

Electronic Supporting Information

Enhanced Colossal Permittivity in Mono-doped BaTiO₃ via Particle Hydroxylation-Induced Defect Dipoles

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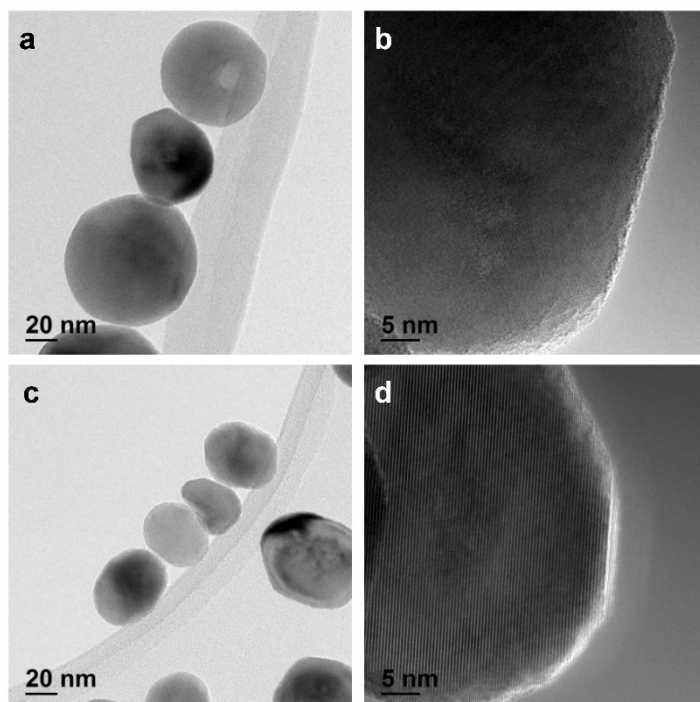


Fig. S1 Bright field and HRTEM images of as-synthesized (a, b) PHO and (c, d) FH0. Pore-free particles were synthesized with high crystallinity irrespective of the OH- concentration.

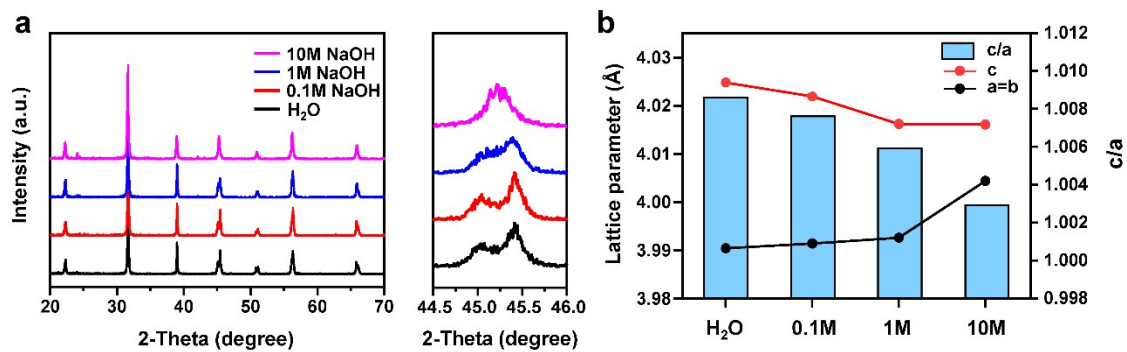


Fig. S2 a) XRD patterns and b) calculated lattice parameters of hydrothermally synthesized BaTiO₃ NPs at various pH conditions.

