Theory-guided discovery of pressure-induced transitions in the fast-ion conductor $BaSnF_4$

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Abstract

Fast-ion conductors such as $BaSnF_4$ are of significant interest for next-generation solid-state battery technologies due to their high ionic conductivity and chemical stability. However, the behavior of these materials under extreme conditions remains poorly understood, despite the relevance of pressure-induced modifications for tuning functional properties. In this study, we combine density functional theory (DFT) calculations with high-pressure experiments to investigate the structural evolution of $BaSnF_4$ up to 40 GPa [1]. As shown in Figure 1, DFT predicts two pressure-induced phase transitions: from the ambient-pressure tetragonal $P4$/nmm phase to a monoclinic $P2_1/m$-I structure at 10 GPa, and subsequently to a denser monoclinic $P2_1$/m-II phase at 32 GPa. The first transition is experimentally confirmed via angle-dispersive X-ray diffraction (carried out using a diamond-anvil cell at ALBA synchrotron), Raman spectroscopy, and electrical resistivity measurements, all performed at ambient temperature. The second transition is supported by distinct changes in high-pressure Raman modes and resistivity behavior, consistent with a further structural reorganization. These findings not only clarify the high-pressure phase diagram of $BaSnF_4$ but also shed light on the potential for pressure-tuned ionic transport in fluorostannate-based solid electrolytes.

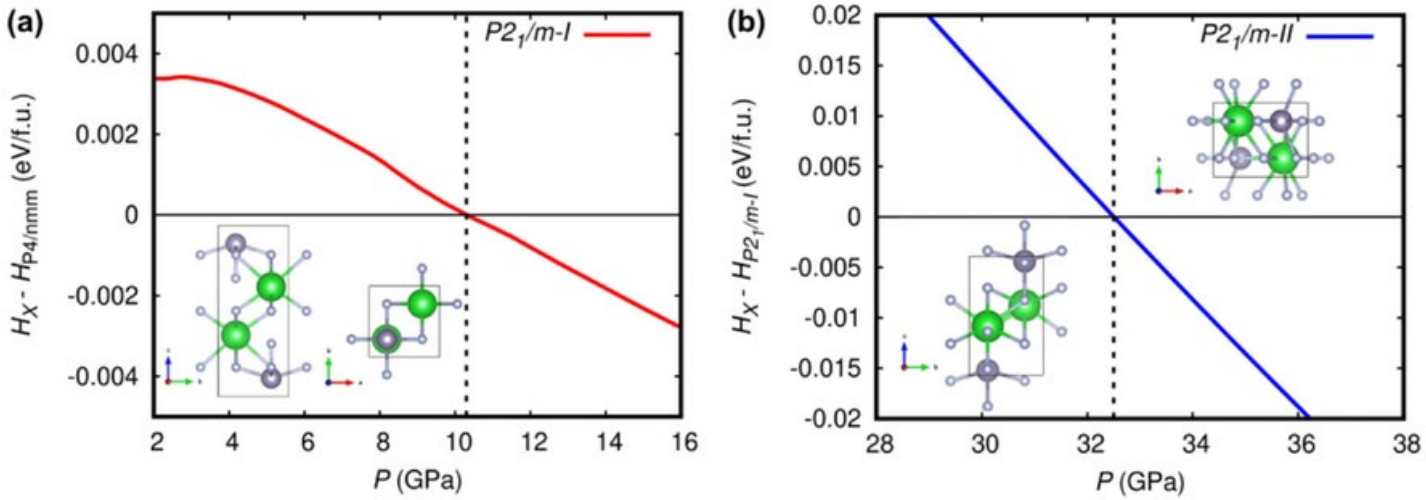

Figure 1: (a) Calculated enthalpy of the high-pressure monoclinic ($P2_1$/m-I) $BaSnF_4$structure relative to the ambient-pressure fluorite-type tetragonal structure ($P4/nmm$). (b) Calculated enthalpy of the higher-pressure monoclinic ($P2_1$/m-II) $BaSnF_4$structure relative to the lower-pressure monoclinic ($P2_1$/m-I) $BaSnF_4$structure. The overall sequence of phase transitions predicted by DFT is $P4/nmm$ (ambient pressure) → $P2_1$/m-I (>10 GPa) → $P2_1/m$-II (>32 GPa).

[1] R. Turnbull *et al.* Phys. Rev. B 112, 184104 (2025). DOI: https://doi.org/10.1103/sk37-q99z