Supporting Information for

Out-of-plane Piezoelectricity and Ferroelectricity in Layered α-In_2Se_3 Nano-flakes

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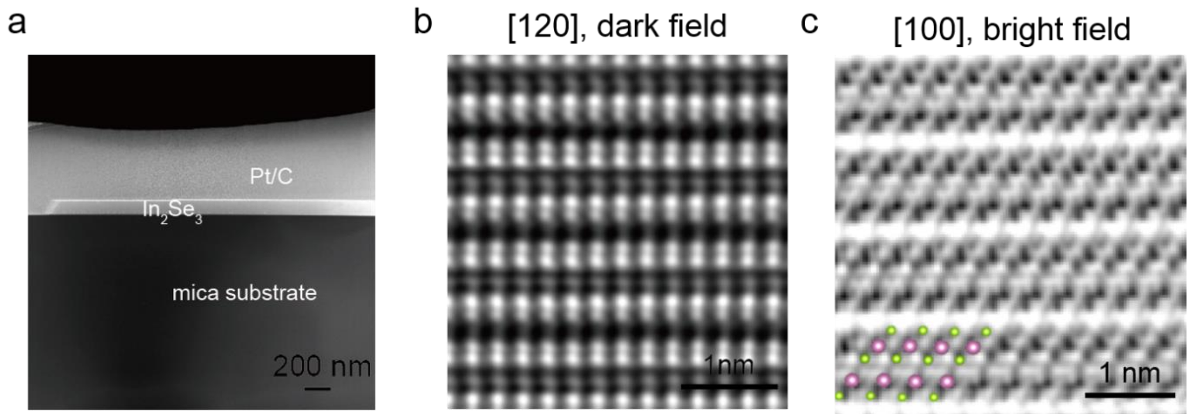

Figure S1| TEM images of In_2Se_3 flakes. (**a**) Scanning electron microscopy image of an In_2Se_3 flake cut along the [120] zone axis. (**b**) High angle annular dark field (HAADF) STEM image of an In_2Se_3 flake cut along the [120] zone axis. The Se atomic column has lower contrast possibly due to lower occupancy or larger disorder. (**c**) Cross-sectional annular bright-field (ABF) STEM image of the In_2Se_3 flake cut along the [100] direction.

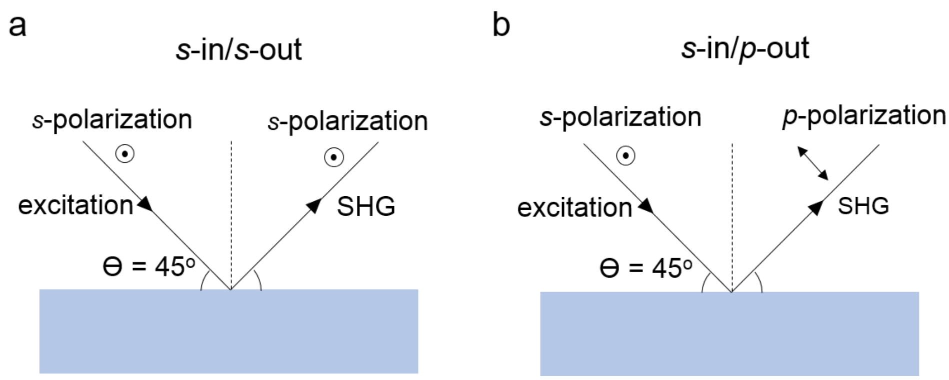

Figure S2| Configurations of SHG measurements. (**a**) For s-in/s-out configuration, both incident light and the collected SHG signal are *s*-polarized (electric field oriented perpendicular to the plane of incidence). (**b**) For s-in/p-out configuration, the incident light is s-polarized, whereas SHG signal is p-polarized (electric field direction parallel to the plane of incidence and perpendicular to wave vector).

