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

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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 ε(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 (Δrmax≈0.015 Å) explaining the low polarization values obtained. Additionally, Electric Filed 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

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