Local positional and spin symmetry .a150..TF11Tf5.9776005.9776138.219646.563Tm[2]TJ31Tf8.966400

We consider theoretically the paramagnetic phases of EuTiO

$_3$ that represent configurations created by two sets of microscopic degrees of freedom (mDOFs) positional symmetry breaking due to octahedral rotations and magnetic symmetry breaking due to spin disorder. The effect of these sets of mDOFs on the electronic structure and properties of the para phases is assessed by considering sufficiently large (super)cells with the required nominal global average symmetry, allowing, however, the positional and magnetic symmetries to be lowered. We find that tendencies for local symmetry breaking can be monitored by following total energy lowering in mean-fieldlike density-functional theory, without recourse for strong correlation effects. While most nominally cubic O_3 perovskites are known for their symmetry breaking due to the B-atom sublattice, the case of -electron magnetism in $EuTiO_3$ is associated with -sublattice symmetry breaking and its coupling to structural distortions. We find that (i) paramagnetic $EuTiO_3$ has an intrinsic tendency for both magnetic and positional symmetry breaking, while paramagnetic $EuTiO_3$ has only magnetic symmetry lowering and no noticeable positional symmetry lowering with respect to low-temperature antiferromagnetic tetragonal phase. (ii) Properly modeled paramagnetic tetragonal and cubic $EuTiO_3$ have a nonzero local magnetic moment on each Eu ion, consistent with the experimental observations of local magnetism in the para phases of $EuTiO_3$ significantly above the Néel temperature. Interestingly, (iii) the local positional distortion modes in the short-range ordered para phases are inherited from the long-range ordered low-temperature antiferromagnetic ground-state phase.

DOI: 10.1103/PhysRevMaterials.6.034604

I. INTRODUCTION

O_3 oxide perovskites have attracted significant research

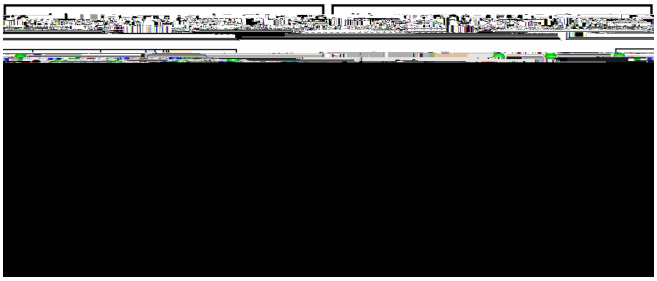

FIG. 1. The α phase of $EuTiO_3$: antiferromagnetic tetragonal insulator. (a) Crystal structure of tetragonal $EuTiO_3$. (b) The orbital and atom projected density of states (DOS) for AFM-G type tetragonal EuTiO

