Probing Electrostructural Coupling on Magnetoelectric $CdCr_2S_4$

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Magnetoelectric materials experienced a renewed interest in the last decade. Their enhanced multifunctional physical properties are extremely appealing for technological application into memories that could be written electrically and read magnetically or vice-versa[1,2]. These properties can be affected by local distortions, therefore requiring the role of the local, polar and magnetic clusters to be ascertained[3]. The local distortions are of special importance into a class of disordered materials, the relaxor-like ferroelectrics, where at this level, we find in many cases the competition/coexistence between short-range and long-range orders. Among these relaxor-like systems appears $CdCr_2S_4$ chalcogenide[4].
Our experimental findings, based on complementary Pair Distribution Function (PDF), Perturbed Angular Correlations (PAC), Magnetization [M(T)] and Dielectric $\varepsilon(T)$ 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 Cr^{3+} well above the ordering temperature. These new insights directly prove the Cr^{3+} ion off-center displacements, *i.e.*, with the onset of local polar distortions, activated by thermal mechanism. PDF analysis shows small displacement of the Cr^{3+} site ($\Delta r_{max} \approx 0.011$ 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 responsible also for the observed magnetic correlations between Cr^{3+} neighbors. This correlation between electric and magnetic orders is shown to justify the peculiar low-field χ^{-1} (T) measurements. Additionally, when it is considered the Landau theory of phase transitions and an expansion of the free energy of a multiferroic system including a linear magnetoelectric coupling term the simulated results match the experimental ones.
Finally, we highlight that the discovery of the present atomic displacement mechanism is related to the relaxor behavior in the PM regime. This work open wide perspectives for the future understanding of anomalous behavior recently observed above the T_C and towards the novel design of multiferroic materials.

References

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