

Supporting Information

Self-organized ferroelectric domains controlled by a constant bias from AFM tip

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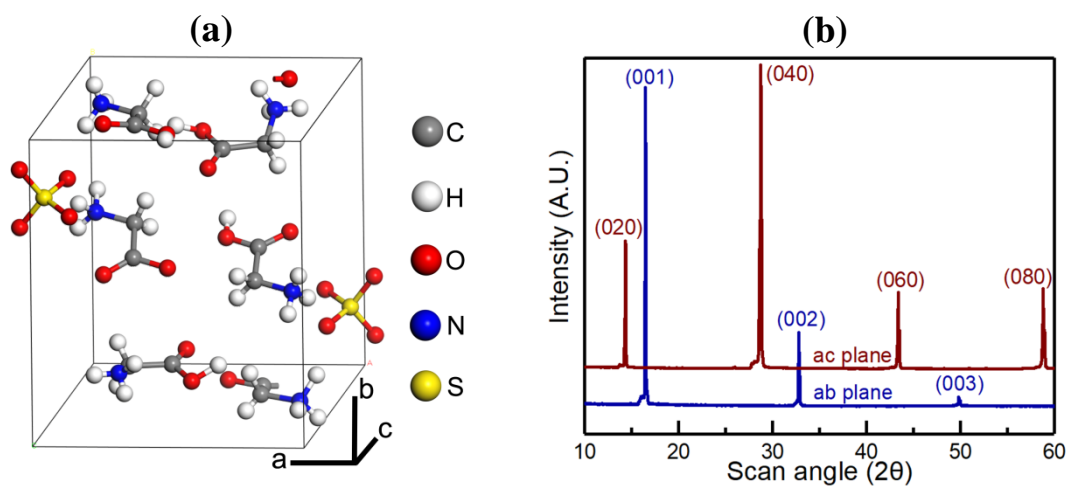


Figure S1. (a) Crystal structure of TGS crystal. (b) XRD patterns of the (010) and (001) TGS crystals.

