Engineering the Magnetic Phase Diagram and Unequal Antipolar Displacement in Double-Perovskite Oxide Heterostructures

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The double-perovskite RE_2NiMnO_6 (RE= rare earth) family is characterized as being insulating ferromagnets, an unusual combination of properties. The Curie Temperature of La_2NiMnO_6 is T_c=280K, and for the other members of the family, T_c decreases linearly with the size of the *RE* ionic radius.

When grown as thin films [1], the ferromagnetic behavior occurs down to ultra-low thicknesses of (at least) 3 unit cells [2,3]. However, below 10 unit cells, the magnetic properties deteriorate due to an interfacial charge transfer caused by the polar discontinuity at the film/substrate interface.

The growth of tailor-made superlattices employing 2 distinct double perovskites RE_2NiMnO_6 (La and *RE* = Nd or Sm) featuring distinct Curie temperatures allows us to engineer the magnetic phase diagram and investigate the couplings at the magnetic interfaces [4]. Large periodicity superlattices conserve the individual para- to ferromagnetic transitions of the parent compounds. However, the Curie temperatures of the superlattice constituents collapse into a single transition for the lowest period samples, illustrating that low-periodicity samples behave as a unique material. This is a consequence of the magnetic order parameter propagating across the superlattice interfaces, as supported by a minimal Landau theory model. We also show that the superlattice interfaces enhance the Nd–Ni–Mn exchange interaction.

Further, scanning transmission electron microscopy combined with first-principles calculations confirms the predicted unequal antipolar displacement in our superlattices, strongly suggesting the presence of electric polarization. This is an important step towards establishing hybrid improper ferroelectricity in artificially layered heterostructures. In combination with our demonstration of robust ferromagnetism, the double perovskite superlattices bear large potential for future multiferroic systems.

References:

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