Charge Order, Dynamics, and Magneto-structural Transition in Multiferroic $LuFe_2O_4$

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Introduction

Complex oxides take advantage of the unique and flexible properties of transition metal centers to govern bonding and local structure. Further, the delicate interplay between charge, structure, and magnetism yields important consequences for functionality and cross-coupling. Iron-based materials such as multiferroic $BiFeO_3$ and $LuFe_2O_4$ have attracted recent attention. In this work, we focus on $LuFe_2O_4$, a frustrated system with a series of phase transitions that gives rise to electronically-driven multiferroicity.

Results and Discussion

To elucidate the charge excitations and understand how they correlate with structure and magnetism, we measured the optical and Mossbauer spectra, magnetization, and x-ray scattering of $LuFe_2O_4$. We compare our comprehensive results to recent electronic structure calculations and to spectral data on classical magnetite. We demonstrate that strong Fe^{2+} to Fe^{3+} charge fluctuation persists even in charge ordered states characterized by superstructure reflections, and it persists down to T_{LT} below which Bragg splitting indicates that strong monoclinic distortions occur. These observations are consistent with the "order by fluctuation" mechanism, in which case $\sqrt{3} \times \sqrt{3}$ charge order is preferable for entropy reasons and stabilized by the charge fluctuation in this geometrically frustrated system. As in magnetite, we analyze the results in terms of a polaron picture, extracting a large effective mass for the charge carriers. On the other hand, Fe^{2+} on-site excitations are sensitive to the magnetic transition at 170 K and display a sizable magneto-optical effect. Combining our spectral, magnetic, and structural data, we generate an H-T phase diagram and show that the low temperature transition can also be driven by a magnetic field. These results demonstrate the important interplay between charge, structure, and magnetism.

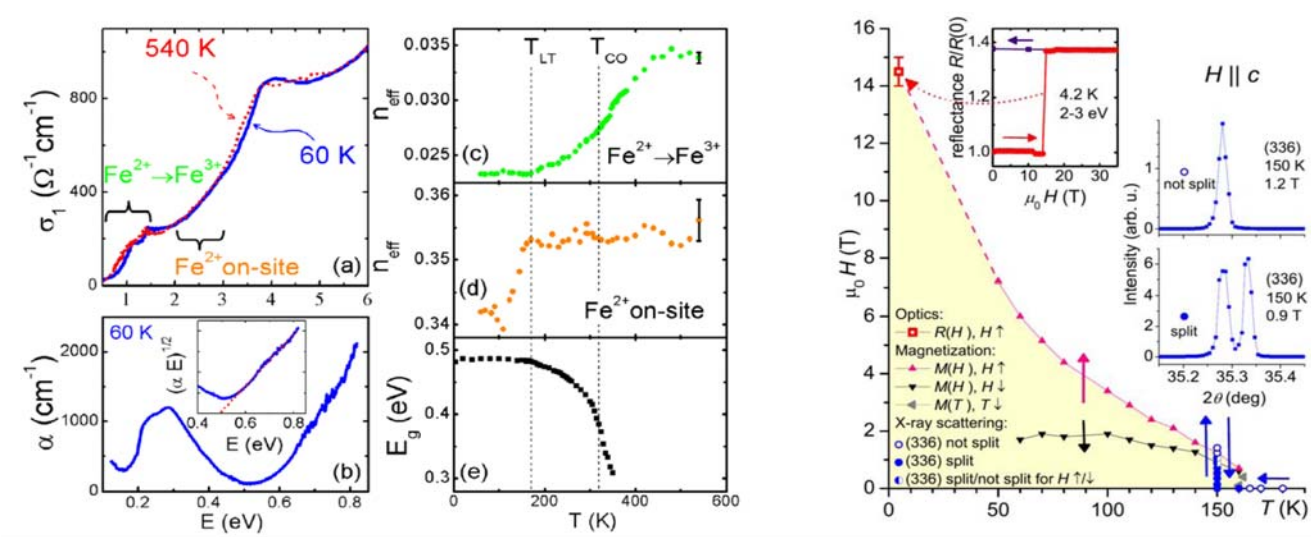

Figure 1: Optical properties of $LuFe_2O_4$ and H-T phase diagram generated from combined optical, magnetic, and structural studies. (a) σ_1 vs energy E at 540 and 60 K calculated from a Kramers-Kronig analysis of reflectance. Brackets indicate assignment from electronic structure calculation, (b) α vs E calculated from transmittance, showing the charge transfer edge starting from ~0.5 eV, (c, d) $n_{eff}(T)$ calculated by sum rules for Fe^{2+} to Fe^{3+} charge transfer and Fe^{2+} on-site excitations. (e) $E_g(T)$ calculated from an absorption edge fit assuming an indirect gap. We generate an H-T phase diagram.

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