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Structure and magneto-electric properties of tetragonal $BiFeO_3$ with high-spin, low spin and intermediate-spin Fe^{3+} under biaxial stress by first-principles calculations

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In this paper we use first-principles calculations made under the density functional theory to study the impact of various spin configurations of Fe on lattice constants, atomic displacements, polarization and magnetization of BiFeO3 (BFO). Experimental and theoretical research revealed the existence of BFO phases with monoclinic and tetragonal symmetries in addition to the rhombohedral one corresponding to its ground state structure, with dramatic changes in lattice constants when passing from one phase to another. This rich crystal phase portrait allows the existence of several Fe spin configurations in BFO, with ab-initio-calculated total energies generally higher than that corresponding to conventional high-spin (HS) state of Fe. We have identified two different low-spin (LS) and intermediate-spin (IS) configurations of the Fe^{3+} ion in the tetragonal structure of BFO, which impact on the c/a ratio, atomic displacements and ultimately on spontaneous polarization according to the specific way the majority and minority t_{2g} and e_g orbitals are filled. A key finding is that only one of the identified LS configurations of Fe strongly reduces the tetragonality ratio of BFO, while the structures with IS-Fe maintain values of c/a similar to that of BFO with HS-Fe. Furthermore, the energetics, atomic displacements, the distribution of tetragonality ratio and magnetization were examined for different BFO geometries with mixed spin configurations of Fe^{3+}, including the effect of external constraint parameters such as biaxial stress. We observed that the tetragonality ratio and Fe displacements with respect to O along the polar axis are virtually homogeneous in superlattices with one kind of LS and IS Fe^{3+}, with a small variation in case of the other kind. Also, superlattices with IS-Fe are more stable against external biaxial stresses (Fig. 1).

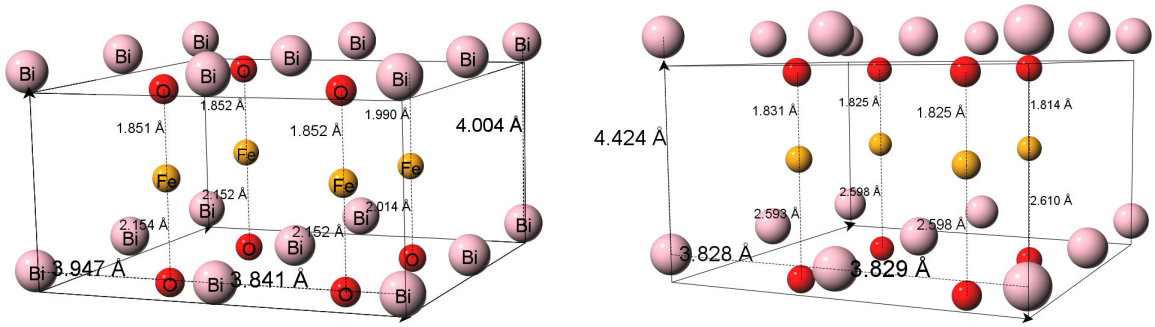

Fig. 1: Optimized structure of tetragonal BFO with 25% LS-Fe^{3+} (left) and 25% IS-Fe^{3+} (right), under 8 GPa tensile stress in (001) plane.