

Supplementary Information

Table S1 Electrical resistance between Pt 1 and Pt 2 before and after poling under various illumination intensities.

Illumination intensity	Resistance before poling	Resistance after poling
0 mW/cm² (dark)	3.1 <i>MΩ</i>	2.7 <i>MΩ</i>
200 mW/cm²	2.7 <i>MΩ</i>	2.2 <i>MΩ</i>
400 mW/cm²	2.5 <i>MΩ</i>	2.0 <i>MΩ</i>
600 mW/cm²	2.3 <i>MΩ</i>	1.8 <i>MΩ</i>
800 mW/cm²	2.1 <i>MΩ</i>	1.6 <i>MΩ</i>

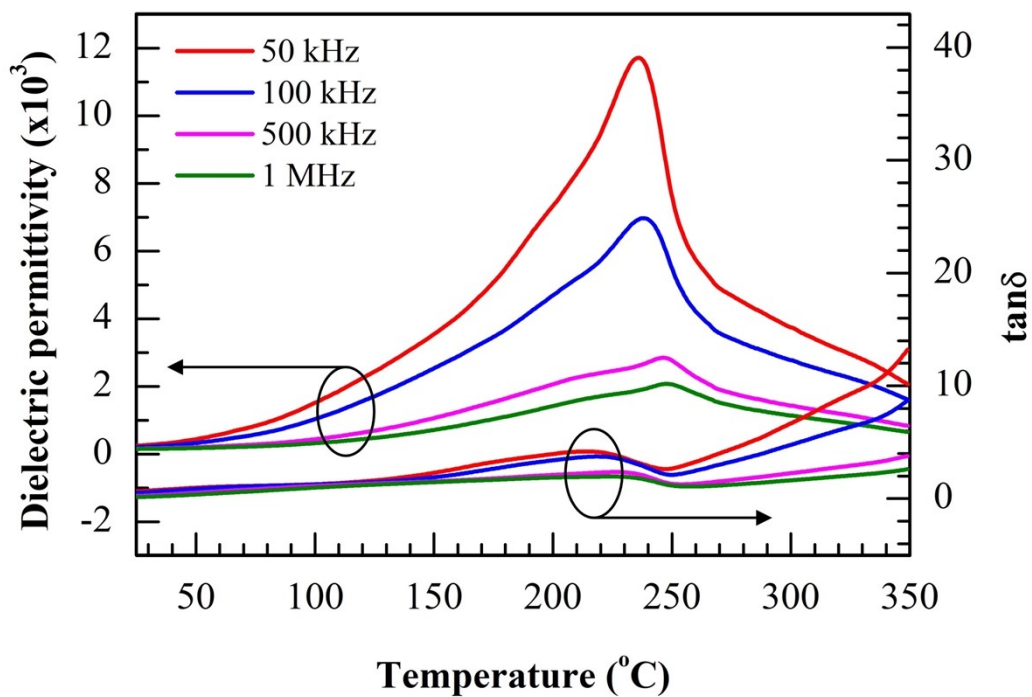


Figure S1 Curves of the dielectric permittivity and dielectric loss ($\tan\delta$) as functions of temperature.

