Rare earth Ionic size Driven Evolution of Magnetic Structure in A2CoMnO6 (A = Lu, Ho) geometrically frustrated double perovskites

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Double perovskites with the general formula $A_2B'B''O_6$ have attracted significant attention due to their diverse functional properties and potential applications in spintronics, magnetoelectrics, and energy-related technologies. Derived from the simple perovskite ABO_3 structure, these compounds exhibit remarkable chemical and physical versatility, including insulating to metallic electronic behavior, multiple magnetic ground states, and magnetically frustrated phases arising from the inherent structural and compositional flexibility of the perovskite framework.

Rare-earth-based double perovskites are of particular interest in spin-frustrated multiferroics, where ferroelectricity emerges from complex magnetic ordering rather than non-centrosymmetric crystal symmetry. Substitution at the A-site and/or B-site provides an effective route to tailor physical properties by modifying exchange pathways, inducing lattice distortions, and tuning magnetic interactions. Such controlled substitutions can generate competing magnetic interactions, leading to exotic magnetic states such as frustration-driven ferromagnetism and multiferroicity.

In this work, we report the synthesis and structural and magnetic properties of A_2CoMnO_6 (A = Lu, Ho) double perovskites prepared via the conventional ceramic route. Powder X-ray diffraction and neutron diffraction measurements confirm phase-pure samples crystallizing in a monoclinic structure with space group $P2_1/n$. Low-temperature X-ray diffraction reveals structural stability over the temperature range 12–300 K. Rietveld refinement indicates the presence of antisite disorder between Co and Mn ions, introducing geometric frustration that strongly influences the magnetic behavior.

Magnetization measurements reveal dominant ferromagnetic interactions mediated by Co^{2+}–O–Mn^{4+} superexchange pathways. A significant enhancement of the Curie temperature is observed upon substitution of Ho^{3+} for Lu^{3+}, with T_C increasing from approximately 30 K in Lu_2CoMnO_6 to about 80 K in Ho_2CoMnO_6. This enhancement is attributed to the larger ionic radius and magnetic moment of Ho^{3+}, which modify lattice distortions and magnetic exchange interactions. These results highlight the crucial role of A-site rare-earth ions in tuning structure–property correlations in Co–Mn-based double perovskites.