Magnetic Ground State of Bilayer Nickelates $La_3Ni_2O_7$ and $La_2PrNi_2O_7$ via Neutron Diffraction

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The discovery of pressure-induced high-temperature superconductivity in Ruddlesden-Popper nickelates has positioned these materials as a critical platform for exploring unconventional superconducting mechanisms [1,2]. While a magnetically ordered spin-density wave (SDW) ground state has been suggested by local probes [3], its direct observation via neutron diffraction—the definitive method for probing magnetic order—has remained elusive due to challenges in sample stoichiometry and signal intensity [4].

In this study, we report the first direct observation of long-range SDW order in oxygen-stoichiometric bilayer $La_3Ni_2O_7$ and its Pr-substituted analogue $La_2PrNi_2O_7$ using high-intensity cold neutron powder diffraction [5]. Magnetic Bragg scattering emerges in both compounds below approximately 150 K. Our refined magnetic models reveal a structure characterized by alternating high- (0.66 μ_B) and low- (0.07 μ_B) magnetic moment stripes within single Ni layers, which stack antiferromagnetically along the out-of-plane direction.

A key finding is the presence of two distinct magnetic propagation vectors (q_1 and q_2) in pristine $La_3Ni_2O_7$, which we attribute to magnetic stacking polymorphs arising from out-of-plane stacking disorder. We demonstrate that partial Pr substitution suppresses this disorder, lifting the degeneracy between competing magnetic arrangements and stabilizing a single ground-state order (q_1). This improved structural and magnetic coherence correlates with the transition from filamentary to bulk superconductivity observed in these systems. These results provide a comprehensive understanding of the magnetic precursor states and their pivotal role in the emergence of superconductivity in layered nickelates.

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