Supporting Information

Bulk photovoltaic effect in two-dimensional ferroelectric semiconductor α-In_2Se_3

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Section 1: Ferroelectricity of 3R α-In2Se_3 flakes

Section 2: Characterization of thin graphite/α-In_2Se_3/thin graphite device

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Section 6: 2D α-In_2Se_3 BPV device under different light excitations

Section 1: Ferroelectricity of the 3R α-In_2Se_3 flake

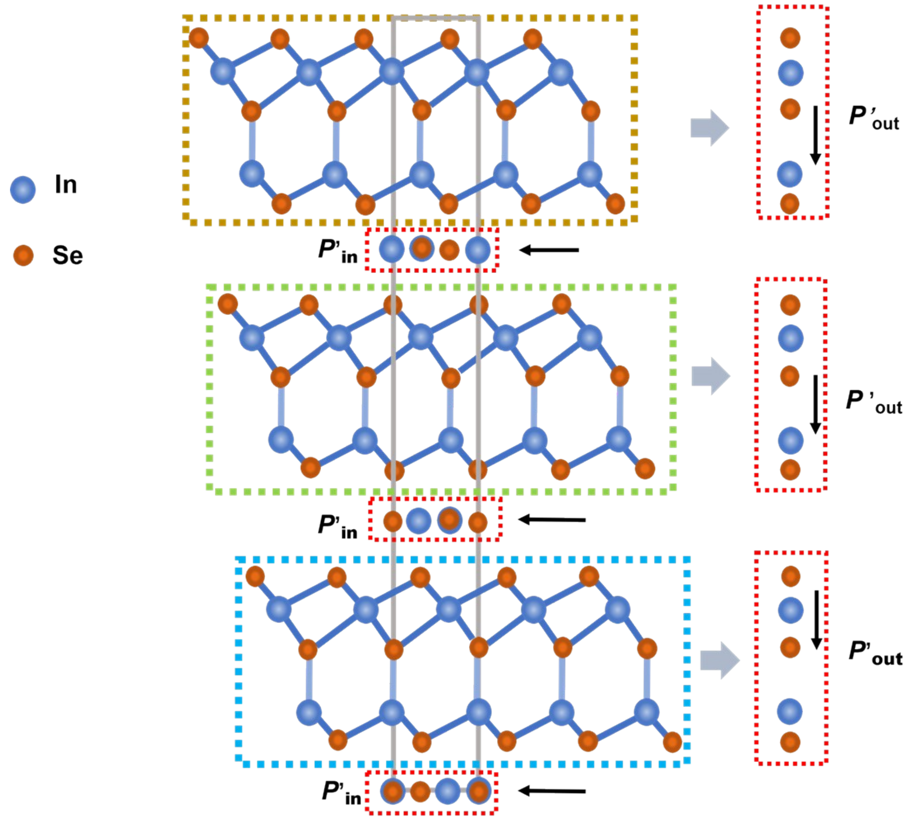

Figure S1 |Side view of a triple-layer rhombohedral (3R) α-In_2Se_3. The blue and orange balls represent In and Se atoms, respectively. The black arrows represent possible spontaneous polarizations.

Section 2: Characterization of thin graphite/α-In_2Se_3/thin graphite device

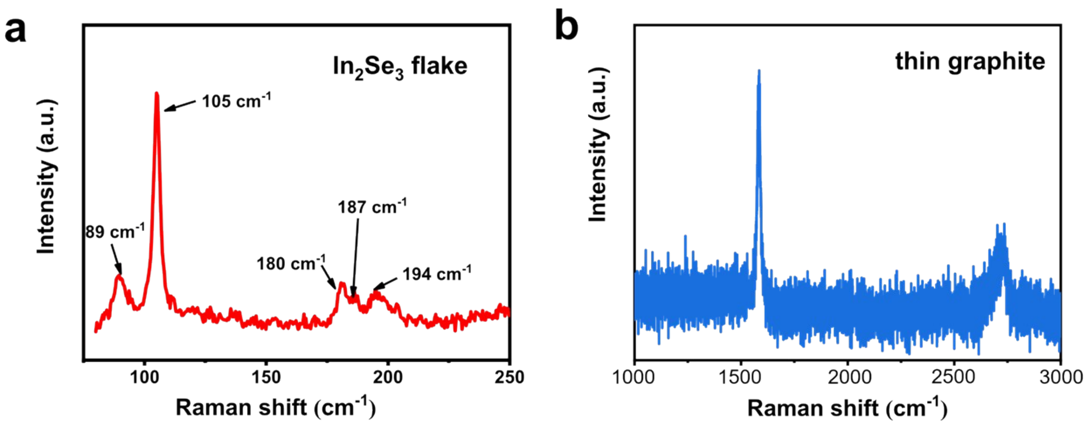

Figure S2 | XRD pattern of a α-In_2Se_3 flake (a) and thin graphite (b) from a thin graphite/α-In_2Se_3/thin graphite device.

