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Charge and magnetic ordering dynamics under pressure in LuFe₂O₄

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Abstract content
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Multiferroics offer the possibility to combine ferroelectric and magnetic ordering in materials, hence the realisation of multifunctional devices [1]. LuFe₂O₄ is one such compound. Initially thought to be the prototypical compound for charge-order (CO) based ferroelectricity, nowadays it is attracting interest because of its pronounced magneto-electronic coupling, as well as the spin/charge frustration anticipated in the triangular network of Fe atoms within the bilayers of the rhombohedral unit cell. Previous work has shown that the low pressure (LP) phase has a T_N ≈ 250 K and a CO temperature of 330 K in the highly stoichiometric sample investigated here [2]. In this work pressure has been used as a thermodynamic variable to tune the magnetic-electronic properties of LuFe₂O₄. Generation of hydrostatic pressure was by means of a diamond anvil cell, and ⁵⁷Fe Mössbauer spectroscopy (MS) was used to probe magnetic-electronic phase transformations. Measurements were performed up to ˜30 GPa at room temperature. We find evidence of an electron hopping component Fe²⁺<->Fe³⁺ already present at ambient pressure on the time scale of the Mössbauer effect, with a hopping frequency of 1.4 MHz. At ˜5 GPa the CO breaks down, and the hopping component completely dominates, with the hopping frequency increasing to ˜10 MHz. The MS spectral line shape at 7 GPa shows new emerging features, supposed to be evidence of a new structural phase initiating at this pressure, consistently with previous x-ray diffraction studies [3]. Furthermore, at 10 GPa new magnetic hyperfine structure is observed in the high pressure (HP) phase at room temperature. This suggests a change of the LP spin frustrated state to that of a new magnetic-state in the HP phase 'with reduced frustration'. The MS spectral envelope of the HP phase suggests that two magnetic-electronic states co-exist, a new CO state is stabilised at HP and unusual spin states are also involved. This is consistent with the claims of previous x-ray and electron diffraction studies of this HP phase recovered to ambient conditions [4].

[1] X. S. Xu et al., PRL 101, 227602 (2008).
[2] S. Lafuerza et al., PRB 88, 201304 (2013).
[3] A. D. Christianson et al., PRL 100, 107601 (2008).
[4] J. Rouquette et al., PRL 105, 237203 (2010).

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