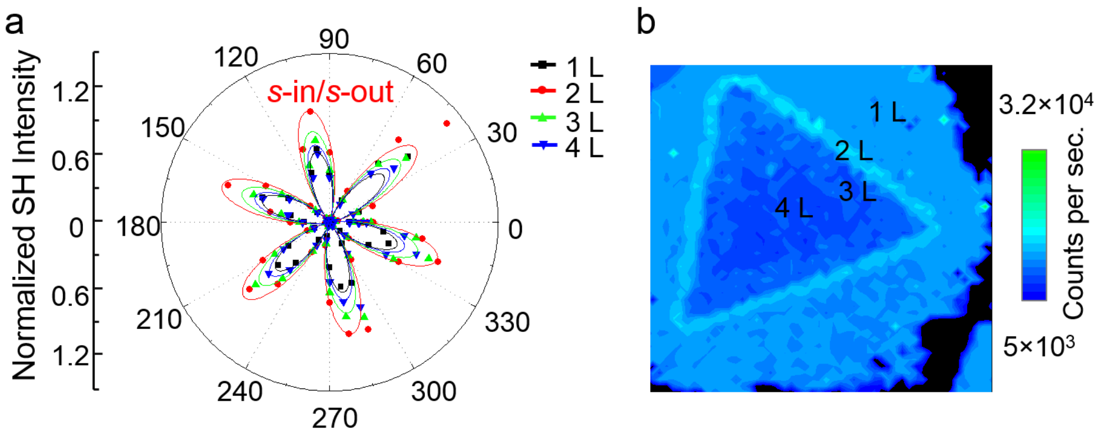

Figure S3| Layer-dependent SHG signals of VPD grown flakes. (**a**) Dependence of SHG signals on sample azimuthal angle θ in the s-in/s-out configuration for monolayer (1L), bi-layer (2L), tri-layer (3L) and four-layer (4L). The similar patterns suggest that all the layers have the same stacking orientation. (**b**) SHG signal mapping of the VPD grown flake with an azimuthal angle 20 ^{o} in the s-in/s-out configuration. The spatially resolved SHG data show that all the flakes have prominent SHG intensity, indicating that the structure of In_2Se_3 is non-centrosymmetric.

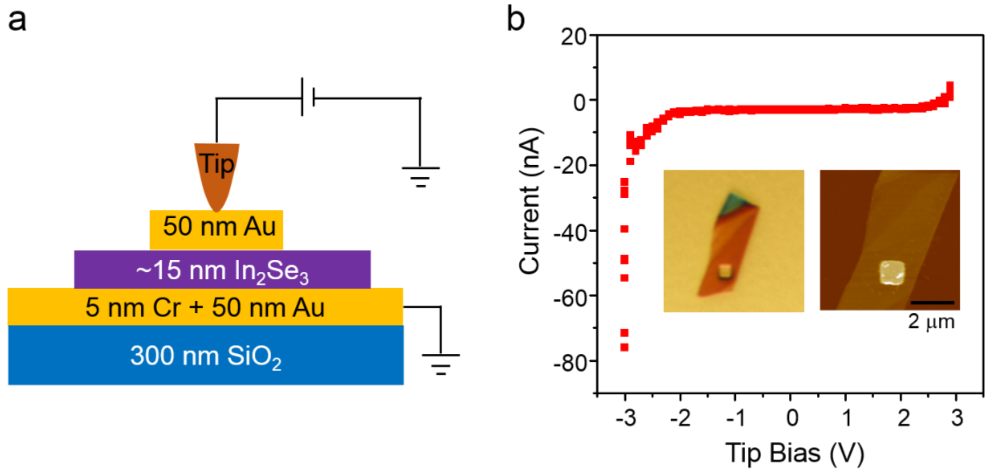

Figure S4| Leakage measurement of an In_2Se_3 flake with both top and bottom electrodes. (**a**) Schematic of the configuration for vertical leakage measurements. The bottom electrode is 50-nm-thick Au film, and top electrode is an isolated 50-nm-thick Au pad. Voltages are applied through a conducting AFM tip in contact with the top Au pad. (**b**) A typical *I-V* curve of a 15-nm-thick In_2Se_3 flake. The current becomes substantial once the tip bias is beyond ±3 V. Insert are optical and AFM images of the flake with a 1 μm × 1 μm Au pad on the surface.

