

Dynamic off-centering of Cr³⁺ ions and short-range magneto-electric clusters in CdCr₂S₄

Wednesday, 27 November 2013 09:50 (20 minutes)

Magnetoelectric materials experience a renewed interest due to its enhanced multifunctional properties, which are extremely appealing for technological applications into memories that could be written electrically and read magnetically or vice-versa [1,2]. Such properties can be affected by local distortions at the atomic scale, therefore requiring understanding the role of the local, polar and magnetic clusters³. Local distortions are of special importance on a class of disordered materials, the relaxor-like ferroelectrics, where in many cases we find the competition/coexistence between short-range and long-range order. Among these relaxor-like systems appears CdCr₂S₄ chalcogenide⁴.

Our experimental findings, based on complementary Pair Distribution Function (PDF), Perturbed Angular Correlations (PAC), Magnetization [M(T)] and Dielectric $\epsilon(T)$ measurements, address the presence of a new dynamic state caused by the presence of simultaneous polar and magnetic clusters, “multiferroic clusters”. The nature of the effects described in the recent literature is set to arise from the atomic displacement of Cr³⁺ well above the ferromagnetic ordering temperature. These new insights directly prove the existence of Cr³⁺ ion off-center displacements, to happen with the onset of local polar distortions. PDF analysis shows small displacement of the Cr³⁺ site ($\Delta r_{\max} \approx 0.015 \text{ \AA}$) explaining the low polarization values obtained. Additionally, Electric Field Gradient (EFG) measurements evidence the dynamic character of the off-centering which leads to the formation of local electric dipoles, which are also responsible for the observed magnetic correlations between Cr³⁺ neighbors. This correlation between electric and magnetic orders is shown to justify the peculiar low-field χ^{-1} (T) measurements. Additionally, upon considering the Landau theory of phase transitions with an expansion of the free energy of a multiferroic system including a bi-linear magnetoelectric coupling term, the simulated results match the experimental ones.⁵

References

- [1] Chu, Y.H., Martin, L.W. et al. Electric-field control of local ferromagnetism using a magnetoelectric multiferroic, *Nature Materials* 7, 478 –482 (2008).
- [2] Bibes, M. and Barthélémy, A. Multiferroics: Towards a magnetoelectric memory, *Nature Materials* 7, 425 – 426 (2008).
- [3] Lopes, A. M. L. et al. New Phase Transition in the Pr_{1-x}CaxMnO₃ System: Evidence for Electrical Polarization in Charge Ordered Manganites. *Phys. Rev. Lett.* 100, 155702 (2008).
- [4] Sun, C. P. et al. Colossal electroresistance and colossal magnetoresistance in spinel multiferroic CdCr₂S₄. *Appl. Phys. Lett.* 96, 122109 (2010).
- [5] G. N. P. Oliveira et al, Dynamic off-centering of Cr³⁺ ions and short-range magneto-electric clusters in CdCr₂S₄, *Phys. Rev. B* 86, 224418 (2012).

Primary author: Mr OLIVEIRA, Gonçalo (CFNUL-Centro de Física Nuclear, Universidade de Lisboa, Av. Prof. Gama Pinto, 2, 1649-003, Lisboa, Portugal & IFIMUP and IN-Institute of Nanoscience and Nanotechnology, Department of Physics and Astronomy of FCUP, University of Porto, Rua do Campo Alegre, 687, 4169-007 Porto, Portugal)

Co-authors: Dr PEREIRA, André M. (IFIMUP and IN-Institute of Nanoscience and Nanotechnology, Department of Physics and Astronomy of FCUP, University of Porto, Rua do Campo Alegre, 687, 4169-007 Porto, Portugal); Dr SANTOS, António m. (Quantum Condensed Matter Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6393, USA); Dr LOPES, Armandina M. L. (CFNUL-Centro de Física Nuclear, Universidade de Lisboa, Av. Prof. Gama Pinto, 2, 1649-003, Lisboa, Portugal); Dr SOUSA, Célia T. (IFIMUP and IN-Institute of Nanoscience and Nanotechnology, Department of Physics and Astronomy of FCUP, University of Porto, Rua do Campo Alegre, 687, 4169-007 Porto, Portugal); Dr CORREIA, Guilherme M. (CTN/IST, Instituto Superior Técnico, Universidade de Lisboa, Estrada Nacional 10, 2686-953 Sacavém, Portugal.); Dr ESTEVES DE ARAUJO, Joao Pedro (Universidade do Porto Laboratório de Física); Dr AMARAL, João S. (CICECO and Department of Physics, Universidade de Aveiro, 3810-193 Aveiro, Portugal); Dr MENDONÇA, Tânia M. (IFIMUP and IN-Institute of Nanoscience and Nanotechnology, Department

of Physics and Astronomy of FCUP, University of Porto, Rua do Campo Alegre, 687, 4169-007 Porto, Portugal); Dr AMARAL, Vitor S. (CICECO and Department of Physics, Universidade de Aveiro, 3810-193 Aveiro, Portugal); Dr REN, Yan (Advanced Photon Source, Argonne, Argonne National Laboratory, Illinois 60439, USA)

Presenter: Mr OLIVEIRA, Gonçalo (CFNUL-Centro de Física Nuclear, Universidade de Lisboa, Av. Prof. Gama Pinto, 2, 1649-003, Lisboa, Portugal & IFIMUP and IN-Institute of Nanoscience and Nanotechnology, Department of Physics and Astronomy of FCUP, University of Porto, Rua do Campo Alegre, 687, 4169-007 Porto, Portugal)

Session Classification: Applications I