Multiferroicity in Bulk and Thin-Film Hexagonal LuFeO$_3$

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The hexagonal manganites such as LuMnO$_3$ are prototypical examples of type-I multiferroics, where ferroelectricity stems from a structural distortion at high temperatures ($T_{FE} \sim 1000$ K) with antiferromagnetic order occurring only at a much lower temperature ($T_N \sim 100$K). The isostructural hexagonal LuFeO$_3$ is a promising candidate for enhancing many of the magnetic properties of this system, including the ordering temperature, as the isovalent Fe exhibits a larger spin relative to Mn as well as the potential for enhanced super-exchange interactions. Interest in this material has been bolstered by recent first-principles calculations predicting an enhanced ferromagnetic moment parallel to the ferroelectric moment as well as a mechanism for establishing robust magnetoelectric coupling$^1$. However, detailed studies of this material have been lacking to date due in large part to the instability of the hexagonal phase of LuFeO$_3$ in bulk, which instead forms in the orthorhombically distorted perovskite structure. In light of this, we have performed an extensive study of this material in two forms: (1) pure hexagonal LuFeO$_3$ thin-films grown by molecular beam epitaxy, and (2) bulk polycrystalline and single crystals stabilized by Mn-substitution on the Fe-site. We use a variety of techniques in to study these samples including magnetometry, neutron scattering, Raman spectroscopy, STEM and PFM. Through these, we find that both films and bulk samples are ferroelectric at room temperature and order magnetically with an enhanced $T_N$ relative to LuMnO$_3$. However, despite recent claims of room temperature magnetic order$^2$, we find that the maximum intrinsic ordering temperature for hexagonal LuFeO$_3$ is only 150 K. A weak ferromagnetic moment is observed along the c-axis of thin-films and bulk samples that vanishes rapidly with increasing Mn concentration, consistent with theoretical predictions. Lastly, we will discuss how our determination of the magnetic phase diagram as a function of temperature and Mn concentration provide insight into microscopic models put forth to explain the various transition and spin-reorientations in this family of compounds.

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