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Perovskites in low dimensional multi-layer structure types

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This study introduces examples of structure property relationships within the multi-layered Sillen-Aurivillius family (shown in Figure) and aims to investigate the effect of chemical doping and lattice matching effects. The first example involves doping 1/3 of the n = 3 ferroelectric perovskite layers with magnetic transition metal cations in Bi₃PbTi₃O₁₄Cl [1] with charge balancing by removing Pb²⁺ for Bi³⁺. A statistical 1:2 distribution of M³⁺ and Ti⁴⁺ across all three perovskite layers was found in Bi₆Ti₂MO₁₄Cl, M = Cr³⁺, Mn³⁺, Fe³⁺, resulting in highly strained structures (enhancing the ferroelectricity compared to Bi₅PbTi₃O₁₄Cl) and pronounced spin-glass behavior below Tₘ(0) = 4.46 K. Ferroelectric transitions were observed at high temperature for each of the new compounds. Ferroelectric properties were also measured on Bi₆Ti₂FeO₁₄Cl using piezoresponse force microscopy showing hysteretic phase behavior. A new n = 2 Sillen-Aurivillius compound Bi₃Sr₂Nb₂O₁₁Br, based on Bi₃Pb₂Nb₂O₁₁Cl [2], was synthesized by simultaneously replacing Pb²⁺ with Sr²⁺ and Cl⁻ with Br⁻. Inter-layer mismatch prevented the formation of Bi₃Sr₂Nb₂O₁₁Cl and Bi₃Pb₂Nb₂O₁₁Br. Sr²⁺ doping reduces the impact of the stereochemically active 6s² lone pair found on Pb²⁺ and Bi³⁺, resulting in a stacking contraction in the lattice parameters by 1.22 % and an expansion of the a-b plane by 0.25 %, improving inter-layer compatibility with Br⁻. X-ray Absorption Near Edge Structure spectra analysis shows that the ferroelectric distortion of the B-site cation is less apparent in Bi₃Sr₂Nb₂O₁₁Br compared to Bi₃Pb₂Nb₂O₁₁Cl. Variable-temperature neutron diffraction data show no evidence for a ferroelectric distortion.


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