Contribution ID: **86**

Type: **Poster (In person)**

Dion-Jacobson ANdNb2O7 (A = Rb,Cs) Layered Perovskite Systems: Bulk Study, Ion Exchange and Exfoliation

Wednesday 3 December 2025 18:25 (1 minute)

The $RbNdNb_2O_7$ (RNNO) and $CsNdNb_2O_7$ (CNNO) systems exhibit a sequence of phase transitions before reaching their hig temperature $P4/mmm$ aristotype phase. In RNNO, the structure evolves from a polar $I2cm$ phase at room temperature to an antipolar $Cmca$ phase at $790\,K$, and finally to a distorted $I4/mcm$ structure at $865\,K$ [1]. These transitions arise from rotations and tilts of the NbO_6 octahedra, which stabilize the polar ferroelectric phase but also induce negative thermal expansion (NTE) in the c-axis. However, such distortions may not be fully resolved by macroscopic techniques, making local-probe studies essential to clarify the microscopic mechanisms involved. To address this, we performed Raman spectroscopy and Perturbed Angular Correlation (PAC) studies across wide temperature ranges, aiming to investigate the phase behaviour and structural evolution of these materials. The PAC technique, in particular, employs a local probe that is highly sensitive to atomic-scale dynamics and allows for the measurement of the electric field gradient (EFG) at specific sites, capturing information on local symmetry and NbO_6 rotational environments.

The large size of Rb and Cs ions produces an interlayer with a large spacing and weak bonding, which opens up the possibility for exfoliation through facile and scalable methods. We performed liquid-phase sonication of the RNNO sample mixed in N-methyl-2-pyrrolidone (NMP), and collected the exfoliated products from the supernatant after centrifugation of the sonicated solution [2]. We've also used an intermediate step of protonation of the RNNO powder with HNO_3, in order to replace the Rb ions and weaken the interlayer bonds even further.

The polar behaviour via hybrid improper ferroelectricity present in RNNO, CNNO and similar compounds already make them highly interesting for energy applications, but their chemical exfoliation opens up even more possibilities in this domain due to enhanced surface area, tunable electronic properties, and integration into next-generation nanoscale devices. We'll present our results both for the bulk sample characterisation and the exfoliated material.

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Session Classification: Poster Session