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Multiferroic phase boundaries and properties of BiFeO₃-based solid solutions¹

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The presence of morphotropic phase boundary in ferroelectric solid solutions (FE-MPB) is known to be crucial for high piezoelectricity. Similarly, magnetic MPB (M-MPB) is found in a few ferromagnets and is proved to be greatly beneficial to the magnetostrictive response. One naturally asks if in multiferroics that exhibit both ferroelectricity and (ferro-/antiferro-)magnetism, the FE-MPB and M-MPB could exist simultaneously, and if so, what the relation between these two kinds of MPB would be, and how they would affect the properties. In this paper, we report the studies of ferroelectric and magnetic double morphotropic phase boundaries in BiFeO₃-based multiferroics. The effects of dysprosium ion on the structure and local polar domains of the BiFeO₃-based systems were investigated firstly in the Dy-substituted solid solutions of 0.66Bi_{1-x}Dy_xFeO₃-0.34PbTiO₃. It is found that the substitution of Dy affects the structural symmetry and phase component of the multiferroic solid solution, and thereby enhances its ferroelectric order. A (weak) ferromagnetic state is induced at room temperature for the rhombohedral compositions with $x \geq 0.10$. The introduction of Dy into 0.66BiFeO₃-0.34PbTiO₃ leads to the breaking of its antiferromagnetic order below Néel temperature and thereby the formation of (weak) ferromagnetic ordering at room temperature when the substitution rate exceeds a critical value ($x \geq 0.10$), making the 0.66Bi_{1-x}Dy_xFeO₃-0.34PbTiO₃ system one of rare room-temperature ferromagnetic and ferroelectric materials, i.e. a true multiferroic. A comprehensive ferroelectric-magnetic phase diagram is established in terms of temperature and composition, which depicts the coexistence of a FE-MPB and a FM-MPB. These two kinds of MPBs overlap with each other. Such unusual coincidence of both magnetic MPB and ferroelectric MPB, the so-called double MPB, points to new kinds of couplings among the multiple physical quantities so that such effects as magnetoelectricity, magnetostrictive and piezoelectricity, could be enhanced near the overlapping MPB region. In addition, we find an unusual magnetic pole inversion behavior in multiferroic (1-x)BiFeO₃-xDyFeO₃ solid solution, which can be tuned by varying the concentration of the magnetic ion Dy³⁺ in the solid solution in a wide composition range.

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