Field-Induced Behavior in Multiferroic $LuFe_2O_4$

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Materials that offer the possibility to simultaneously control magnetic and electric degrees of freedom are the subject of intense interest [1]. Recently multiferroic materials have been identified that show a large coupling between electric and magnetic degrees of freedom. Ferroelectricity driven by either a magnetic or charge ordering process appears to be the origin of the large coupling and hence understanding the underlying electronic interactions is crucial for further insight into multiferroicity [1].

$LuFe_2O_4$ has attracted attention as a novel ferroelectric material where ferroelectricity is driven by the electronic process of charge ordering of Fe^{2+} and Fe^{3+} ions and for indications of coupling between electronic and magnetic degrees of freedom [2-6]. $LuFe_2O_4$ is a member of the RFe_2O_4 (where R denotes rare earth element) family, the physical properties of which depend strongly on oxygen stoichiometry. For example, nearly stoichiometric YFe_2O_4 exhibits three-dimensional (3D) magnetic order while oxygen deficient YFe_2O_4 exhibits two-dimensional (2D) magnetic order [7]. $LuFe_2O_4$ exhibits multiple phase transitions. 2D charge correlations are observed below 500 K, while below ~ 320 K 3D charge order is established, roughly coinciding with the onset of ferroelectricity [2,8]. Magnetic order appears below 240 K and 2D ferrimagnetic order has been suggested based upon neutron scattering studies [9]. However, strong sample dependent behavior observed in other members of RFe_2O_4 [7] suggests that unraveling the interesting behavior of $LuFe_2O_4$ requires paying due attention to sample quality.

Recently, we have performed neutron diffraction measurements on highly stoichiometric single crystal specimens ([10] and Fig. 1). These measurements show that, in contrast to previous work, below 240 K 3D magnetic correlations exist with magnetic intensity appearing at (1/3 1/3 L) where L may take on integer *and* half integer values. We were able to refine the magnetic structure as a ferrimagnetic spin configuration with a propagation vector of (1/3 1/3 0). The magnetic intensity appearing on peaks where L is a half-integer is a consequence of the charge ordering at ~320 K. In addition we found a second transition at 175 K where significant changes in magnetic peak intensities occur, many reflections are broadened, and a new set of satellites appear, for which polarized neutron scattering experiments indicate [11] a significant magnetic component.

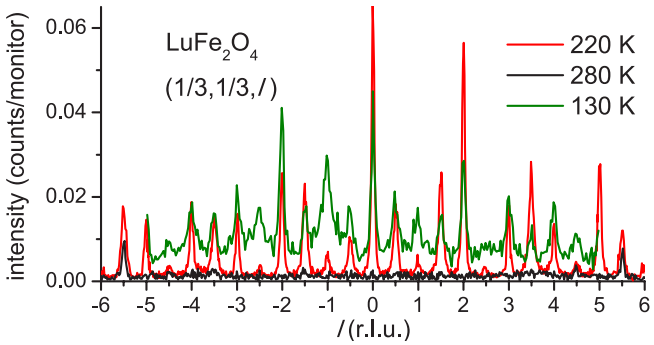

Fig 1. Zero field scattering along (1/3 1/3 L).

Further understanding of the magnetoelectric behavior presented by $LuFe_2O_4$ can be gained through neutron diffraction measurements under applied fields. To that end we have performed additional experiments on the C5 triple axis with the M2 horizontal magnet. We used vertically focusing monochromator and flat analyzer PG-002 crystals with a fixed final energy of E_f = 30.5 meV. Two PG filters were located in the scattered side to eliminate higher order wavelength contamination from the beam. In addition a liquid nitrogen cooled sapphire filter was used in the main beam to reduce the fast neutron background. Horizontal collimations were set to [none, 0.48°, 0.55°, 1.2°]. With the M2 horizontal magnet, magnetic fields of up to 2.6 T were applied from room temperature down to the lowest temperature of ~ 1.8 K. The sample was aligned in the (HHL) plane and with the 350° horizontal access of the M2 magnet we were able to apply the field parallel to the c-axis while probing a large number of wavevectors in this scattering plane.

