Tailoring magnetism in double-perovskite RE_2NiMnO_6 heterostructures

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Atomically engineered transition-metal-oxide heterostructures provide a versatile platform for probing and tailoring magnetic properties at the ultrathin limit and across interfaces. We focus on the double-perovskite family RE_2NiMnO_6 (RE = rare earth), a class of insulating ferromagnets —an unusual combination of properties. La_2NiMnO_6 exhibits a Curie temperature (T_c) of 280 K, while T_c decreases linearly across the series with decreasing RE-radius.

In RE_2NiMnO_6 thin films, ferromagnetic order persists down to at least 3 unit cells in epitaxial layers, and we further show that this behavior is retained in free-standing membranes.

To engineer the magnetic phase diagram and probe interfacial coupling, we fabricated atomic-scale-controlled superlattices comprising La_2NiMnO_6 and RE_2NiMnO_6 (RE = Nd, Sm) layers, which have distinct Curie temperatures. Large-period superlattices display two separate magnetic transitions characteristic of the parent compounds. In contrast, reducing the period yields a single transition, indicating that low-period heterostructures behave as a unique material. Interfaces also enhance the Nd-Ni-Mn exchange interaction.

Moroever, in these superlattices, scanning transmission electron microscopy and first-principles calculations reveal unequal antipolar displacements consistent with electric polarization, suggesting hybrid improper ferroelectricity. Together with robust ferromagnetism, these results highlight double-perovskite heterostructures as a promising platform for multiferroic devices.

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