Enhanced dielectric permittivity utilizing ferrorestorable polarization

Yuji Noguchi1* and Hiroki Matsuo 2

[1] Division of Information and Energy, Faculty of Advanced Science and Technology, Kumamoto University, 2-39-1, Kurokami, Chuo-ku, Kumamoto 860-8555, Japan

[2]International Research Organization for Advanced Science & Technology (IROAST), Kumamoto University, 2-39-1, Kurokami, Chuo-ku, Kumamoto 860-8555, Japan

*Corresponding author, e-mail: ynoguchi@cs.kumamoto-u.ac.jp

A self-powered system with a long lifetime offers an opportunity to develop a next-generation, standalone Internet of Things. Ceramic capacitors are promising candidates for energy storage components because of their stability and fast charge/discharge capability. Even for state-of-the-art capacitors, the energy density needs to be increased markedly. Improving breakdown electric fields provides a potential solution, but operations at such high fields relying on unchanged dielectric permittivity sacrifice the lifetime to some degree. Here, we report a ferrorestorable polarization engineering capable of enhancing effective permittivity over twice. Our experiments and *ab initio* calculations demonstrate that a defect dipole composed of 3d transition metal acceptors such as Cu^{3+} and oxygen vacancy in a prototypical ferroelectric $BaTiO_3$ ceramic is coupled with spontaneous polarization[1)]. The resultant ferrorestorable polarization delivers an extraordinarily large effective relative permittivity beyond 7,000 with a high energy efficiency up to 89 %[2)]. Our work paves the way to realizing efficient ceramic capacitors for self-powered applications.

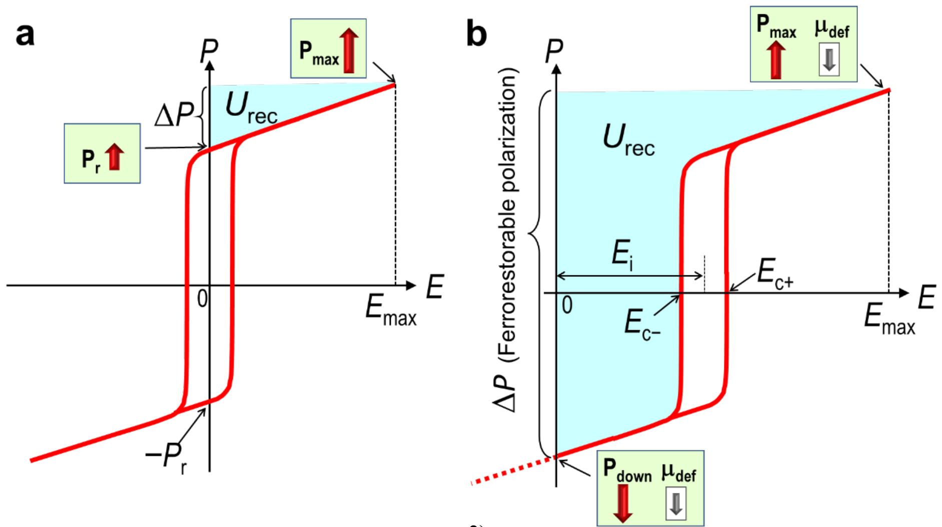

Fig. 1 | Ferrorestorable polarization[2)]. a, Typical *P-E* loop of ferroelectrics (pristine). **b,** Shifted *P-E* loop with an internal electric field (E_i) caused by the ground-state configuration of $\boldsymbol{\mu}_{def} \parallel \mathbf{P}_s$ (controlled). The controlled sample has a large U_{rec} as a result of ΔP, which is termed ferrorestorable polarization. The interaction between $\boldsymbol{\mu}_{def}$ and $\mathbf{P}_s$ stabilizes the downwards polarization (P_{down}) at zero field, i.e., $P_0 = P_{down}$, because the *P-E* loop shifts to a positive field by the magnitude of E_i. E_i is defined as the average of E_{c+} and E_{c-}, that is, $E_i = (E_{c+} + E_{c-})/2$, where E_{c+} and E_{c-} are the electric fields at the extreme polarization switching currents in the positive and negative field sweeps, respectively.

1) Y. Noguchi, Y. Taniguchi, R. Inoue and M. Miyayama, *Nat. Commun*. **11**, 966 (2020).
2) H. Matsuo, M. Utsunomiya and Y. Noguchi, *NPG Asia Mater*. **14** [1], 80 (2022).