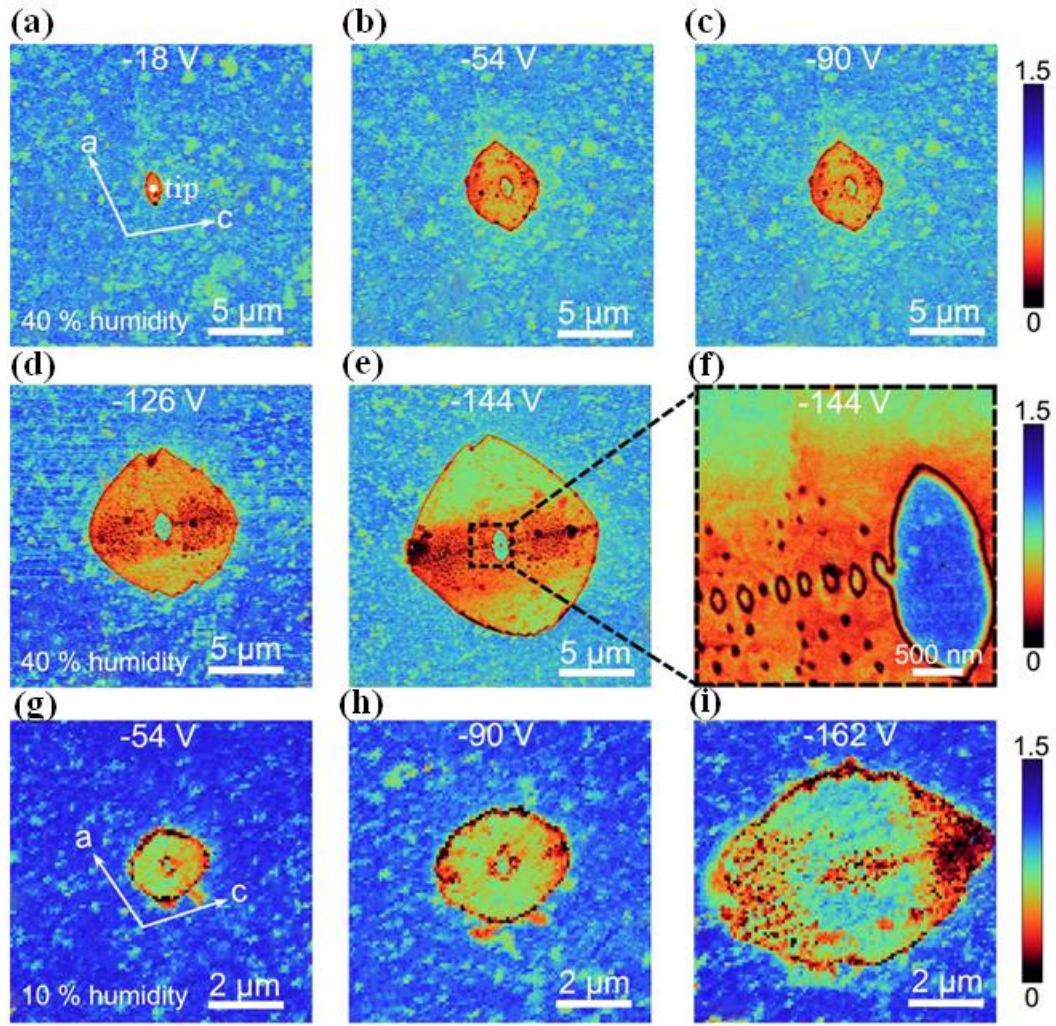


Figure S2. (a-e) Out-of-plane amplitude images after the (010) TGS crystal with a downward domain was polarized by a conductive PFM tip with (a) -18 V, (b) -54 V, (c) -90 V, (d) -126 V and (e) -144 V for 10 s and then 0 V for 1 minute in the 40% humidity ambience, respectively. (f) Amplified amplitude image of a region in (e). (g-i) Out-of-plane amplitude images after the (010) TGS center was polarized by a conductive PFM tip with (g) -54 V, (h) -90 V and (i) -162 V for 10 s in the 10% humidity ambience, respectively.