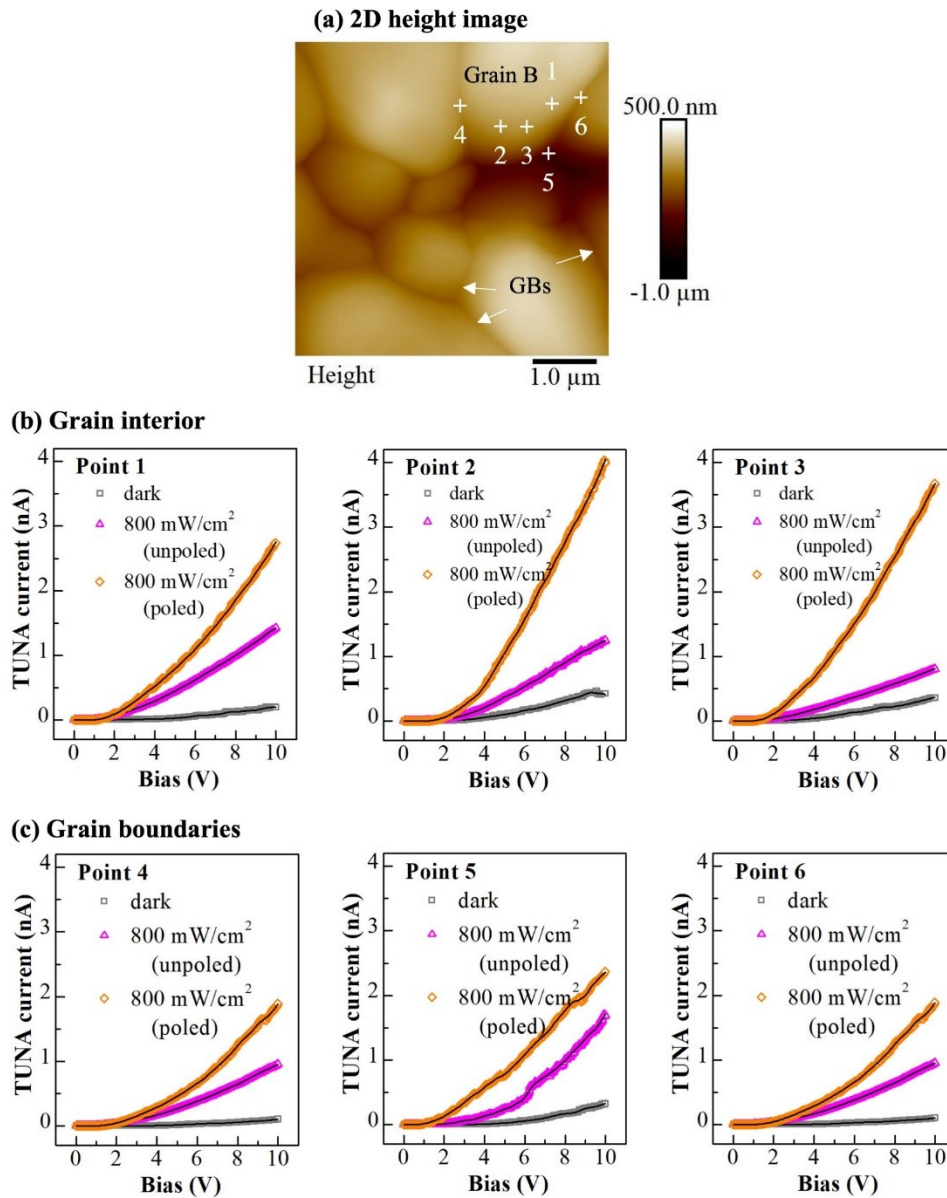


Figure S2 Curves of the photocurrent vs. bias voltage acquired at various points in the grain interior and GBs of grain B at dark and 800-mW/cm² illumination before and after *E*-field poling.

