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# Probing the local structure in Multiferroic $SmCrO_3$

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Rare-earth orthochromites of the formula $RCrO_3$ R=Dy, Pr, Ho, Yb, Er, Y, Lu, Sm are currently at the center of great controversy regarding ferroelectricity. While dielectric constant anomalies near $400 - 500$ K in the heavier rare-earth chromites were associated with non-centrosymmetry, others claim that the polarization observed in these systems is due to the combined effect of the electric applied field, that breaks the symmetry, and exchange-field on the R ion from the Cr sub-lattice. Accordingly to these claims, no spontaneous ferroelectric polar-order exists in these systems and the presence of a magnetic R-ion is essential to induce a metastable ferroelectric state. Contrarily, the appearance of ferroelectricity without direct correlation to the magnetic order, arising from polar octahedral rotations and/or cation displacements, was recently claimed. [1,2]

Clearly, additional efforts are needed to definitely validate these claims. Since these properties might emerge from local structural landscapes that are not well described by long-range average structural methods, the use of local probe studies, such as Perturbed Angular Correlation (PAC) spectroscopy, provide relevant knowledge. In this work the $SmCrO_3$ compound was studied. The temperature dependent of the electric field gradient (EFG) on $SmCrO_3$ compound was followed, using the $^{111}$Cd PAC probe, in the 16 K<T< 723 K temperature range. A temperature range that spans over the important transition temperatures, namely the reported ferroelectric transition ($T_{FE}$ ≈220 K), the magnetic ordering of Cr atoms sub-lattice ($T_N^{\mathrm{Cr}}$ =133 K), the spin reorientation ($T_{SR}$ =34 K) and magnetic ordering of Sm atoms sub-lattice ($T_N^{\mathrm{Sm}}$ =20 K). The $^{111m}$Cd implantation and $^{111}$Cd->$^{111}$In diffusion was followed by an annealing at high temperatures in air.

At high temperatures, T>300 K, a frequency triplet corresponding to a single EFG, $i.e.$, one probe local environment, was observed and in this temperature range no significant changes occur in the spectra when the temperature is lowered. However, below 300 K visible changes can be observed in the perturbation function ($R(t)$) data and in the corresponding Fourier transforms. In detail, a second EFG emerges and its relative abundance increases with decreasing temperature. Accordingly, the fits to the $R(t)$ experimental data were performed considering only one static EFG distribution, which was assumed to be Lorentzian-like, for T>300 K while two EFG distributions had to be considered to account for the features that emerge below that temperature.

The spectra obtained at high temperatures revealed an EFG characterized by a $V_{ZZ}^{\mathrm{Sm_1}}$ ≈76 V/m$^2$ and an asymmetry parameter $\eta \approx 0.2$ in good agreement with similar systems. The second EFG, that emerges at low temperatures, is characterized by a similar fundamental frequency but a higher asymmetry parameter $\eta \approx 0.6$.

From our data we observed that a distortion of the high temperature local environment start to develop below 300 K within the paramagnetic phase. Although our data might be compatible with the most recent reports, where polar octahedral rotations and/or cation displacements are at the origin of a polar order in the paramagnetic state, remarkably, our results point to a more subtle scenario, where locally an inhomogeneous state emerges. In this new state regular and distorted environments (most probably polar and non polar states) coexist.

[1] https://doi.org/10.1103/PhysRevB.86.214409.

[2] https://10.1209/0295-5075/107/47012.

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