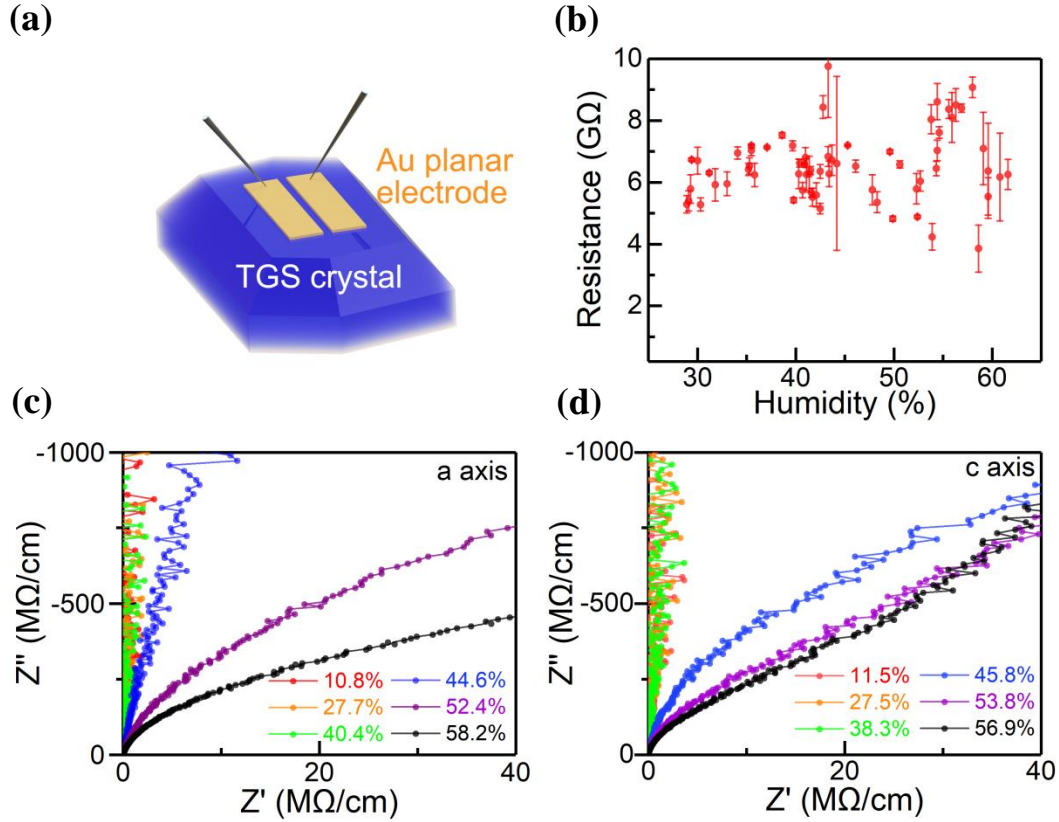


Figure S3. Resistance and impedance of (010) TGS crystal. (a) Scheme of two planar electrodes at the surface for electrical measurements. (b) Dependence of resistance on humidity. (c,d) Dependence of impedance (c) along [100] axis and (d) [001] axis on humidity, respectively.

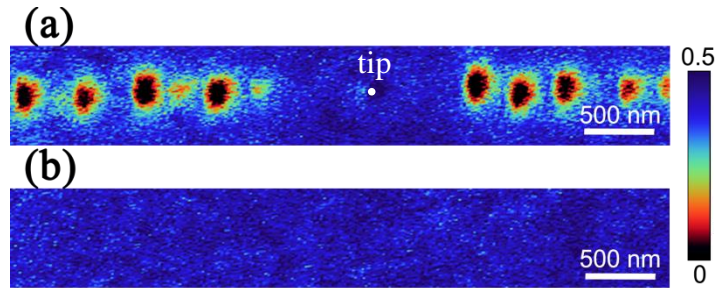


Figure S4. Amplified amplitude images after the granular domains along the tip-*c* line were introduced by a -220 V tip bias for 10 s and then they were relaxed for (a) 330 minutes and (b) 360 minutes, respectively, where previous images were already shown in Figure 3.