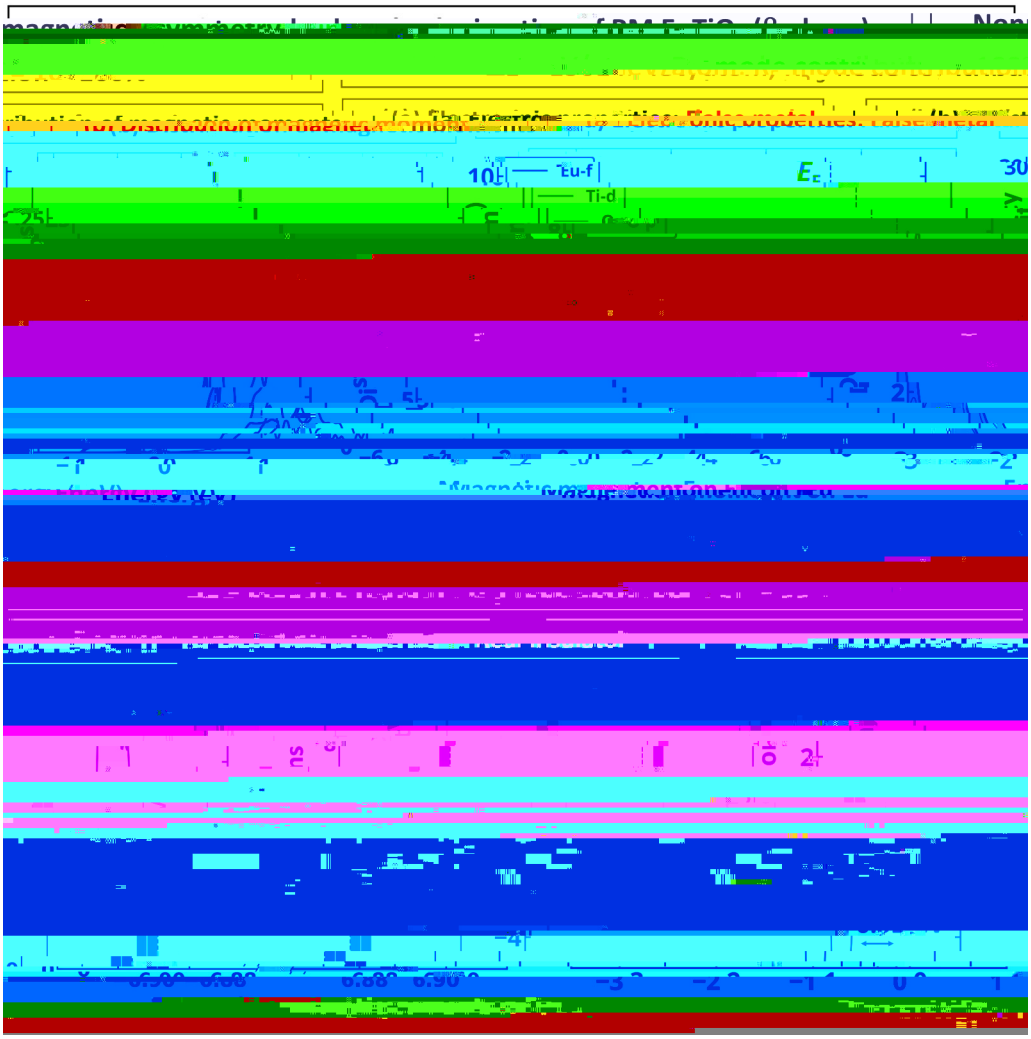

FIG. 2. The β phase of $EuTiO_3$: paramagnetic tetragonal insulator. (a), (c) Electronic and (b), (d) magnetic properties of the paramagnetic β phase of $EuTiO_3$ computed for (a), (b) nonmagnetic symmetry unbroken model and (c), (d) spin polymorphous symmetry broken model. Spin polymorphous symmetry broken model is reproduced by the nudging of 160-atom SQS supercell of tetragonal $EuTiO_3$ allowing internal

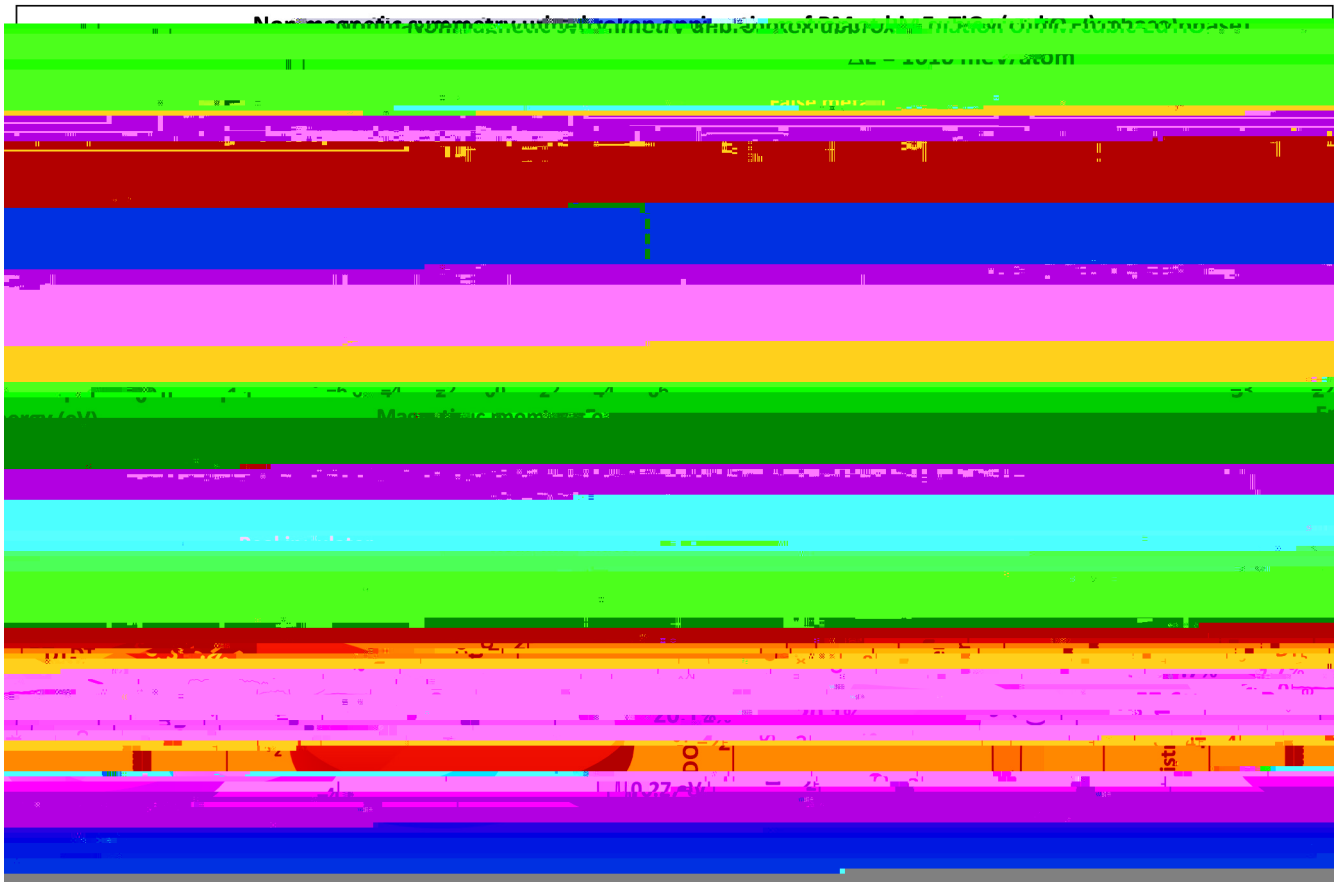

FIG. 3. The γ phase of $EuTiO_3$: paramagnetic cubic insulator. (a), (d) Electronic, (b), (e) magnetic, and (c), (f) structural properties of paramagnetic γ phase of $EuTiO_3$ computed for (a)–(c) nonmagnetic symmetry unbroken model and (d)–(f) spin polymorphous symmetry broken model. The spin polymorphous symmetry broken structure is obtained by the nudging of 160-atom SQS supercell of cubic $EuTiO_3$ allowing internal relaxation and volume optimization but keeping cubic lattice vectors. The band-gap region in (d) is shown in yellow. The dashed black line in (e) shows the magnetic moments in the α phase. Contribution of different modes to symmetry breaking in paramagnetic γ phase is computed as