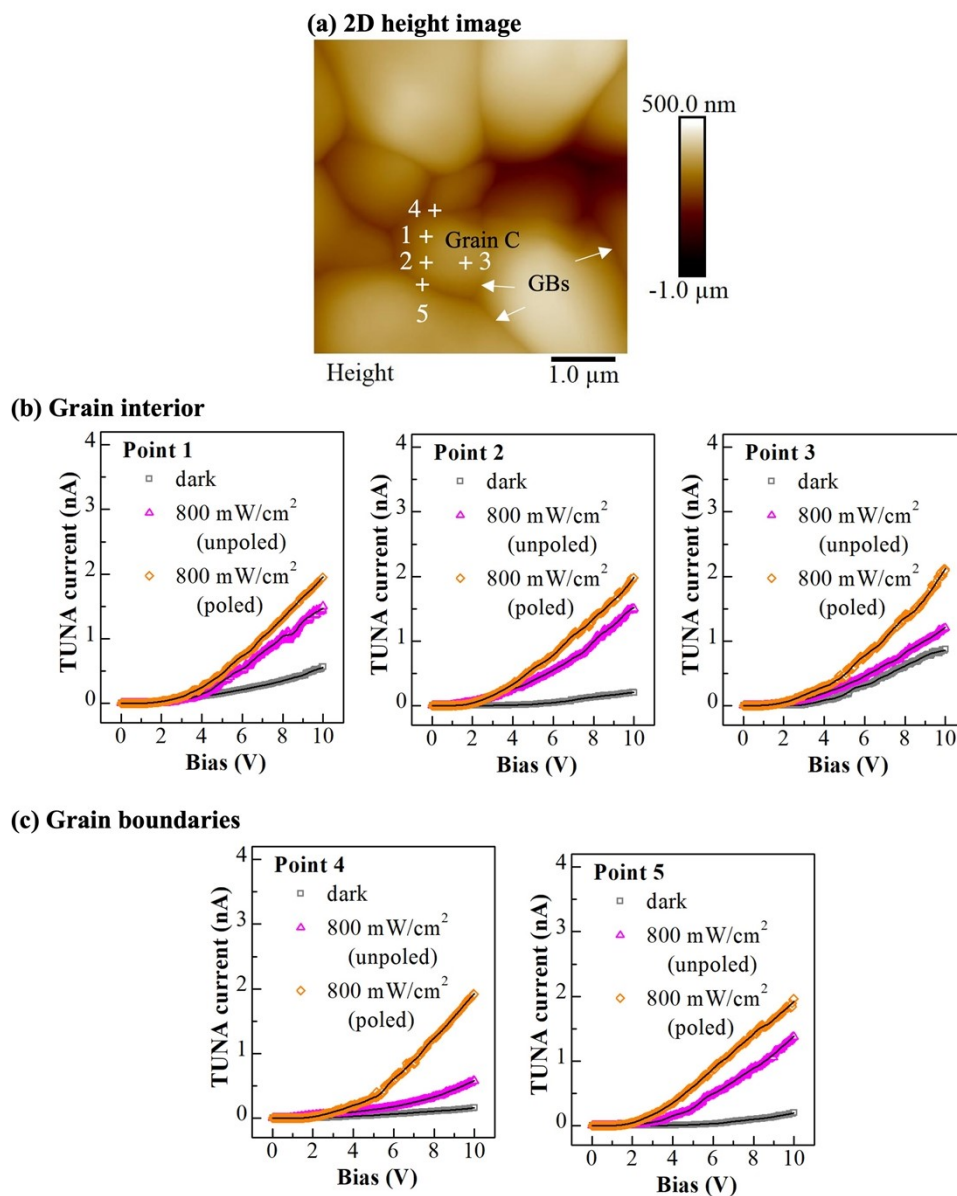


Figure S3 Curves of the photocurrent vs. bias voltage acquired at various points in the grain interior and GBs of grain C at dark and 800-mW/cm² illumination before and after *E*-field poling.

