Interplay of charge order and magnetism in magnetoelectric LuFe₂O₄

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Four years ago, LuFe₂O₄ was proposed to be a multiferroic, with ferroelectricity arising from charge order (CO) [1], a novel mechanism providing a potential route to practical magnetoelectric devices. Applications potential was also suggested by the subsequent discovery of a giant magneto-dielectric effect at room temperature. Understanding magnetoelectric coupling in this material requires elucidating the microscopic origins of electric and magnetic properties, i.e., charge and spin order. We have investigated these ordering phenomena mainly with (non-resonant and resonant) x-ray and (unpolarized and polarized) neutron diffraction on highly homogeneous single crystals, partly under the influence of electric or magnetic fields.

Both charge and spin correlations lead to competing ordering instabilities, with superstructure reflections of type A:(1/3,1/3,0) and B:(1/3,1/3,3/2), and superimposed apparent long-wavelength modulations. Long-range CO realized below 320 K is of type B, surprisingly corresponding to an <u>antiferroelectric arrangement of polar subunits</u>, whereas short-range correlations at higher T are of the (ferroelectric) type A [2]. Type A long-range CO could not be stabilized by electric fields due to (likely intrinsically) too high leakage at high T. Spins order below 240 K, with type A in zero magnetic field [3]. Moderate magnetic fields instead stabilize a type B spin order – though with no apparent effect on CO.

Hallmark of a sizeable spin-charge coupling is an additional phase transition at 170 K. It corresponds to a structural transition driven by CO and the complete freezing of remaining charge fluctuations [4], as well as to the transition to a less well-ordered magnetic state [3] with dynamically "glassy" features.

References

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