$CdCr_2S_4$: Randomly oriented dipoles arising from Cr^{3+} dynamic off-centering.

G. N.P. Oliveira (1,2)*, A.M. Pereira (2), A.M.L. Lopes (1), J.S. Amaral (2,3) A.M. dos Santos (4), T.M. Mendonça (2), C. T. Sousa(2), Y. Ren (5), V. S. Amaral (3), J. G. Correia (6), J. P. Araújo (2)

(1) CFNUL – Center Nuclear Physics, University of Lisbon, Av. Prof. Gama Pinto, 2, 1649-003, Lisboa, Portugal, (2) IFIMUP and IN - Institute of Nanoscience and Nanotechnology and Department of Physics and Astronomy, Faculty of Sciences, University of Porto, Rua do Campo Alegre, 687, 4169-007 Porto, Portugal, (3) CICECO and Departament of Physics, University of Aveiro, Campus Universitário de Santiago, 3810-193 Aveiro, Portugal, (4) Neutron Scattering Science Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA, (5) APS - Advanced Photon Source, Argonne National Laboratory, Argonne, Illinois 60439, USA, (6) IST/ITN, Instituto Superior Técnico, Universidade Técnica de LIsboa - EN 10 - Apartado 21, 2686-953 Sacavém, Portugal
*Presenting author : goliveira@fc.up.pt

The quest for improved multiferroic materials is, nowadays, a hot topic of research promising routes for the development of several technological applications based on the magneto-electric coupling [1-4]. In this respect, the most appealing multiferroic materials are those, which display a strong coupling between the magnetic and polar degrees of freedom [5-7].

Our studies provide experimental evidence for the origin of the relaxor-like behavior on the cubic spinel $CdCr_2S_4$. Here we definitely settle that this behavior arises from Cr off-centering displacement from its coordination sphere being thus responsible for the observed colossal magneto-electrical effects. Our findings were achieved by a singular combination of local probe techniques namely Pair Distribution Function (PDF) and Perturbed Angular Correlation (PAC). We further show that the off centering of the magnetic Cr-ion gives rise to a peculiar entanglement between the polar and magnetic degrees of freedom, stabilizing, in the paramagnetic phase, short range magnetic clusters. This phenomenon was unveiled by low-field high-resolution magnetization measurements, analyzed using a modified Landau theory with a linear coupling between the magnetic and polar order parameters.

Our work provides fundamental comprehension of the magneto-electric coupling on the $CdCr_2S_4$ system, essential for opening new routes to tune similar materials for suitable applications namely in the Spintronics industry. Furthermore, we point out the PDF and PAC techniques importance in unveiling the behavior presented by this system.

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