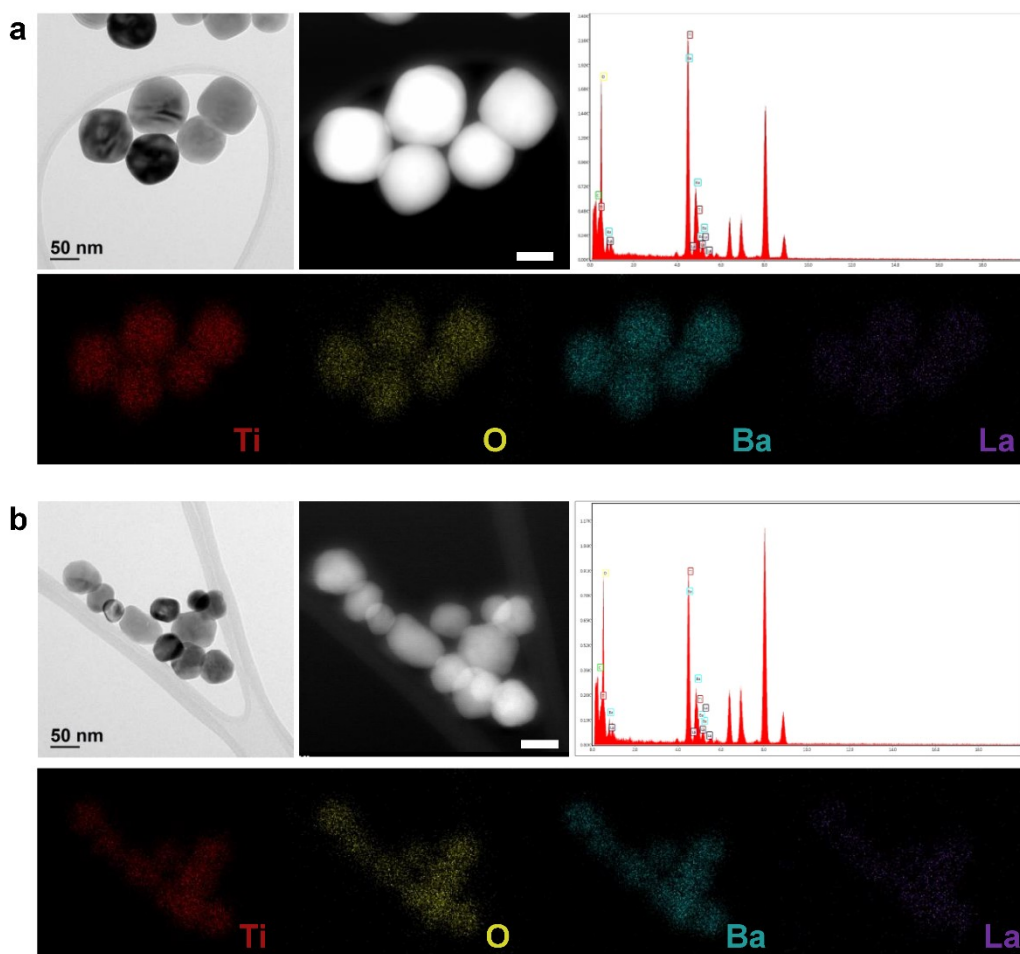


Fig. S3 TEM, STEM, and EDS results of a) PH1 and b) FH1 nanoparticles. Elemental mapping results for Ti, O, Ba, and La indicate that La^{3+} was incorporated under both synthesis conditions.

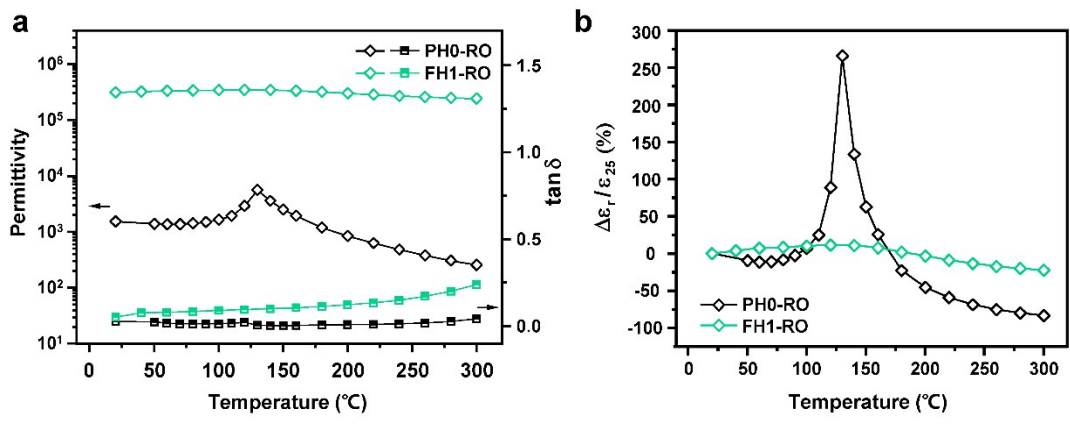


Fig. S4 (a) Temperature-dependence of dielectric properties and (b) temperature-coefficients of capacitance values of PH0-RO and FH1-RO ceramics.

