

Toward Room Temperature Ferroelectric Ferromagnets in (LuFeO₃)_n(LuFe₂O₄)_m Superlattices

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Introduction

The dream of room temperature ferroelectric ferromagnets is at the heart of the field of multiferroics and magnetoelectrics. Interface engineering is gaining traction in this regard. The idea is that by controlling composition and strain, room temperature exotic properties might emerge. One candidate for ferroelectric ferrimagnetism is a set of superlattices of the form $(LuFeO_3)_n:(LuFe2O4)_m$. Here, *n* and *m* are layer indices that run from 0 to 9. The parent compounds, $LuFe_2O_4$ and $LuFeO_3$, are well studied. $LuFe_2O_4$ is an antiferroelectric with a complex phase diagram, a series of charge ordering transitions above room temperature, and a 240 K ferrimagnetic ordering temperature. $LuFeO_3$ is polar at 300 K and orders antiferromagnetically at 147 K in a pattern in which symmetry allows a slight canting of the spins giving rise to weak ferromagnetism. Recent findings of inter-grown layers suggests the tantalizing prospect of room temperature ferroelectric ferrimagnetism with ordering temperatures up to 280 K in certain members of the $(LuFeO_3)_n:(LuFe_2O_4)_m$ series. The need to understand the electronic structure of these engineered materials, the precise charge ordering pattern, and the origin of the high temperature magnetism invites the approach taken here.

Results and Discussion

In order to investigate magnetoelectric coupling in a homologous series of epitaxial thin film superlattices with the prospect of room temperature ferrimagnetic ferroelectricity, we measured the optical absorption and magnetic circular dichroism of the (3,1) superlattice [Fig. 1] and compared our findings with complementary first principles calculations. Features in the dichroic rotation $\Delta \alpha(E)$ are directly proportional to magnetization and can be correlated to the density of states and to specific bands, so the spectrum reveals precisely which iron centers are involved in the magnetic response. Constant energy cuts in the region of the Fe²⁺ \rightarrow Fe³⁺ charge transfer gives optical hysteresis loops, which close with increasing temperature. Bringing the coercive field obtained by magnetic circular dichorism together with those from bulk magnetization² reveals the B - T phase diagram and demonstrates a significant portion of the magnetism originates in the LuFe₂O₄ layer. In order to test these ideas, we performed complementary measurements on the (7, 1) and (9, 1) superlattices. Although $T_{\rm C}$ is strongly enhanced in the (9, 1) material, the net magnetization - which is proportional to the MCD signal - is diminished. At the same time, we reveal fascinating new regions in the B - T phase diagram as well as the energy dependence of the coercive field.



Fig. 1: (a) STEM image of the (3,1) superlattice.¹ (b) Optical absorption of $(LuFeO_3)_3$: $(LuFe_2O_4)_1$. (c) Dichroic response taken on the split helix. (d) Optical hysteresis loop of the (3,1) superlattice closing with increasing temperature. (e) Coercive field vs. temperature for the (3,1) superlattice combining magnetization² with MCD data.

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References

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