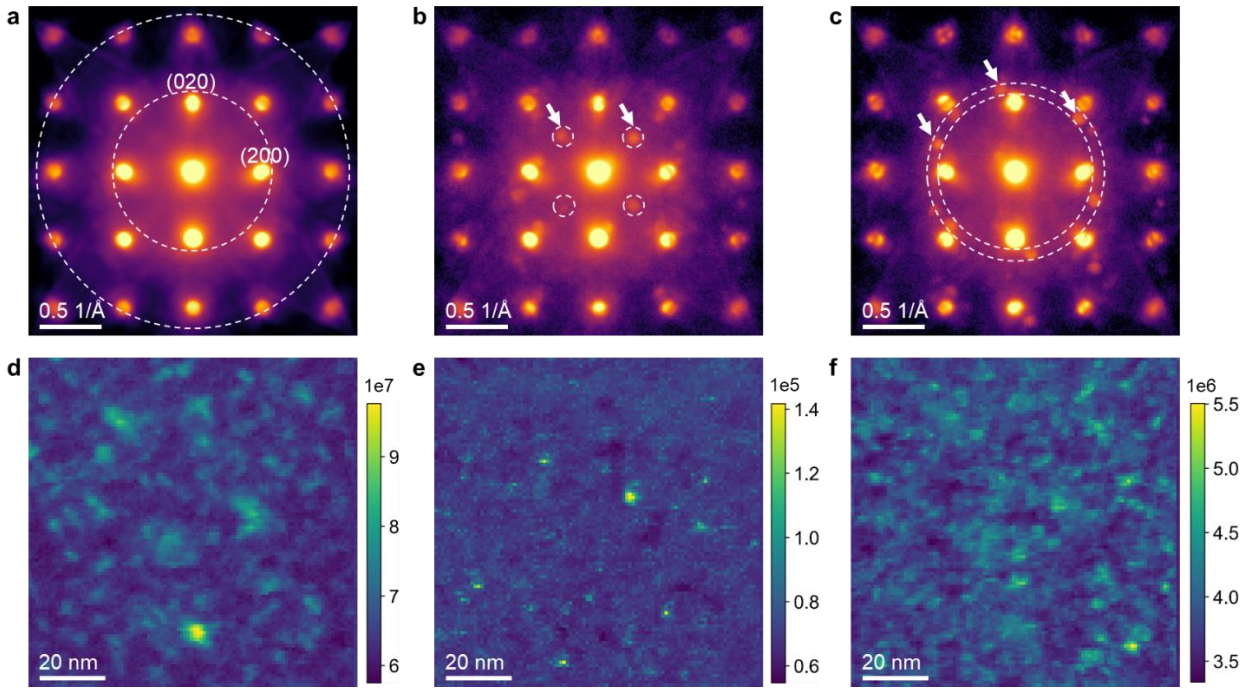
Extended Data Fig. 1 Chemical disorder in $Al_{0.1}CrCoFeNi$. *(a) An averaged EDS spectrum from the STEM/EDS spectral dataset acquired over the 90x90 nm^2 sample area with a step size of 1 nm. (b) An example of Cr-K peak fitting. (c-g) The peak-ratio maps extracted from the EDS dataset for each element in the HEA. The maps are displayed with the low and upper limits of ±3 times of standard deviation about the average.*



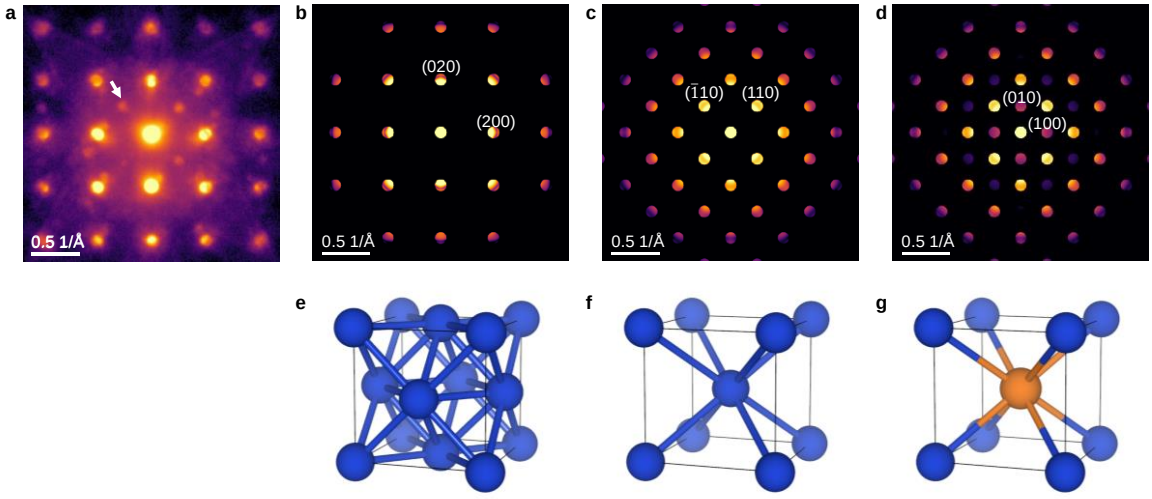
Extended Data Fig. 2 Cross-correlation of elemental combinations. *For zero-pixel image shift, the red curve shows that Cr, Co, Fe and Ni have a negative correlation with each other, while the correlation of Al with other elements is under the limit of random model (red shadow region). At 1-pixel distance apart, the cross-correlation intensity falls within the limit of random model (gray shadow region) for all elements.*



