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Epitaxial Engineering of Domain Walls and Distortions in Ferrite Heterostructures.

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The defining feature of ferroics is the ability of an external stimulus—electric field, magnetic field, or stress—to move domain walls. These topological defects and their motion enables many useful attributes, e.g., memories that can be reversibly written between stable states as well as enhanced conductivity, permittivity, permeability, and piezoelectricity. Although methods are known to drastically increase their density, the placement of domain walls with atomic precision has until now evaded control. Here we engineer the location of domain walls with monolayer precision and exploit this ability to create a novel multiferroic in which ferroelectricity enhances magnetism at all relevant length scales. Starting with hexagonal LuFeO_3 , a geometric ferroelectric with the greatest known planar rumpling, we introduce individual extra monolayers of FeO during growth to construct formula-unit-thick syntactic layers of ferrimagnetic LuFe_2O_4 within the LuFeO_3 matrix, i.e., $(\text{LuFeO}_3)_m/(\text{LuFe}_2\text{O}_4)_1$ superlattices. The severe rumpling imposed by the neighboring LuFeO_3 drives the ferrimagnetic LuFe_2O_4 into a simultaneously ferroelectric state and reduces the LuFe_2O_4 spin frustration. This increases the magnetic transition temperature significantly—to 281 K for the $(\text{LuFeO}_3)_9/(\text{LuFe}_2\text{O}_4)_1$ superlattice. Moreover, LuFeO_3 can form charged ferroelectric domain walls, which we align to the LuFe_2O_4 bilayers with monolayer precision. Charge transfers to these domain walls to alleviate the otherwise electrostatically unstable polarization arrangement, further boosting the magnetic moment. Our results demonstrate the utility of combining ferroics at the atomic-layer level with attention to domain walls, geometric frustration and polarization doping to create multiferroics by design.