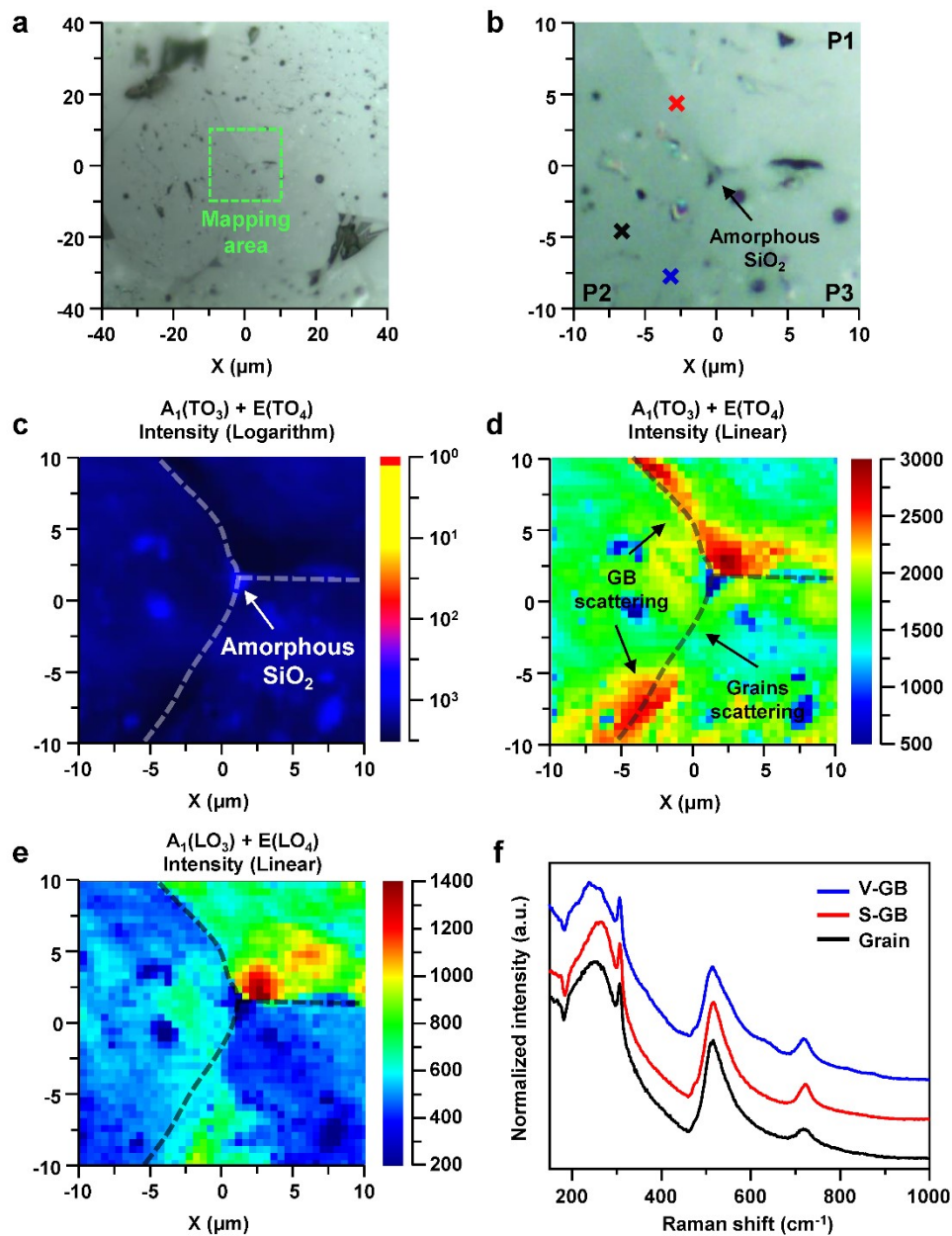


Fig. S5 Microstructural effects on Raman scattering. (a, b) Low and high magnification of optical image of the PHO-C polished surface. The sintering additive (amorphous SiO_2) was observed at the triple junction. (c, d) Raman maps of the absolute intensity at $A_1(\text{TO}_3) + E(\text{TO}_4)$ in logarithmic and linear scales. The interface scattering varies with the scanning spot of the grain boundary. (e) Raman maps of the absolute intensity near $A_1(\text{LO}_3) + E(\text{LO}_4)$. The grains P2 and P3 exhibit low scattering intensity, indicating that their polar axes are tilted with respect to incident wavevector. (f) Raman spectra obtained at the grain and grain boundaries in PHO-C. Scanning spots are marked with cross sign (x) in the OM image.

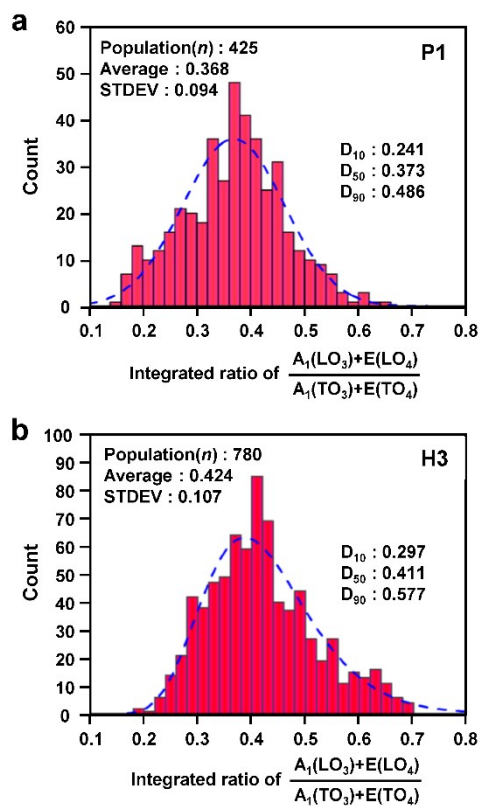


Fig. S6 Histogram of the integrated ratio between $A_1(LO_3)+E(LO_4)$ and $A_1(TO_3)+E(TO_4)$ in a) P1 and b) H3. D_{10} , D_{50} , and D_{90} denote the values of the integrated ratio at the 10%, 50%, and 90% of the overall population, respectively.