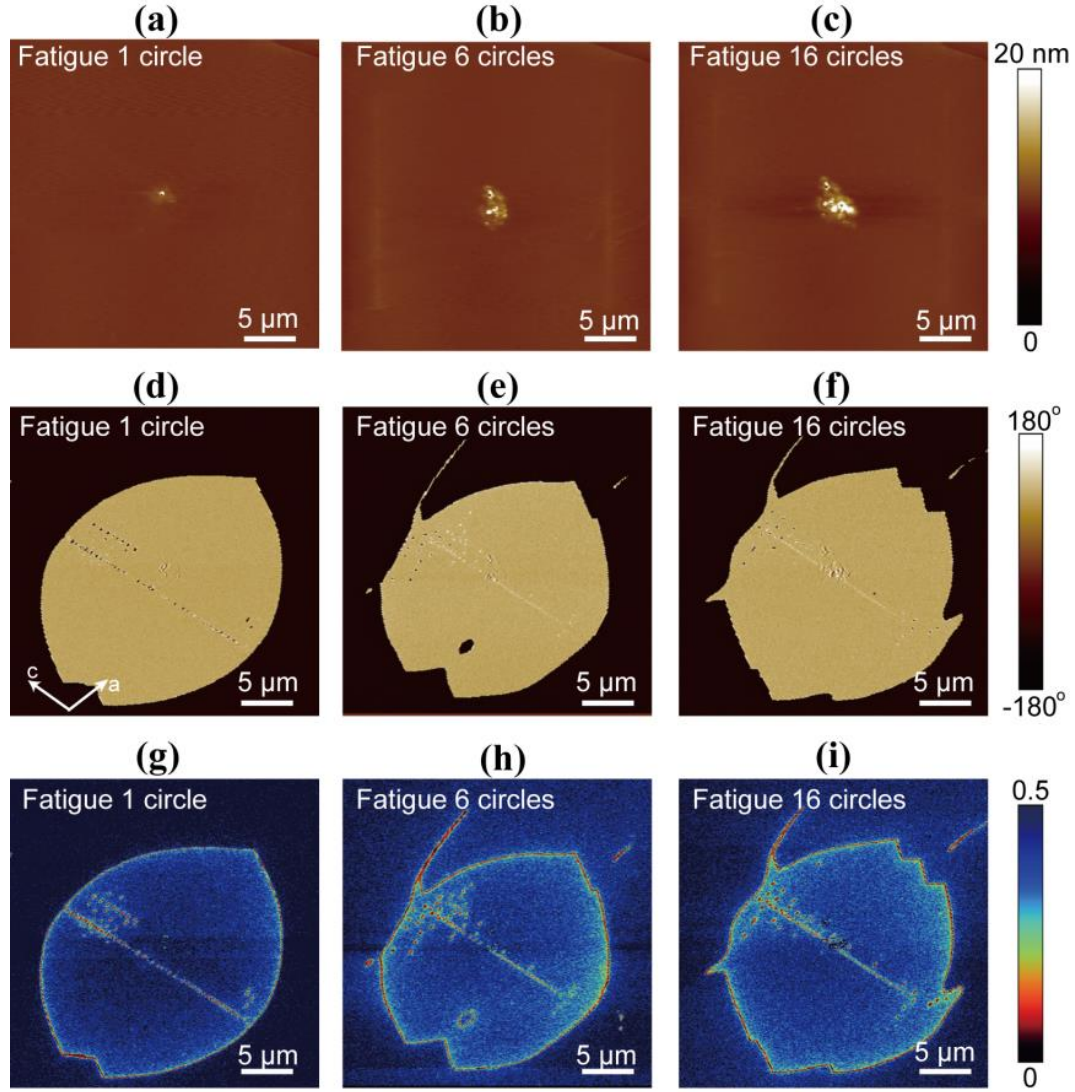


Figure S5. Repetitive experiments of domains triggered by tip bias. (a, b, c) Morphology, (d, e, f) PFM phase and (g, h, i) amplitude images after ± 180 V tip biases were applied upon its center for 1 circle, 6 circles and 16 circles, respectively.