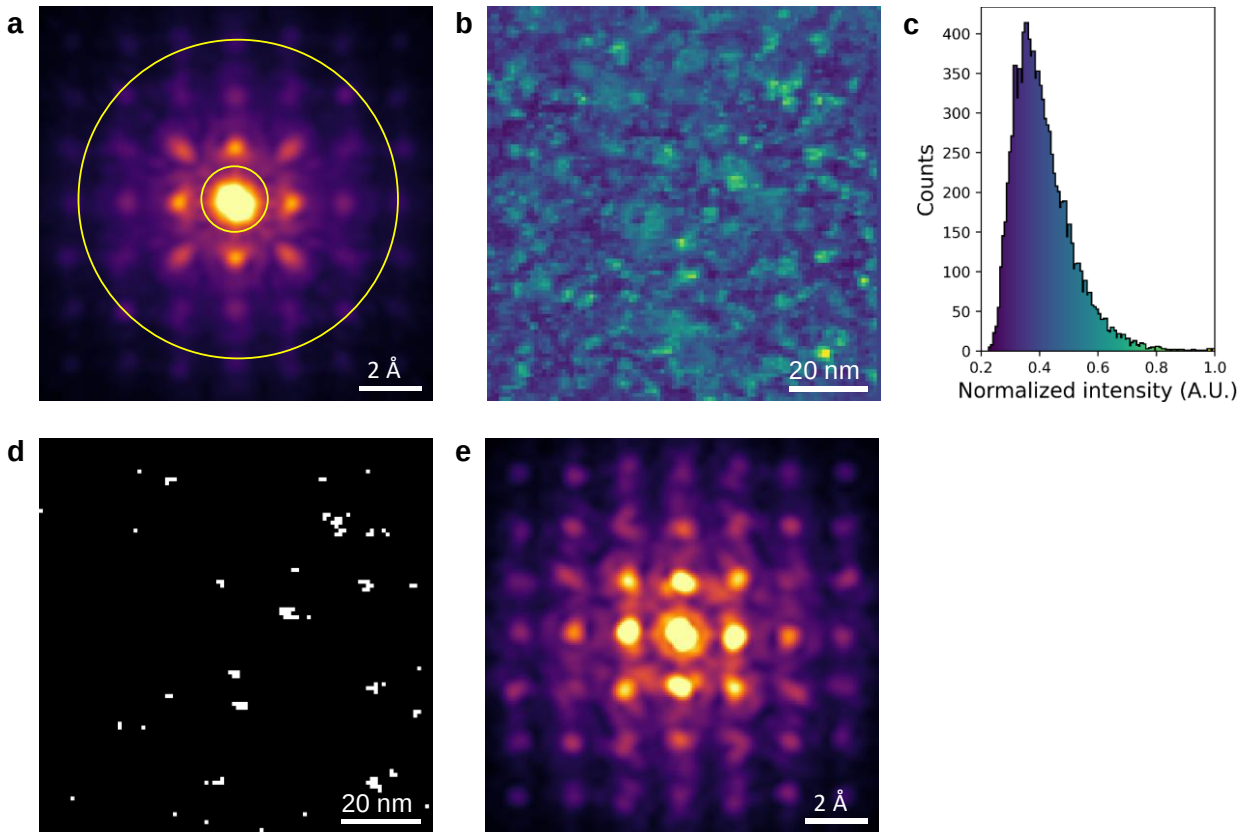
Extended Data Fig. 3 Effect of non-randomness on autocorrelation function (ACF). *(**a**) The plot of ACF coefficient at r = 1 versus the percentage of non-randomness as obtained from simulations. Red dash lines indicate the ACF value of each element at 1 nm distance obtained from the STEM-EDS ratio maps. The bar in (a) marks $\pm\sigma$ and $\pm 3\sigma$, estimated from the random model (Suppl. Note 2). (**b**) An example random image with 25% non-randomness sites.*



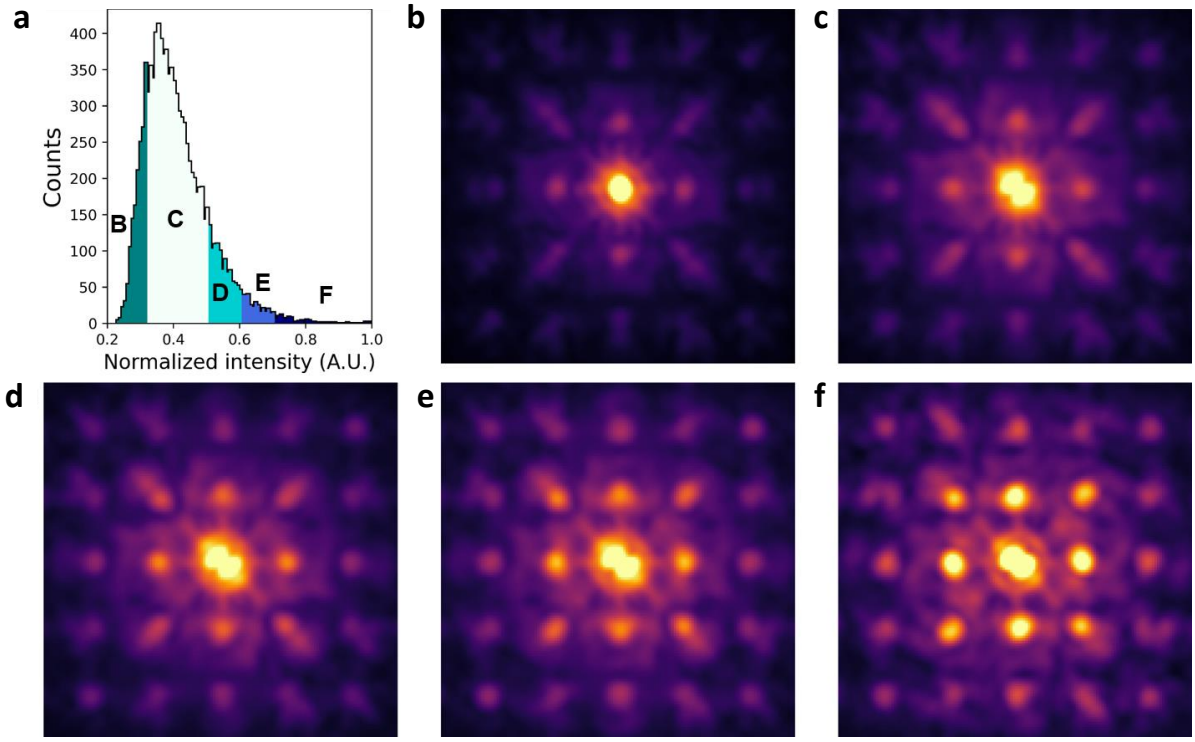
Extended Data Fig. 4 Electron nanodiffraction imaging of structural inhomogeneity*. (**a**) Averaged diffraction pattern of an 100x100 SEND dataset, with step size of 1 nm. Selected individual diffraction patterns shown in logarithmic scale having (**b**) extra <110> reflections, and (**c**) extra reflections