

*Tailoring molecular multiferroic compounds: neutron studies on formate frameworks*

Laura Cañadillas-Delgado<sup>1</sup>, Lidia Mazzuca<sup>1</sup>, Oscar Fabelo<sup>1</sup>, José Alberto Rodríguez-Velmazán<sup>1</sup>, Francesc Lloret<sup>2</sup>, Juan Rodríguez-Carvajal<sup>1</sup>

<sup>1</sup>Institut Laue Langevin, Grenoble, France, <sup>2</sup>Instituto de Ciencia Molecular/Departament de Química Inorgànica, Universitat de València, Valencia, Spain  
E-mail: lcd@ill.fr

Considerable attention is currently focused on metal-organic polymers combining two or more functionalities, e.g., conductivity and magnetism, magnetic and optical properties, porosity and magnetism. The inclusion of two different metallic atoms into the niccolite-like structure framework have led to the formation of isostructural compounds of formulae  $[\text{NH}_2(\text{CH}_3)_2]_n[\text{M}(\text{II})\text{Fe}(\text{III})(\text{HCOO})_6]_n$  with  $\text{M}(\text{II}) = \text{Fe}(1), \text{Co}(2)$  and  $\text{Mn}(3)$ .

The nuclear characterization of these compounds has been done combining high resolution neutron diffraction together with synchrotron X-ray diffraction at 45K.

In compound 1 neutron studies show a sequence of phase transition involving nuclear transformation as well as a long range magnetic order. The nuclear phase transition involves an ordering of the  $\text{NH}_2(\text{CH}_3)_2$  counteranion within the cavities framework which is related with a change in the electric behaviour from paraelectric to antiferroelectric.[1] However, in compounds 2 and 3, multipattern refinements combining X-rays and neutron radiation, permit the localization of the hydrogen atoms of the  $[\text{NH}_2(\text{CH}_3)_2]$  counter ions, and discards the occurrence of a structural phase transition.[2]

Below TN, the magnetic order in these compounds vary from an antiferromagnetic behaviour with a weak spin canting for compound 3 to a ferrimagnetic behaviour for 1 and 2 compounds, due to the competition of the different magnetic moments present in each sublattices (the different spin state involved in those complexes are  $S = 5/2$ , for  $\text{Fe}(\text{III})$  and  $\text{Mn}(\text{II})$  while it is  $3/2$  and  $2$  for  $\text{Co}(\text{II})$  and  $\text{Fe}(\text{II})$ , respectively).

Neutron studies using single crystal and powder diffraction techniques have been carried out at Institut Laue Langevin (Grenoble, France) to elucidate the different magnetic behaviour present in these compounds.

[1] Cañadillas-Delgado, L. et al. (2012) J. Am. Chem. Soc, 134(48), 19772-19781.

[2] Mazzuca, L. et. al. (2017), Inorg. Chem. 56, 197–207.

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