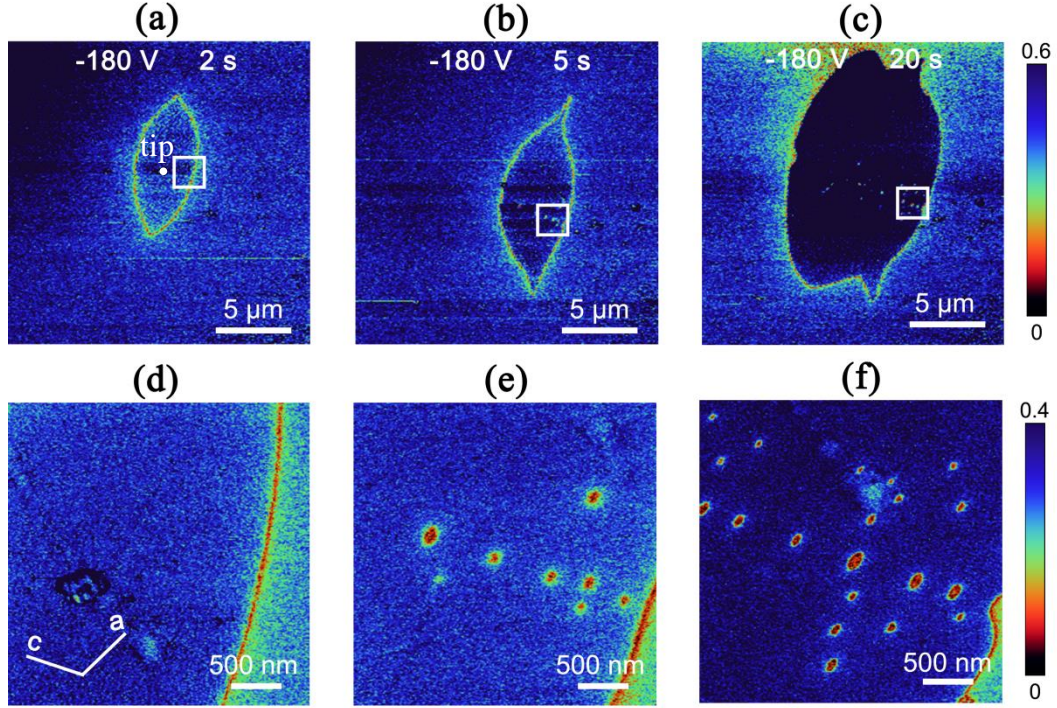


Figure S6. PFM amplitude images of the (010) TGS measured after its center were polarized by a -180 V tip bias for (a, d) 2 s, (b,e) 5 s and (c,f) 20 s, where (d-f) are the amplified images of the square regions in (a-c). Granular domains along c-axis are observed in (e) and (f) however they do not appear in (d), which proves that the 2 s tip bias is too short to induce polarization backswitching along tip-c line.

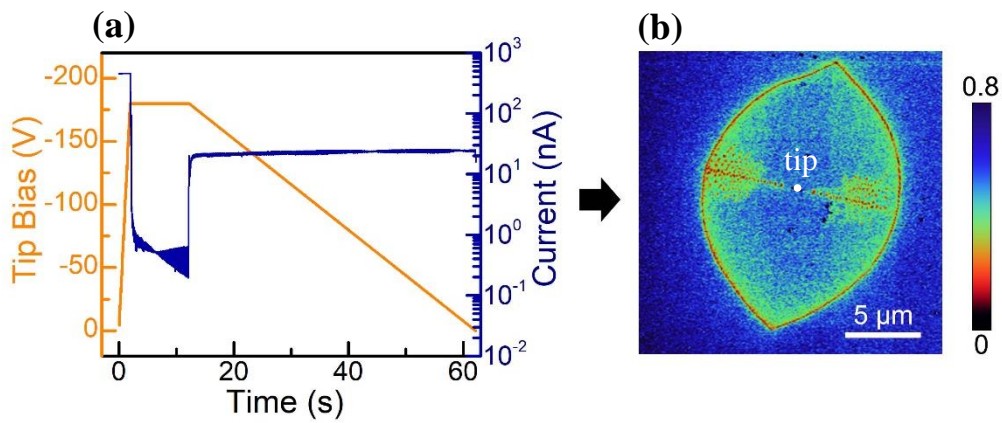


Figure S7. A ladder-type tip bias introducing depolarization along the tip-*c* line. (a) Tip bias (yellow) and the corresponding current through the tip (blue). (b) Out-of-plane PFM amplitude image.

