



Abstract

The Structural Origin of Composition-Driven Magnetic Transformation in BiFeO₃-Based Multiferroics [†]

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Perovskite-like compounds are widely known as model systems for studying the relationships between crystal structure and physical properties. Among them, magnetically-ordered ferroelectric oxides have attracted much attention in recent years. Such materials combine spin and electric dipole ordering in the same phase, thus providing the technologically important possibility to control magnetism with an electric field. While BiFeO3 is the most thoroughly studied magnetic ferroelectric compound, the properties of its solid solutions remain a matter of intensive debate. In this work we show how variation in the chemical composition of Bi_{1-x}AE_xFe_{1-x}Ti_xO₃ (AE= Ca, Sr, Ba) multiferroics affects their crystal structure and magnetic behavior. The polycrystalline samples have been studied by X-ray diffraction, neutron powder diffraction, VSM-magnetometry, electron microscopy, and scanning probe microscopy techniques. It has been found that Ca/Ti and Sr/Ti substitutions suppress the cycloidal antiferromagnetic structure specific to the parent compound, thus stabilizing a weak ferromagnetic and ferroelectric state. The Ba/Ti-doped solid solutions retain the magnetic behavior characteristic of the pure BiFeO₃. The composition-driven changes in the magnetic properties of the Bi_{1-x}AE_xFe_{1-x}Ti_xO₃ perovskites correlate with the structural evolution, confirming the existence of a tight coupling between the type of magnetic ordering and electric polarization/magnitude of oxygen octahedra tilting in these materials. The magnetostructural correlations reflect the pattern of chemical substitution-induced changes in the polarization- and oxygen octahedra rotation-related components of the Dzyaloshinskii-Moriya interaction affecting the magnetic structure. The investigation sheds light on the conditions favoring the coexistence of spontaneous magnetization and polarization in BiFeO₃-based multiferroics.

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