Low-dimensional Atomic Multiferroics: Defects in Nonmagnetic Ferroelectric PbTiO3

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Multiferroics in nanoscale dimensions are promising for novel functional device paradigms, such as magnetoelectric memory, due to intriguing cross-coupling between coexisting ferroelectric and (anti-)ferromagnetic order parameters. However, the ferroic order is inevitably destroyed below the critical dimension of several nanometers. Here, we demonstrate a new path toward ultimately-small multiferroics while resolving the origin of ferromagnetism that emerges in nanoparticles of nonmagnetic ferroelectric PbTiO₃. Systematic exploration using hybrid Hartree-Fock density functional calculations as well as the DFT+U calculations with a theoretical Hubbard $U_{\text{cRPA}}$ derived from the constrained random phase approximation (cRPA) successfully identifies that oxygen vacancies formed at surfaces/grain boundaries induce ferromagnetism due to local non-stoichiometry and orbital symmetry breaking. The localized character of emerged magnetization allows an individual oxygen vacancy to act as an atomic-scale multiferroic element with a nonlinear magnetoelectric effect that involves rich FM-AFM-NM phase transitions in response to switching spontaneous polarization. Moreover, we also demonstrate that the local (anti-)ferromagnetism can emerge at dislocations as a line defect. Therefore, defects in ferroelectric oxides can behave as atomic-scale low-dimensional multiferroics. Engineering these multiferroic features opens a new avenue to the design of ultrahigh-density integration for atomic-scale multiferroics.