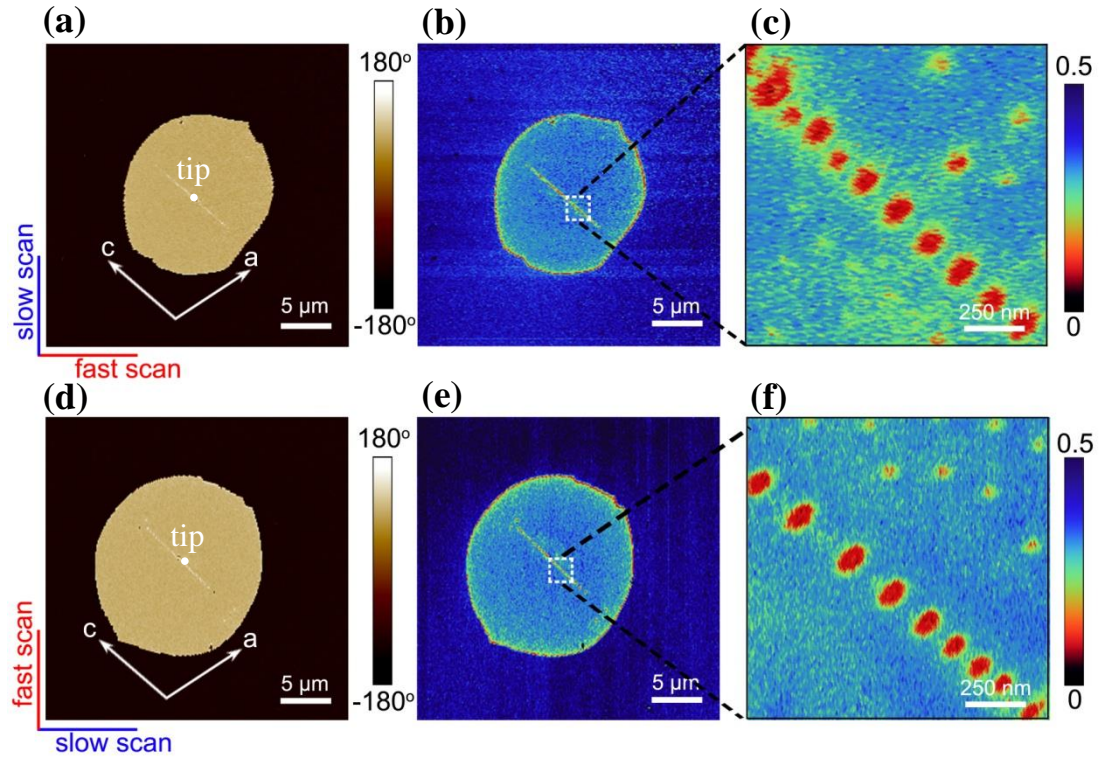


Figure S8. PFM images of the (010) TGS crystal (a,b,c) before and (d,e,f) after it rotates 90° on [010] axis. Out-of-plane (a,d) phase and (b,c,e,f) amplitude images after -162 V tip bias was applied on TGS (010) surface for 10 s and then tip moved away from the TGS surface, where the tip fast traces along red line, moves a small step along blue line, retraces along red line, and these processes are repeated until 512 lines are completed to form a high-resolution images.

