Structure and magnetism of non-equimolar compositionally complex oxides

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Compositionally complex oxides (CCOs) are a class of ceramic materials that incorporate five or more metallic ions into their crystal lattice [1], offering structural stability alongside wide compositional flexibility. Most studies on spinel CCOs have focused on equimolar transition-metal compositions [2-4], while the structure and properties of stoichiometrically non-equimolar systems remain largely unexplored. In this work, we investigate a series of non-equimolar spinel CCOs containing the same magnetic principal elements as in previously reported systems, while systematically varying the stoichiometry of each constituent element (Figure 1).

To investigate this compositional space, samples were synthesized using a sol–gel method that has been shown to produce highly homogeneous CCOs [5]. Structural characterization by powder X-ray diffraction, neutron diffraction, and high-resolution transmission electron microscopy confirmed that all studied CCOs form highly crystalline, single-phase materials. Additionally, SQUID magnetometry, temperature-dependent neutron diffraction, and electrical transport measurements revealed highly tunable ferrimagnetic transition temperatures and electrical resistivity. These results demonstrate that non-equimolar compositions unlock a wider tunability of magnetic and electrical properties, opening new avenues for energy-related applications.

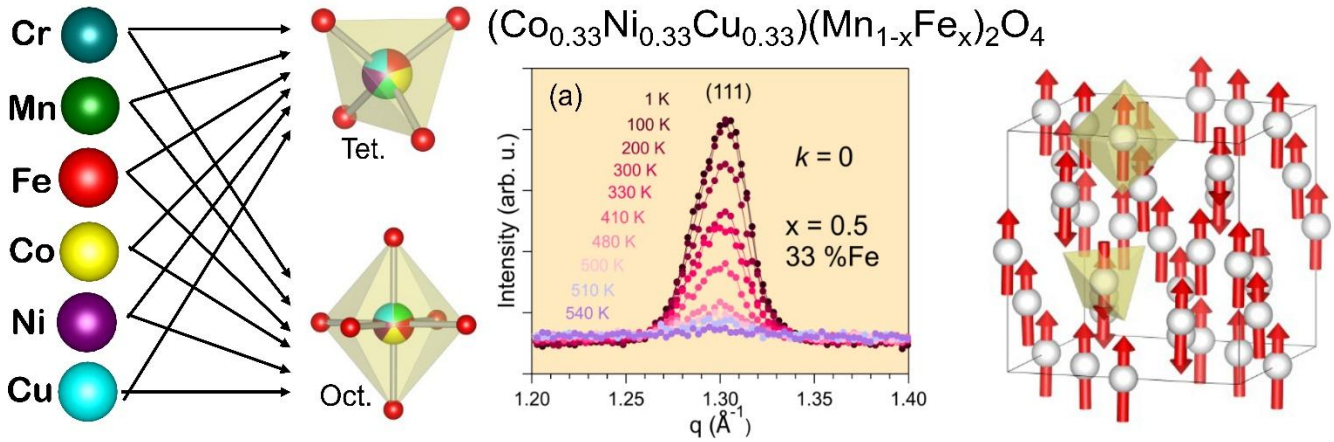


Figure 1. *In spinel compositionally complex oxides (CCOs), the ions will be stabilized in either tetrahedral or octahedral sites depending on their crystal field stabilization energy and the influence of the configurational entropy. The studied system was* $(Co_{0.33}Ni_{0.33}Cu_{0.33})(Mn_{1-x}Fe_x)_2O_4$ *with x = 0.25,0.5, and 0.75. (a) Intensity of (111) Bragg reflection from neutron powder diffraction at temperatures above* T_C *= 510K and down to 2 K, showing onset of k=0 ferrimagnetic ordering for x = 0.25. The obtained magnetic structure corresponds to the ferrimagnetic* $I4_1/am'd'$.

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