

## Piezoelectricity of Strongly Polarized Ferroelectrics in Prototropic Organic Crystals

Sachio Horiuchi,<sup>a</sup> Jun'ya Tsutsumi,<sup>a</sup> Kensuke Kobayashi,<sup>b</sup> Reiji Kumai<sup>b</sup> and Shoji Ishibashi<sup>c</sup>

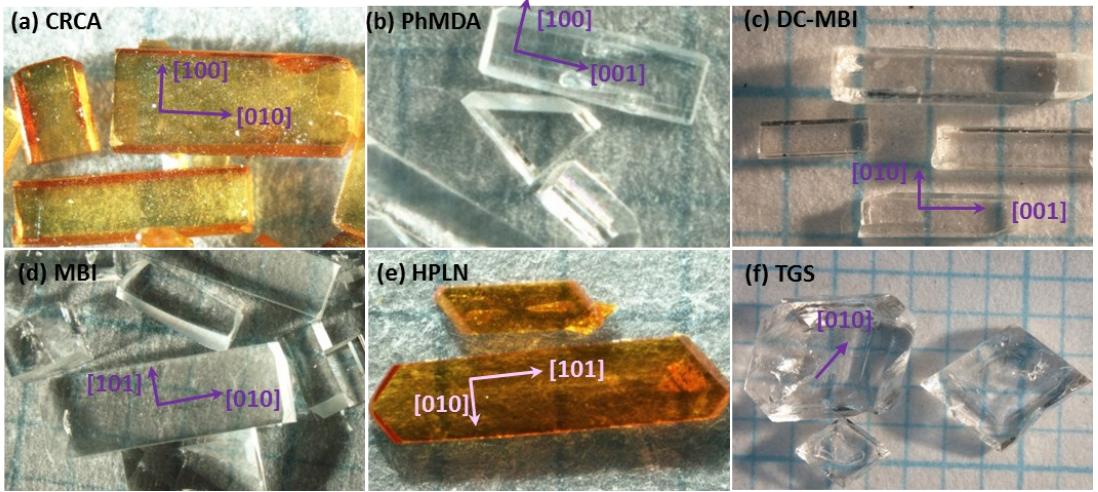
<sup>a</sup>Flexible Electronics Research Center (FLEC), National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Ibaraki 305-8565, Japan

<sup>b</sup>Condensed Matter Research Center (CMRC) and Photon Factory, Institute of Materials Structure Science, High Energy Accelerator Research Organization (KEK), Tsukuba, Ibaraki 305-0801, Japan

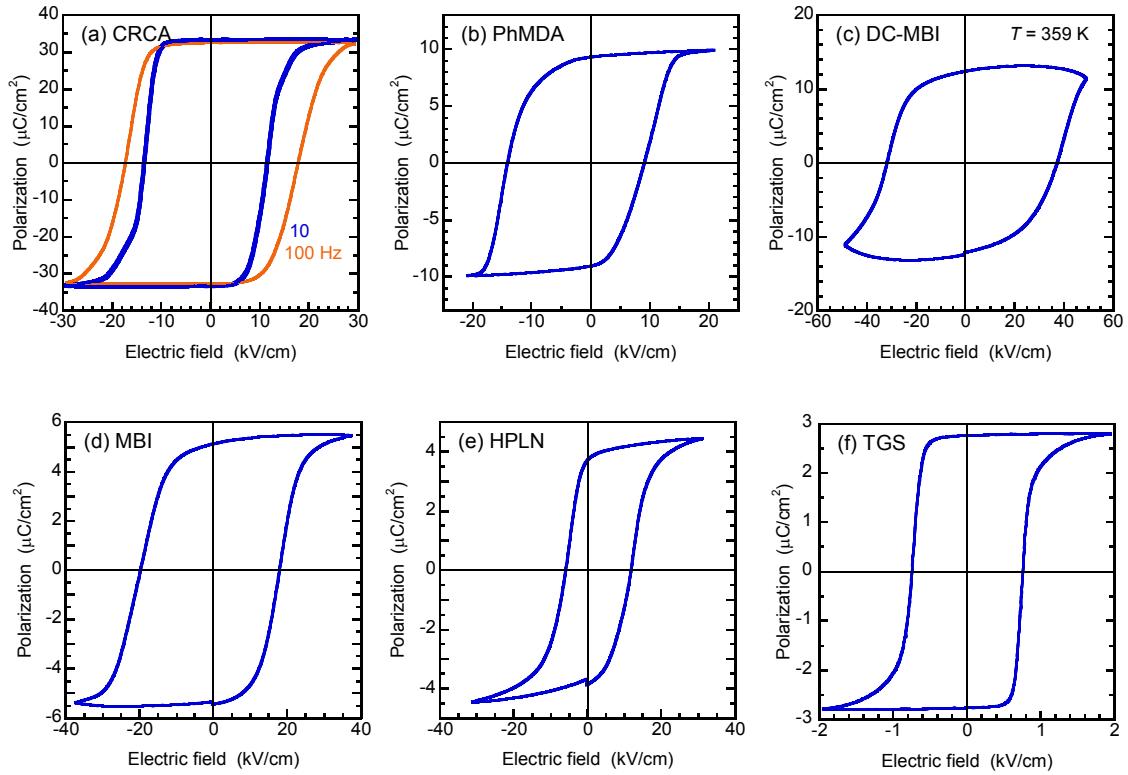
<sup>c</sup>Research Center for Computational Design of Advanced Functional Materials (CD-FMat), National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Ibaraki 305-8568, Japan

## Electronic Supplementary Information

## Experimental details



**Figure S1.** Photographs of single crystals. (a) CRCA, (b) PhMDA, (c) DC-MBI, (d) MBI, (e) HPLN, and (f) TGS.



**Figure S2.** The electric polarization versus electric field hysteresis loops of the ferroelectric crystals examined for electric poling prior to the measurements of the piezoelectric coefficient by the direct stress–charge method. (a) CRCA (with  $E \parallel [001]$  configuration), (b) PhMDA ( $E \parallel [001]$ ), (c) DC-MBI ( $E \parallel [001]$ ), (d) MBI ( $E \parallel [101]$ ), (e) HPLN ( $E \perp [101]$  plane), and (f) TGS ( $E \parallel [010]$ ). The magnitude of remanent polarization is optimized up to the fully polarized level.