inside the dashed circles. (**d**) virtual ADF image constructed from the SEND dataset by integrating intensities between the dashed circles in (**a**), (**e**) Virtual DF image generated by integrating intensities inside the 4 dashed circles in (**b**). (**f**) Virtual ADF image integrated over the dashed circles in (**c**). The white arrows in (b & c) indicate extra reflections which do not belong to the FCC lattice.*



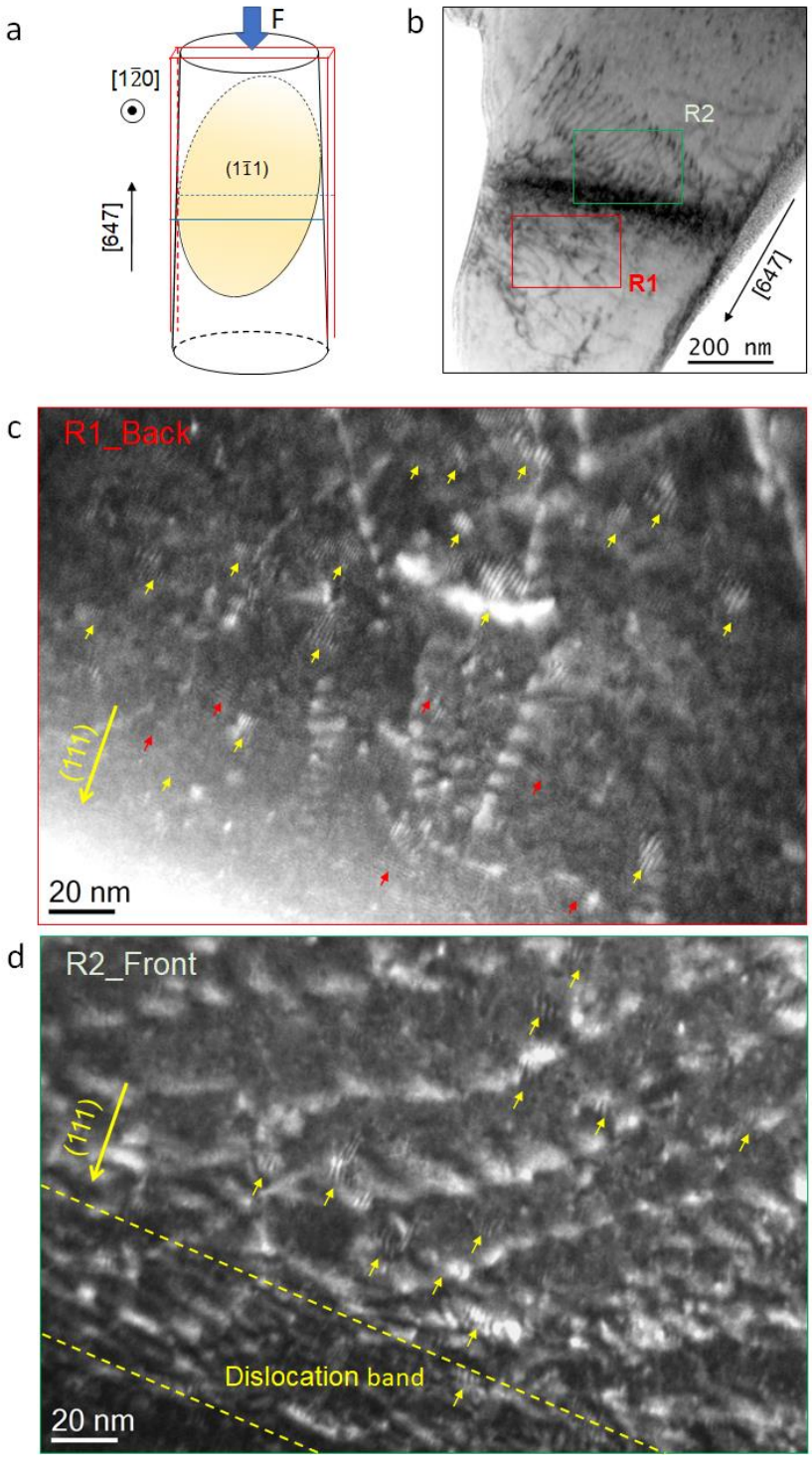
Extended Data Fig. 5 Electron diffraction phase analysis. *(**a**) A recorded nanodiffraction pattern showing the extra <110> reflections. Corresponding simulated nanodiffraction patterns of (**b**) FCC structure, (**b**) BCC structure, and (**d**) B2 structure with Cr at the body center, with the Ball and stick structure models shown below in (**e-g**).*



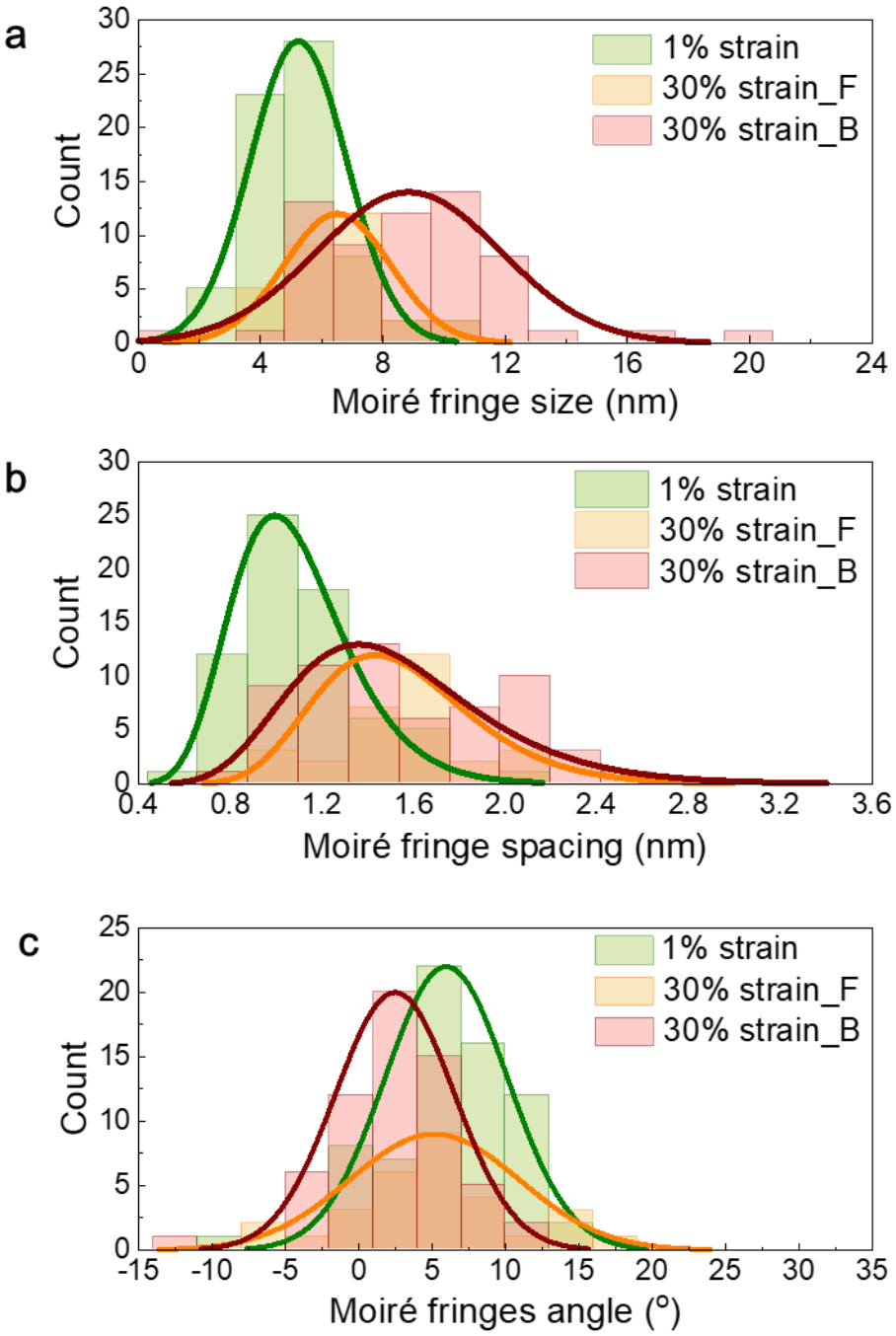