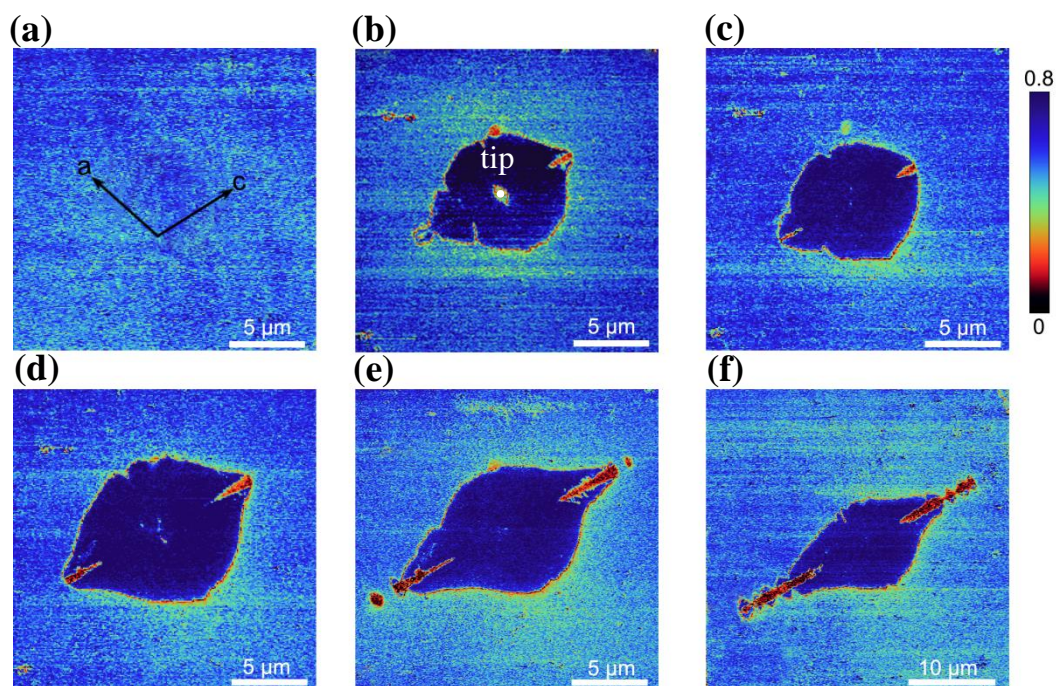


Figure S9. Out-of-plane amplitude images of (010) TGS in 40 % humidity. (a) Original amplitude images of the (010) TGS. Images after the center position was polarized (b) by a tip with 180 V for 5 s and then 0 V for 1 minutes or by a tip with 180 V for (c) 5 s, (d) 10 s, (e) 20 s and (f) 50 s and then withdrawing from the TGS surface.

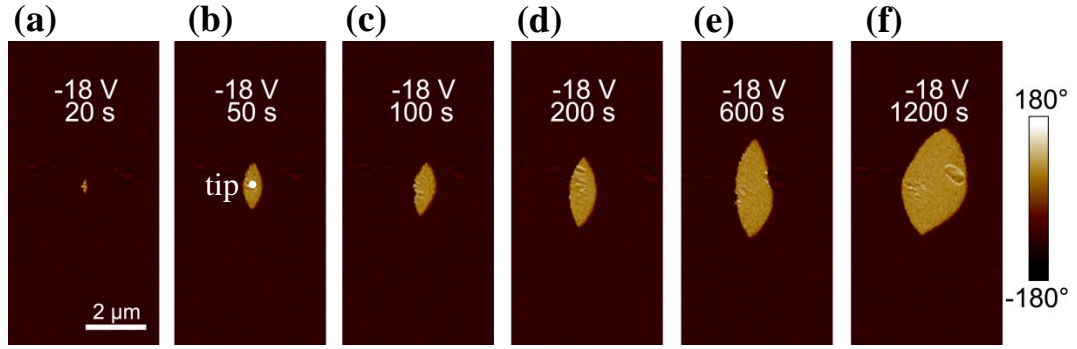


Figure S10. (a-f) Out-of-plane phase images after the (010) TGS crystal with a downward domain was polarized by a PFM tip with a -18V bias for (a) 20 s, (b) 50s, (c) 100s, (d) 200 s, (e) 600 s and (f) 1200 s, respectively.

