

Magnetization distribution in anisotropic antiferromagnetic systems: An example of Co-olivine

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Neutron diffraction plays an important role for studying magnetism. The classical polarized neutron diffraction technique (flipping-ratio measurements in an applied magnetic field) is used to study the magnetization distribution in paramagnetic and ferromagnetic materials [1]. It is much more difficult to make such measurements in antiferromagnetic systems where the cross-section is not often polarization dependent and, therefore, the classical method is not applicable. For some magnetic structures the recently developed zero-field spherical neutron polarimetry technique allows the determination of the magnetization distribution in the ground antiferromagnetic state [2]. Here, we show the possibilities and limitations of the techniques on the example of antiferromagnetic Co-olivine, Co_2SiO_4 [3]. Natural olivine ($\text{Mg}_{1-x}\text{Fe}_x$) $_2\text{SiO}_4$ is a major phase in the solar system. However, besides the fundamental importance for geologists and mineralogists, olivines display a surprising variety of chemical and physical properties. Synthetic Co_2SiO_4 crystallizes in the olivine structure and shows antiferromagnetic ordering below 50 K. Its magnetic structure has been studied by means of polarized neutron diffraction (flipping-ratio measurements) on a single crystal. The data refinement was done by using the recently developed local susceptibility approach, which assigns a site susceptibility tensor to each crystallographically independent site [4]. The results of the model-based refinement were compared with the model-free maximum-entropy reconstruction of the magnetization density. A field-induced magnetization shows anisotropy, which is rather small at high temperatures but becomes strongly enhanced when approaching the ordering temperature. The refinement revealed a significant orbital component to the total magnetic moment of the Co^{2+} ions. In addition, a non-negligible amount of magnetic moment on the oxygen positions was found, indicating a delocalization of the magnetic moment from Co towards neighbouring O owing to superexchange coupling. The relative strength of the exchange interactions is discussed. Some other systems and future perspectives in polarized neutrons are also briefly reported.

[1]. R. Nathans, C. G. Shull, G. Shirane, and A. Andresen, *J. Phys. Chem. Solids* 10, 138 (1959)

[2]. P. J. Brown, J. B. Forsyth, E. Lelièvre-Berna, and F. Tasset, *J. Phys.: Condens. Matter* 14, 1957 (2002)

[3]. A. Sazonov, M. Meven, V. Hutanu, G. Heger, T. Hansen and A. Gukasov, *Acta Cryst. B.* 65, 664 (2009)

[4]. A. Gukasov and P. J. Brown, *J. Phys.: Condens. Matter* 14, 8831 (2002).