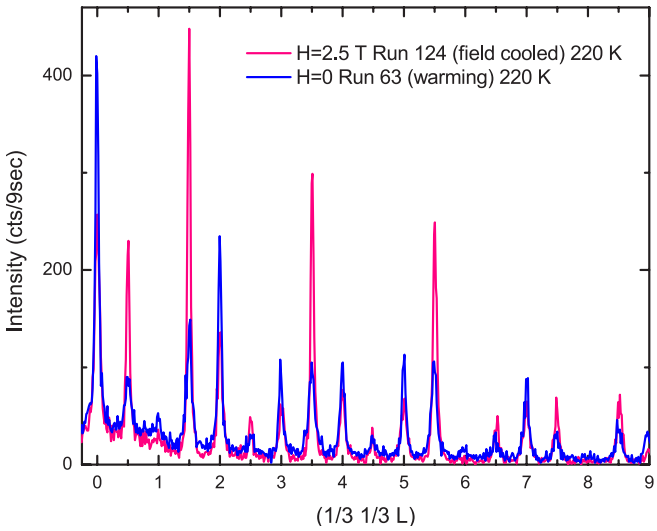

Fig 2. Changes in the scattering along (1/3 1/3 L) cooling with a field of 2.5 T applied along the c-axis.

Figure 2 shows substantial changes to the scattering below the 240 K ferromagnetic transition when cooling with a field applied along the c-axis. The main effect of the field appears

to be an increase in intensity of peaks where L is a half-integer while the peaks where L is an integer decrease in intensity. On the other hand, no change in intensity in peaks due solely to charge order of Fe^{2+} and Fe^{3+} was observed for peaks at high Q. A possible explanation of this is that one of the Fe-sites remains unordered below the ferromagnetic transition at 240 K due to frustrated interactions and that an applied field tips the balance in favor of ordering on all Fe-sites.

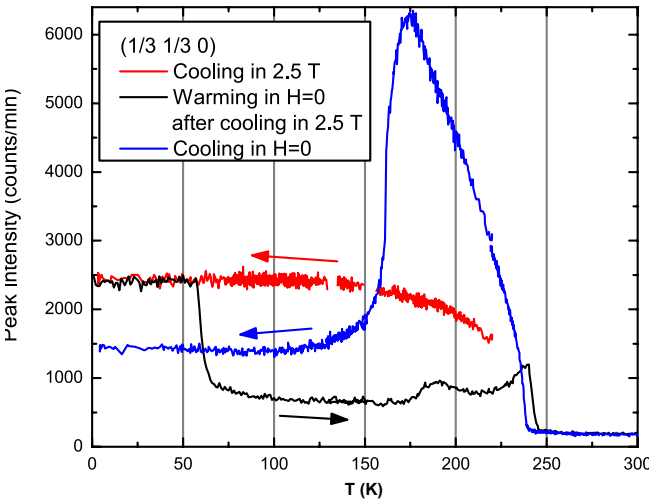

Fig 3. *The scattering at (1/3 1/3 0) for different field configurations.*

Figure 3 displays the changes in the (1/3 1/3 0) peak for various applied field configurations. Of particular note is the strong thermal remanence observed when the sample is cooled under an applied field that is then removed at the base temperature and subsequently warmed to room temperature. When cooled under a field of 2.5 T the sample never enters the low temperature magnetic phase as characterized by the broad peaks appearing below 175 K in the zero field cooled configuration (figure 1). When the field is removed at 4 K no change in intensity is observed for peaks along (1/3 1/3 L). However, when warming the field cooled sample, a magnetic state abruptly locks in that appears to be similar to the low temperate state with extremely broad magnetic peaks which appears below 175 K in the zero field cooled configuration.

In summary, the field dependent behavior is extremely rich and further quantitative work is ongoing to understand this behavior in further detail.

References

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