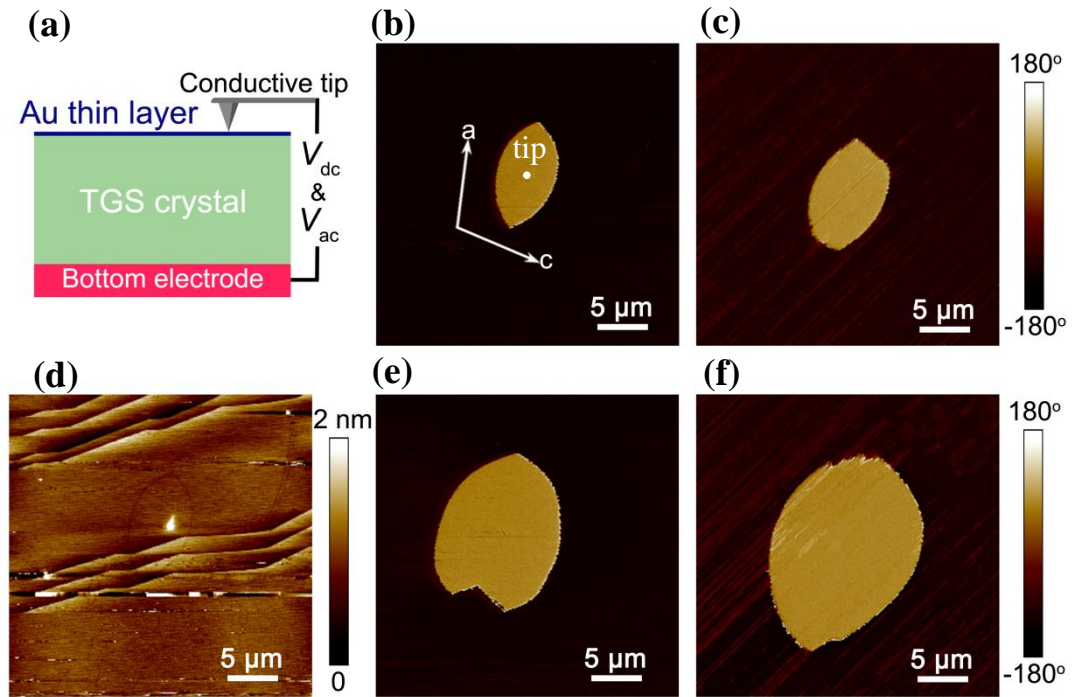


Figure S11. PFM of the (010) TGS crystal with a ~1 nm Au electrode. (a) Scheme of the (010) TGS crystal with a ~1 nm Au layer at top surface that increases surface conductivity by ~10 times. (b) AFM morphology of TGS crystal with the Au layer. Out-of-plane PFM phase images after a -180 V tip bias for (c) 0.1 s and (d) 1s at 40% humidity, (e) 0.1 s and (f) 1 s at 5% humidity, respectively.

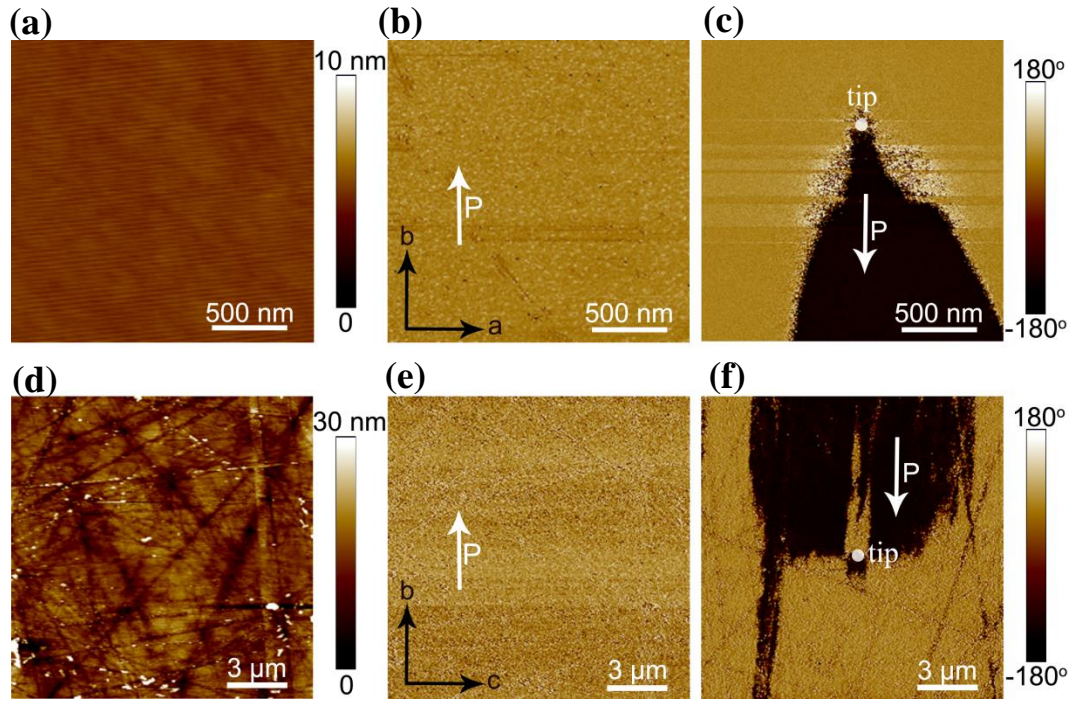


Figure S12. Tip-induced switching on non-polar cut TGS crystals in 40 % humidity. Surface, origin in-plane PFM images, and the PFM images after the (a,b,c) (100) and (d,e,f) (001) TGS crystals were polarized by a 108 V tip bias for 10 s and a -144 V tip bias for 10 s, respectively, where the white arrows show polarization orientations and the white dot represents the position of the biased tip.