Extended Data Fig. 6 Histogram segmentation of the cepstral dataset*. (**a**) Average of 10,000 difference cepstral patterns. (**c**) Histogram of the (**b**) Cepstral virtual ADF image reconstructed from the cepstral-transformed SEND dataset by integrating the correlation distances from r=0.44~4.25 Å. (**d**) A masked image of **(b)** with values larger than 0.7 of the normalized intensity, and (**e**) Cepstral pattern obtained by averaging over the threshold regions in **(d)**.*



Extended Data Fig. 7 Difference Cepstral patterns based on histogram segmentation. *(**a**) Histogram of virtual ADF image reconstructed from the cepstral-transformed SEND dataset by integrating the correlation distances from r=0.44~4.25 Å. (**b-f**) Representative cepstral patterns for each segment indicated in the histogram in (**a**), obtained by averaging over the corresponding clustered regions.*



Extended Data Fig. 8 Dislocation microstructure of a deformed HEA nanopillar. ***(a)*** *Schematic illustration of nanopillar compression and cross-sectional geometry for post-mortem analysis.* ***(b)*** *A BF-TEM image showing the dislocation band and dislocation pileup in front of the dislocation band formed after compression to 30%.* ***(c)*** *and* ***(d)*** *DF-TEM images showing Moiré Fringes contrast regions with size ranging from ~6 to ~14 nm and dislocations in the deformed pillars in the front and back of the dislocation band. The red and yellow arrows mark two types of Moiré Fringes.*



Extended Data Fig. 9 Distribution of Moiré fringes in the deformed nanopillars. *At 30% strain, B and F stands for in the back and front of the dislocation band respectively, as shown in* ***Extended Data Fig. 8****. (**a**) the Moiré fringe size, (**b**) Moiré fringe spacing and (**c**) Angle between Moiré fringes and **g**(111) direction*