STEM-EELS investigations and Lorentz microscopy unravelling the structural evolution of FeCoNi(AlMn)$_x$ high-entropy alloy and its impact on magnetic properties

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High-entropy alloys (HEAs) are a new class of materials exhibiting a superior combination of magnetic, mechanical and electrical properties. FeNiCoAlMn is a promising candidate for a soft magnet, important for applications in electrical systems such as power generation, electromagnets and transmission. However, the high entropy effect is partially true, since other thermodynamic properties can also play a role in phase stability, making the alloys vulnerable to phase separation that could affect their magnetic behavior.

Here, we report on the first nanoscale investigation of FeNiCo(AlMn)$_x$ processed by Laser Metal Deposition, by applying (Scanning) Transmission Electron Microscopy ((S)TEM) and Lorentz TEM combined with Energy-Dispersive X-ray spectroscopy (EDX) and Electron Energy-Loss Spectroscopy (EELS). Experiments were performed on an FEI Titan G2 60-300 kV equipped with a CEOS DCOR probe-corrector, monochromator and Super-X EDX detectors. Quantum mechanical modelling using density functional theory (DFT) was also used to assess the relative stability of different structural models of selected compositions, giving support to and adding detail to the experimental studies. The impact of phase transitions on the magnetic properties was investigated with a vibrating sample magnetometer (VSM) and Lorentz TEM.

A gradual change of the FCC phase towards BCC by increasing x was observed. A thorough analysis employing STEM-EELS revealed a more complicated structural picture on the nanoscale. A coexistence of Full-Heusler $L2_1$ and B2 ordered structures was detected in the BCC phase, and ordered nano-percipitates and grains were detected in the FCC phase. Low-loss EELS revealed a plasmon-peak splitting in FCC indicative of two valence electron densities. This finding in combination with intense Moiré fringes and high-defect density indicate that FCC exhibits intense spinodal decomposition trends. However, ordering and phase separation trends did not have a severe impact on the magnetic behavior of the alloy. All samples exhibited good soft-magnetic behavior since they were easily magnetized to the saturated state with coercivity values of <1000 A/m. Ordering in the BCC phase did not have a negative effect on the soft magnetic properties, indicating that the magnetic behavior of B2 or Heusler alloys with similar compositions to HEAs, is more associated with the specific chemical environment of atoms. Direct visualization of pinning of magnetic domain walls at the ordered nano-precipitates and grains was indicated by Lorentz TEM. However, their impact on the coercivity was trivial in comparison to the impact of the grain size and sub-grain structure of the alloys in μm-scale.

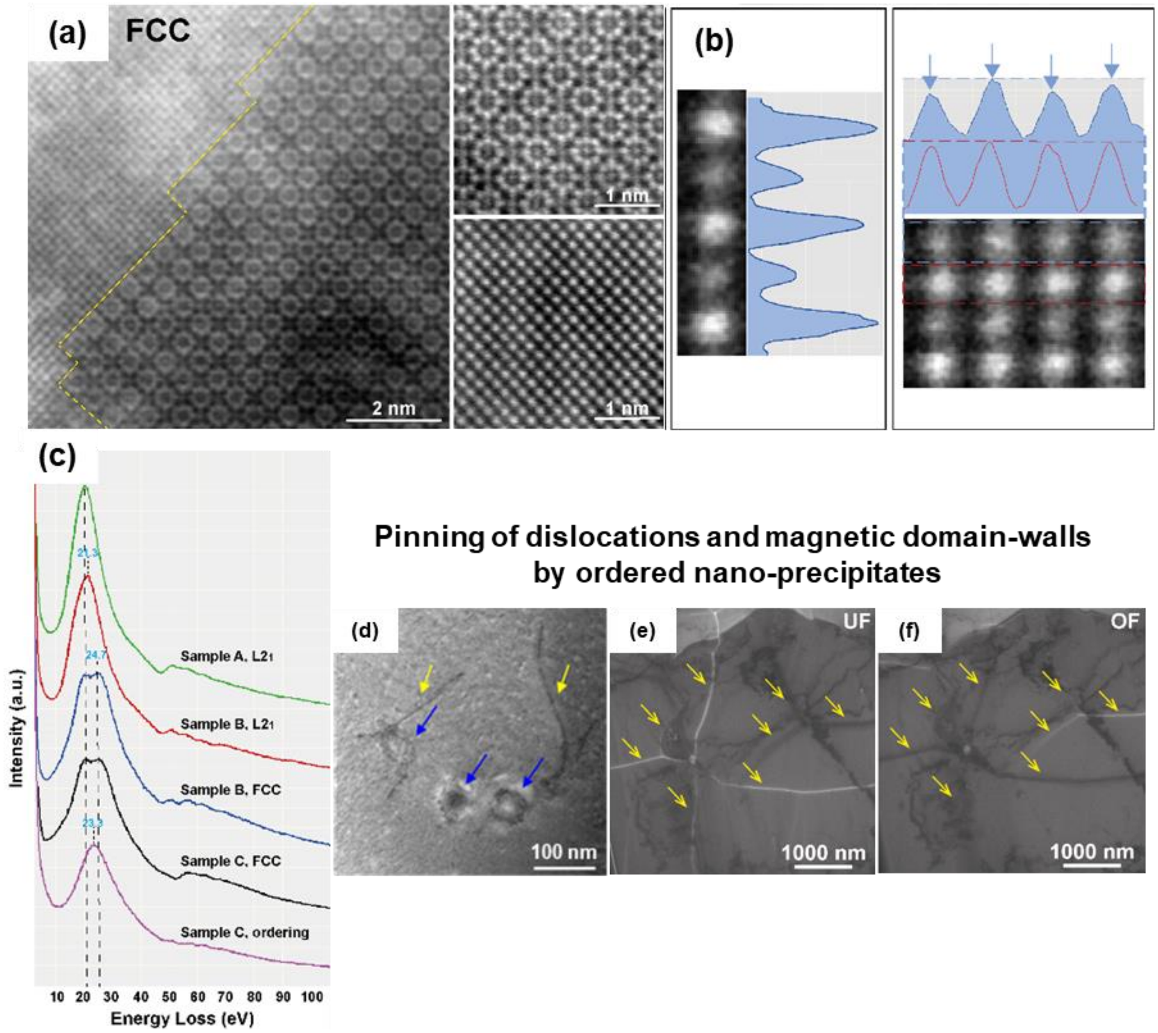

Figure 1. (a) ADF-STEM image showing phase transition of FCC to an ordered phase. A coherent relationship between the FCC and ordering is revealed, with the phase-transition taking place in nano-steps. Close-ups of the FCC along the [001] direction and the ordered area. (b) Close-ups of high-resolution HAADF-STEM image along the [101] direction of BCC with intensity profiles, showing ordering along the (020) and (202) planes. Nanoscale interplay between disordered BCC and ordered B2/$L2_1$ phases was revealed. (c) Comparison of low-loss EELS spectra. A clear plasmon-peak splitting is observed only in the FCC phases indicating two valence electron densities - consistent with the observed spinodal decomposition. The phenomenon is suppressed in all ordered phases. (d) low-magnification TEM image and (e)-(f) Lorentz TEM Fresnel images showing that ordered nano-precipitates act as pinning points for the dislocation and magnetic domain wall movement.

Acknowledgments

This work is part of the Project "MAGNIFICENT - Additively manufactured magnetic high entropy alloys for renewable electricity", funded by the Research Council of Norway (pr. nr 287979) within the NANO2021 Program.