Anion deficiency in perovskite-based ferrites containing lone pair cations Pb2+ and Bi3+ can be accommodated by the formation of crystallographic shear (CS) planes. In this contribution, using (Pb1-\(z\)Sr\(z\))\(1-x\)Fe\(x\)O\(3-y\) as a model system, we demonstrate that the lone pair cations are not only responsible for the formation of the CS planes, but also control their orientation. The isovalent substitution of Pb2+ by stereochemically inactive Sr2+ highlights the influence of the A-cation electronic structure owing to very close ionic radii of these cations. Employing a combination of transmission electron microscopy and X-ray and neutron powder diffraction, we have systematically investigated the structure evolution of the compounds with changing P/Sr ratio. Two compositional ranges with distinct orientations of the CS planes have been identified in the system: 0.05 \(\leq z \leq 0.2\), where the CS planes orientation systematically varies but stays close to (203)p, and 0.3 \(\leq z \leq 0.45\) with the (101)p CS planes. The incommensurate structure of Pb0.792Sr0.168Fe1.040O2.529, a representative compound of the first range, has been refined from the neutron powder diffraction data (S.G. X\(\alpha\)2\(\beta\)(\(\alpha\)0\(\gamma\)), \(X = [1/2,1/2,1/2], a = 3.9512(1)\(\AA\), \(b = 3.9483(1)\(\AA\), \(c = 3.9165(1)\(\AA\), \(\beta = 93.268(2)^\circ\), \(q = 0.0879(1)\(a^*\) + 0.1276(1)\(c^*\), RF = 0.023, RP = 0.029, T = 900K). Comparing the CS structures in both compositional ranges, we demonstrate that the orientation of the CS planes is primarily governed by a specific bonding configuration of the lone pair cations with the anions.

**Keywords:** perovskites, incommensurate compounds, TEM