Contribution ID: **86** Type: **Poster**

First-Principles Calculations of Electric Field Gradients in Hf-Based Perovskites: a Tool to Identify Transition Pathways

Friday 24 September 2021 09:40 (1 minute)

We present an ab-initio study performed by means of Density Functional Theory (DFT), group-subgroup symmetry analysis and lattice dynamics to probe the properties of the octahedral distortions, which occur during the structural phase transitions [1]. We mainly focus our study on the Sr3Hf2O7 (SHO) system, which is characterized by a high-temperature I4/mmm (S.G. 139) centrosymmetric structure and a ground-state Cmc21 (S.G. 36) ferroelectric system. We have probed potential candidate phases that may form the I4/mmm → Cmc21 transition pathways, namely Fmm2 (S.G. 42), Ccce (S.G.68), Cmca (S.G. 64) and Cmcm (S.G. 63). We found that the band gap widths increase as the symmetry of the systems decreases, with the ground-state structure presenting the largest gap width (˜5.95 eV). By probing the Partial Density of States (PDoS), we observe a direct relation regarding the tilts and rotations of the O perovskite cages as the transition occurs; these show large variations mostly of the O p-states which contribute mostly to the valence band maximum. We have also computed the macroscopic polarization and confirm that the Cmc21 phase is ferroelectric with a value of spontaneous polarization of 0.0478 C/m2. The ferroelectricity of the ground-state Cmc21 system arises due to a second order parameter related to the coupling of the rotation and tilts of the O perovskite cages together with the Sr displacements.

The measurement of the Electric Field Gradients (EFG) and asymmetric parameters, through hyperfine techniques, i.e. nuclear quadrupole resonance (NQR) or perturbed angular correlation (PAC), combined with ab-initio DFT calculations, has shown to be a valuable tool to probe the octahedral rotations of the cages during phase transition. Therefore, we have computed the EFG for each structural phase and observe variations of respective parameters which allow the possibility to ascertain for the correct phases/pathways, namely the ones connected by group-subgroup relations. In such a way, these techniques can inform about the subtle octahedral tilting and rotations, which are typically not easily accessible by long-range crystallographic techniques.

References
[1] E. Lora da Silva et al., Nanomaterials 11, 897 (2021) and references therein.

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Session Classification: Quantum Materials and Quantum Technologies (Posters)

Track Classification: Advanced Materials and Processes for Energy