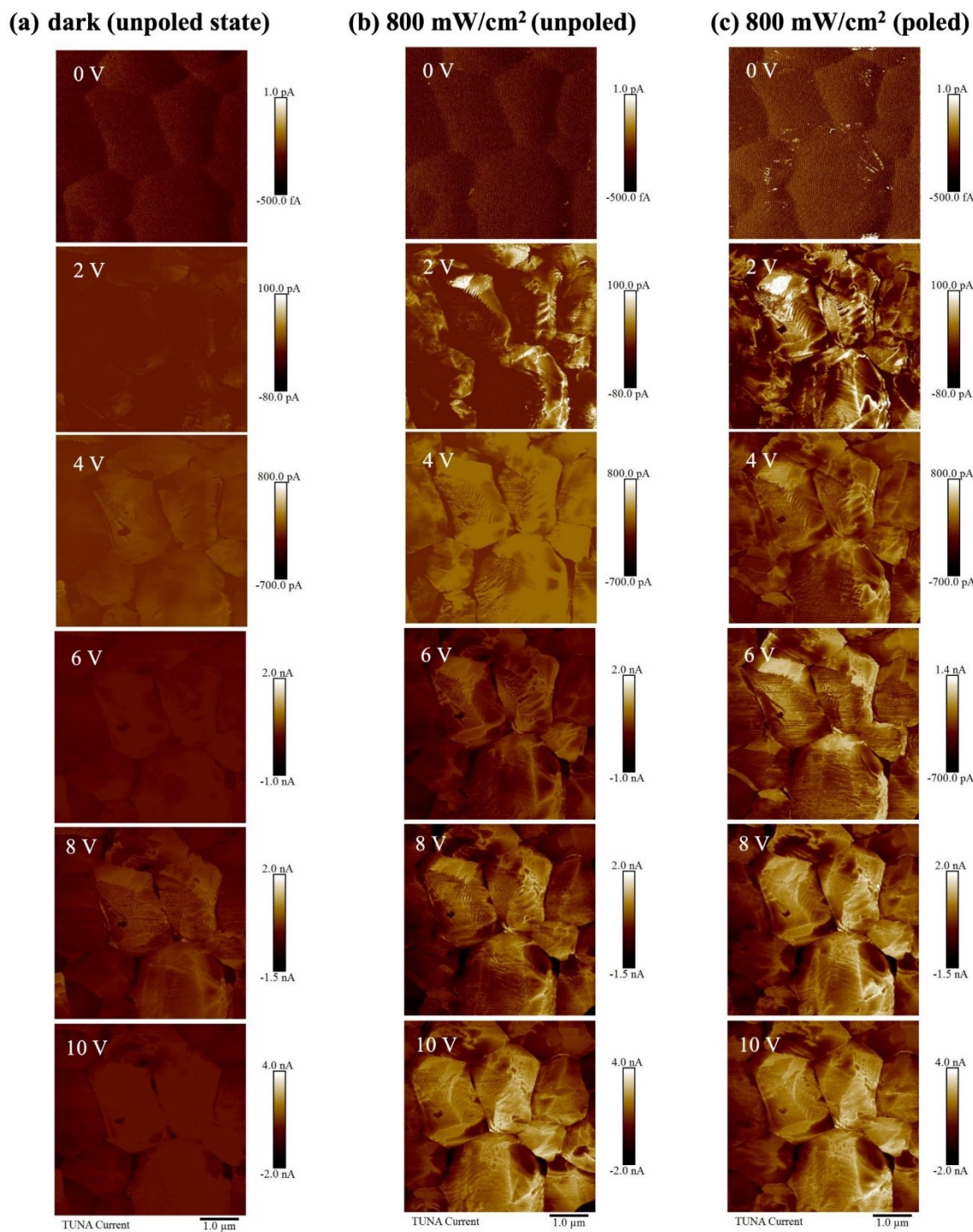


Figure S4 Mapping of photocurrent-voltage characteristics before poling at (a) dark and (b) 800-mW/cm² illumination and after poling (c) 800-mW/cm² illumination.

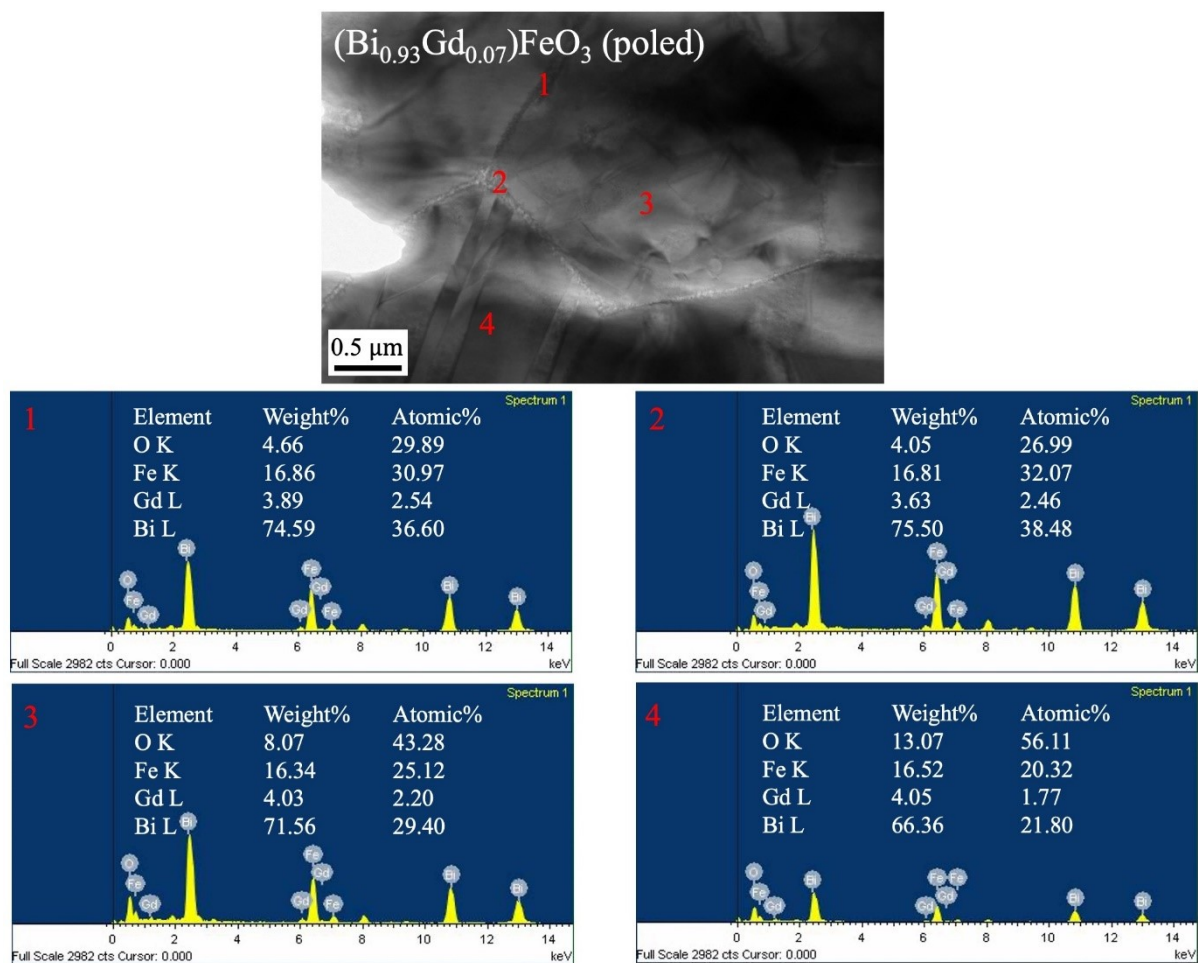


Figure S5 TEM bright-field image of poled $(\text{Bi}_{0.93}\text{Gd}_{0.07})\text{FeO}_3$ ceramic and the corresponding EDS spectra acquired from various points on the grain matrix.