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Structural evolution in the hybrid improper multiferroic $\text{Ca}_{3-x}\text{Sr}_x\text{Ti}_2\text{O}_7$ F. YE, J. A. FERNANDEZ-BACA, Oak Ridge Nat. Lab, JIEMING SHENG, Renmin Univ. of China, BIN GAO, SANG-WOOK CHEONG, Rutgers Univ. — A novel microscopic mechanism has been proposed to search for ferroelectric material for realistic applications. The instability of the polar phonon mode is driven by the simultaneous condensation of two nonpolar lattice modes associated with oxygen octahedron rotation and tilt, and is responsible for the polar symmetry observed in the Ruddlesden-Popper compound. Unlike the well-studied $\text{Ca}_3\text{Mn}_2\text{O}_7$ where an antiferromagnetic order sets in at 120 K and displays strong interplay between the magnetism and structural distortion, the nonmagnetic $\text{Ca}_{3-x}\text{Sr}_x\text{Ti}_2\text{O}_7$ only exhibits a competition between two structural distortion modes. We used single crystal neutron diffraction to investigate the temperature and doping evolution of the crystal structure in a series of Sr-doped $\text{Ca}_3\text{Ti}_2\text{O}_7$ where the in-plane rotation of TiO_6 is gradually suppressed and the out-of-plane tilt persists with increasing Sr concentration for $x < 1.0$. The corresponding structural temperature from the low-T orthorhombic to the high-T tetragonal phase also increases steadily with Sr doping. Our diffraction study provides detailed characterization of spontaneous polarization that can be achieved through tilt engineering of the TiO_6 environment.

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