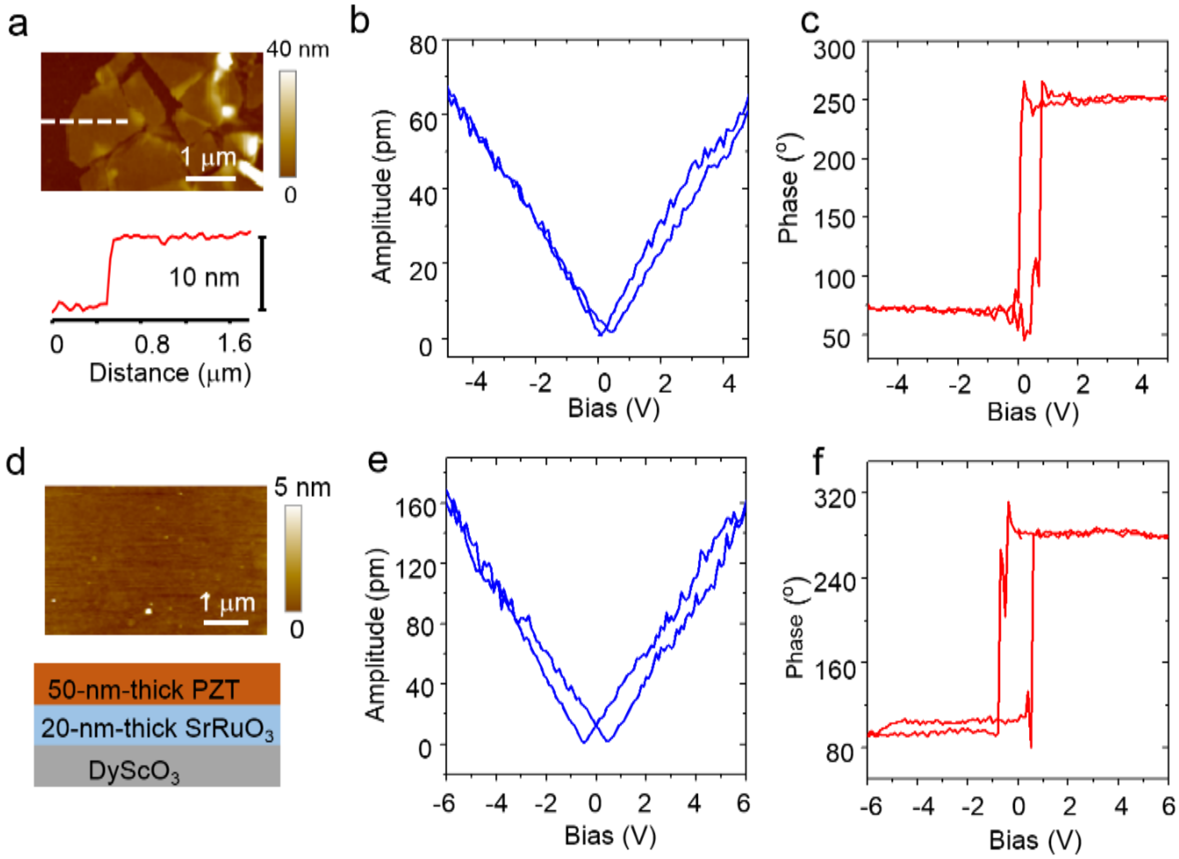

Figure S5| Polarization reversal under external electrical field. (**a**) AFM image (top) and line profile (bottom) of a 10-nm-thick In_2Se_3 flake. (**b**) On-field PFM amplitude and (**c**) PFM phase hysteresis loops on this flake. (d) AFM (top) and schematic (bottom) of a 50-nm-thick ferroelectric $PbZr_{0.48}Ti_{0.52}O_3$ (PZT) film grown on the $SrRuO_3/DyScO_3$ substrate. (**e**) On-field PFM amplitude and (**f**) PFM phase hysteresis loops of the PZT film. The same IrPt tip and bias conditions were used in all these measurements. The resemblance between (b, c) and (e, f) indicates that the ferroelectric switching makes substantial contributions to the bias-on PFM hysteresis in the In_2Se_3 sample.