



Contribution ID: 53

Type: Poster

On the origin of large magnetic anisotropy in ϵ -Fe₂O₃

Thursday, 8 September 2022 19:35 (20 minutes)

The epsilon phase of Fe₂O₃ is a ferrimagnetic polymorph that stands out for a remarkable magnetic anisotropy ($K_u \sim 2 \cdot 10^5$ J/m³) and sizeable remnant magnetization at room temperature (~ 50 kA/m)[1] making it appealing for applications in magnetic recording and electromagnetic applications in the range of millimetre waves [2]. Moreover, ϵ -Fe₂O₃ is also interesting from a more fundamental point of view, featuring low-temperature magnetoelectric properties [3] and three distinct zero-field magnetic orders between 10 K and its paramagnetic state above ~ 850 K [4]. Such a rich phase diagram has much to do with its non-centrosymmetric structure (*Pna21*), more complex than for other ferric oxides, with four different Fe sites in the asymmetric unit cell: three in an octahedral (O_h) and one in a tetrahedral (T_d) local symmetry [1].

Despite having attracted increasing attention over the last decades, the intriguing magnetic order in ϵ -Fe₂O₃ is still poorly understood. Here we present recent results of temperature-dependent X-ray and neutron diffraction, X-ray absorption spectroscopy (XAS) and X-ray magnetic circular dichroism (XMCD) experiments that provide new insights into the origin of magnetic anisotropy in ϵ -Fe₂O₃ and \square -(Fe_{1-x}Cr_x)₂O₃ in the form of nanoparticles (NPs) and thin films. The Fe³⁺ with Cr³⁺ substitution is very revealing due to its expected isovalent and magnetic isotropic character. Although trivalent iron could be thought as behaving in a similar way, a nonzero orbital angular momentum m_L value has been proposed for the Fe₂ crystallographic site, which might act as the primary source for the magnetic anisotropy responsible of the gigantic coercive field. Thus, depending on the preferential entering sites for Cr in the ϵ -Fe₂O₃ structure, a systematic reduction in m_L and a weakening of the spin-orbit coupling along the series may be expected. Our XMCD investigation has addressed this controversy.

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Session Classification: List of posters presented during the conference