Asphericity of magnetisation density and anisotropy in rare-earth pyrochlores via polarized neutron diffraction and iterative entropy maximization

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Lanthanide ions play a crucial role in various research fields. Much theoretical effort, that aims understanding and enhancing magnetic anisotropy in multiferroics and molecular magnetic materials, shows that the variation of magnetisation anisotropy is accompanied by important changes of 4f-electron, spin and orbital distributions. However, the experimental determination of the shape of these distributions is a non-trivial task especially in the case of unquenched orbital moment. Here, the procedure of magnetisation density reconstruction in lanthanides with unquenched orbital moment is developed, based on the iterative entropy maximization and the site susceptibility approach. The calculation was performed by recently developed code written as part of a crystallographic CrysPy library [1].

We illustrate the possibilities of the method by the first joint magnetisation density reconstruction and susceptibility refinement of locally anisotropic lanthanide pyrochlores $R_2Ti_2O_7$ $(R = Tb, Ho, Er \text{ and } Yb)$ [2]. An oblate asphericity of Tb^{3+} density and prolate these of Ho^{3+} and Yb^{3+} was revealed (fig.1). Reconstructed distributions and refined susceptibility parameters are compared with these predicted by the crystal field theory in frame of single ion anisotropy model using McPhase software [3].

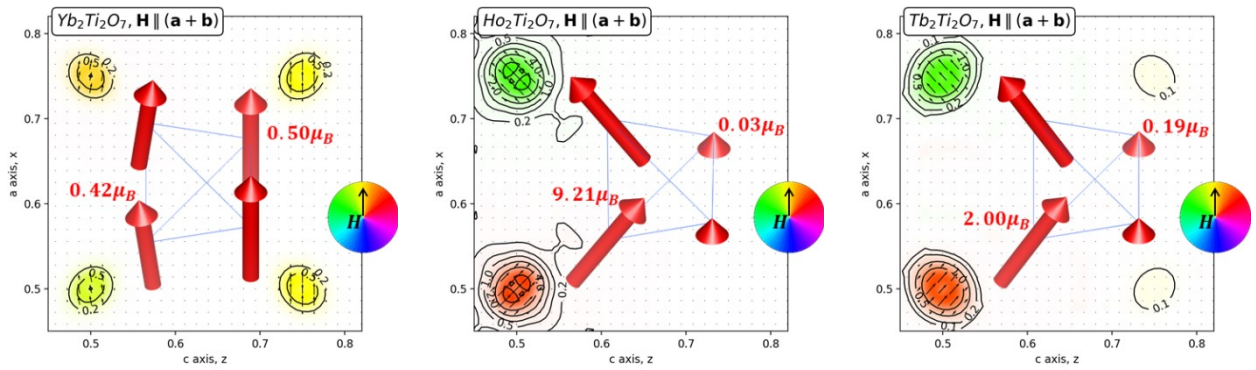

Figure 1. Magnetic moment distributions obtained for $R_2Ti_2O_7$ $(R = Tb, Ho, Yb)$ at 5K 1T by the MEM procedure. The projection of the magnetic moments on the ac-plane is depicted. Resulting (integrated) magnetic moments of R-ions are shown in insets. HSL color wheel with hue equal 60° is used in figures to visualize the deviation of induced magnetic moments from the field direction.

1. GitHub page of CrysPy library: https://ikibalin.github.io/cryspy/
2. H. Cao et al. Phys. Rev. Lett. (2009) **103**, 056402
3. M. Rotter et al J. Phys.: Conf. Ser. (2011) **325** 012005.

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