強誘電体キャパシタにおける酸素空孔分布の制御

Ferroelectric capacitors with a controlled oxygen-vacancy distribution

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Spontaneous polarization (P_s) in ferroelectrics offers opportunities to develop sensors, actuators, medical transducers, and non-volatile memories. Recently, ferroelectric tunnel junctions have attracted much attention owing to their potential applications in non-destructive readout memories[1]. In all these applications, the control of domain structures and polarization states by applying external fields are crucial to achieving desirable properties. Even though ferroelectric bulk has the insulating nature, charged domain walls act as an electrically conductive interface, which leads to possibilities for domain-wall memories[2]. Moreover, utilizing a high mobility of defects enables to create a p–n junction that can be erased and inverted by electric fields[3].

Meanwhile, an accumulation of defects at interfaces deteriorates the behaviour of polarization-switching dynamics[4,5]. For the charged domain walls stabilized by oxygen vacancies, the defect kinetics dominate the process of polarization switching that is accompanied by a vacancy redistribution. The strong interaction with the vacancies gives rise to the clamping of domain walls and eventually causes imprint, retention loss, and fatigue of polarization states, preventing the widespread use of ferroelectric-based memories

Controlling defects in materials provides an extra degree of freedom not only for enhancing properties but also for introducing additional functionalities. In ferroelectric oxides, an accumulation of point defects at boundaries often deteriorates a polarization-switching capability, but on the one hand, delivers interface-driven phenomena. Despite several decades of intensive research, it remains challenging to manipulate oxygen vacancies to result in a desirable defect structure. Here, we report a practical route to designing oxygen-vacancy distributions by utilizing the interaction with transition-metal dopants. Our thin-film experiments combined with *ab-initio* theoretical calculations for $BiFeO_3$ demonstrate that isovalent dopants such as Mn^{3+} with a partly or fully electron-occupied e_g state can trap oxygen vacancies, leading to a complete polarization switching. Our approach to controlling point defects will offer an ability to harness the full potential of switchable polarization and also of domain-wall functions in ferroelectric devices.

1. Garcia, V. *et al.* Giant tunnel electroresistance for non-destructive readout of ferroelectric states. *Nature* **460**, 81–84 (2009).
2. Seidel, J. *et al.* Conduction at domain walls in oxide multiferroics. *Nat. Mater.* **8**, 229–234 (2009).
3. Yang, C.-H. *et al.* Electric modulation of conduction in multiferroic Ca-doped $BiFeO_3$ films. *Nat. Mater.* **8**, 485–493 (2009).
4. de Araujo, C. A. P. A.-P. *et al.* Fatigue-free ferroelectric capacitors with platinum electrodes. *Nature* **374**, 627–629 (1995).
5. Han, M.-G. *et al.* Interface-induced nonswitchable domains in ferroelectric thin films. *Nat. Commun.* **5**, 4693 (2014).