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Magnetostructural coupling in spinel oxides[1]

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Spinels oxides are of great interest functionally as multiferroic, battery, and magnetic materials as well as fundamentally because they exhibit novel spin, structural, and orbital ground states. Competing interactions are at the heart of novel functional behavior in spinels. Here, we explore the intricate landscape of spin, lattice, and orbital interactions in magnetic spinels by employing variable-temperature high-resolution synchrotron x-ray powder diffraction, total neutron scattering, magnetic susceptibility, dielectric, and heat capacity measurements. We show that the onset of long-range magnetic interactions often gives rise to lattice distortions. Our work illustrates that the spinels $NiCr_2O_4$, $CuCr_2O_4$, and Mn_3O_4, which are tetragonal at room temperature due to Jahn-Teller ordering, undergo further spin-driven structural distortions at the onset of long-range ferrimagnetic order. We have also studied the complete structural description of the ground states of several spinels including the geometrically frustrated spinels $ZnCr_2O_4$ and $MgCr_2O_4$. The detailed spin-lattice studies of spinel oxides presented here illustrate the prevalence of structural phase coexistence when magnetostructural changes occur below 50 K. The new understanding of structural ground states in spinel oxides will guide the design of structure-property relationships in these materials. Broadly, this work highlights the importance of variable-temperature high-resolution synchrotron x-ray diffraction in understanding phase transitions in functional materials.

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