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Synthesis and characterization of mixed-valence LuFe₂O_{4-δ}: Effect of stoichiometry δ

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Abstract content
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LuFe₂O₄ exhibits a unique mixed-valence superstructure arising from charge-ordering (CO) within the lattice. The unit cell consists of a characteristic stacking of bi-layers, in which Fe is configured in a triangular network. This results in charge and spin frustration in the material, with a resulting plenitude of degenerate magnetic-electronic ground states. Furthermore the Fe is coordinated with oxygen in a trigonal bi-pyramidal polyhedron, a rather unusual co-ordination geometry for inorganic compounds. This mixed-valence characteristic within the bi-layers has been claimed to give rise to a dipole moment, i.e., ferroelectric effects arising from the CO. However such claims of electronically driven CO are under dispute [1]. Recent work has also demonstrated remarkable oxygen storage capacities in this compound [2]. Previous studies have indeed shown that the Fe²⁺/Fe³⁺ ratio and magnetic ordering temperature T_N are influenced by the oxygen stoichiometry [3-4]. We will report our investigations of the effect of oxygen stoichiometry on the CO and magnetic-electronic properties of LuFe₂O_{4-delta;}, from a comparison of stoichiometric and oxygen deficient samples. Such samples of LuFe₂O_{4-delta;} of varying oxygen stoichiometry, delta;, have been synthesised by solid state reaction as a polycrystalline powder. These have been characterised by x-ray diffraction, ⁵⁷Fe Mouml;ssbauer-effect spectroscopy (MES), SQUID magnetometry and TGA chemical analysis. Using different masses for the overall starting mixture has a radical effect on the purity of the as-synthesised LuFe₂O_{4-delta;} sample. Magnetisation measurements show that T_N is confined to 245-250 K for the synthesised samples, albeit with quite significant differences in the magnetisation-temperature envelopes. Variable cryogenic temperature MES measurements are used to compare the effect of oxygen content on both the CO and magnetic hyperfine structure.

[1]. Angst M, Phys. Status Solidi RRL 7, 375 (2013).
[2]. Hervieu M et al., Nature Materials 13, 74 (2013).
[3]. Yang HX et al., J. Phys. Condens. Matter 24, 435901 (2012).
[4]. Wang F et al., J. Appl. Phys. 113, 06 (2013).

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