

Strain-Driven Bidirectional Spin Orientation Control in Epitaxial High Entropy Oxide Films

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Abstract : High entropy oxides (HEOs), based on the incorporation of multiple-principal cations into the crystal lattice, offer the possibility to explore previously inaccessible oxide compositions and unconventional properties. Here it is demonstrated that despite the chemical complexity of HEOs external stimuli, such as epitaxial strain, can selectively stabilize certain magneto-electronic states. Epitaxial $(\text{Co}_{0.2}\text{Cr}_{0.2}\text{Fe}_{0.2}\text{Mn}_{0.2}\text{Ni}_{0.2})_3\text{O}_4$ -HEO thin films are grown in three different strain states: tensile, compressive, and relaxed. A unique coexistence of rocksalt and spinel-HEO phases, which are fully coherent with no detectable chemical segregation, is revealed by transmission electron microscopy. This dual-phase coexistence appears as a universal phenomenon in $(\text{Co}_{0.2}\text{Cr}_{0.2}\text{Fe}_{0.2}\text{Mn}_{0.2}\text{Ni}_{0.2})_3\text{O}_4$ epitaxial films. Prominent changes in the magnetic anisotropy and domain structure highlight the strain-induced bidirectional control of magnetic properties in HEOs. When the films are relaxed, their magnetization behavior is isotropic, similar to that of bulk materials. However, under tensile strain, the hardness of the out-of-plane (OOP) axis increases significantly. On the other hand, compressive straining results in an easy OOP magnetization and a maze-like magnetic domain structure, indicating perpendicular magnetic anisotropy. Generally, this study emphasizes the adaptability of the high entropy design strategy, which, when combined with coherent strain engineering, opens additional prospects for fine-tuning properties in oxides.

Keywords : high entropy oxides, thin film, strain tuning, perpendicular magnetic anisotropy

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