Section 3: Electrical transport of thin graphite/α-In_2Se_3/thin graphite device

The |*I*|-*V* curve shows a counter-clockwise loop at room temperature.

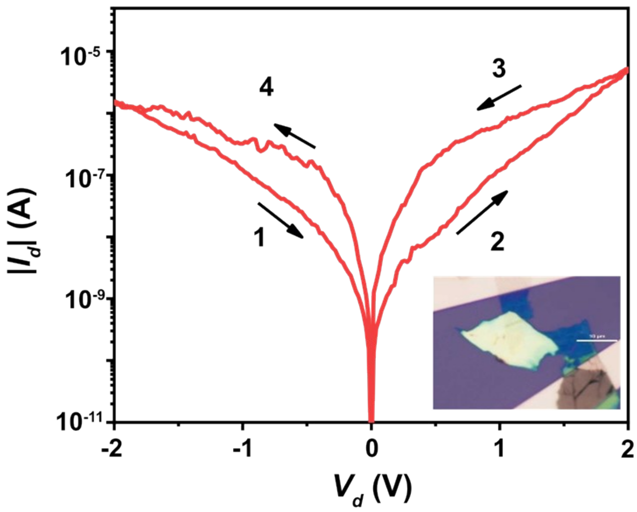

Figure S3 | Current-voltage |*I*|-*V* of a thin graphite/α-In_2Se_3/thin graphite under current sweeping at a rate of 0.1 V/s between -3 and 3 V.

Section 4: Optical characterization of a α-In_2Se_3 flake

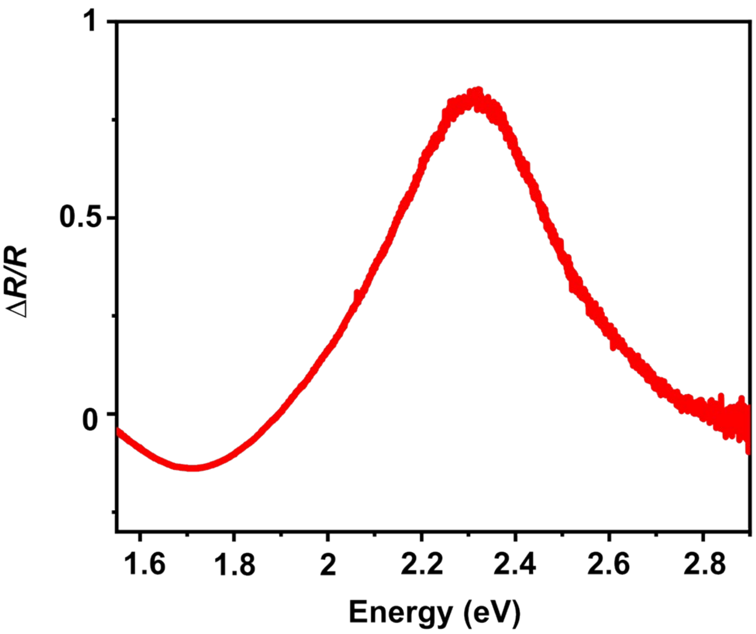

Figure S4 |Differential reflectance of a α-In_2Se_3 flake.

Section 5: Temperature dependence of I_{sc} and V_{oc} for 2D α-In_2Se_3 BPV device and its circuit Model

In order to discuss the possible mechanism underlying the temperature dependence of I_{sc} and V_{oc} under light illumination, we investigate the temperature dependence of I-V curves under dark and light conditions (illustrated in Figure S5). As the device was cooled, the total resistance containing bulk materials resistance (R_{bulk}) and contact resistance ($R_{contact}$) increased. As expected, carrier concentration in bulk semiconductors decreases at low temperatures, leading to the increase of R_{bulk}. The increase of $R_{contact}$ also occurs with decreasing temperature for the Schottky barrier.

Figure S6 displays the equivalent circuit model that may represent the device conditions during measurements of photovoltaic effect. I_{pv} is the BPV photocurrent which works as a current source. I_p and V_p are the output current and voltage of BPV device, respectively. I_b is the current within the bulk materials. The relationship of these parameters can be described by the following equation.

$$I_{pv} = I_b + I_p \qquad (1)$$

$$I_b R_{bulk} = I_p R_{contact} + V_p \qquad (2)$$

The open-circuit photovoltage (short-circuit photocurrent) is output voltage (current) when $I_p = 0$ ($V_p = 0$). Then, V_{oc} and I_{sc} are given by

$$V_{oc} = R_{bulk} I_{pv} \qquad (3)$$

$$I_{sc} = \frac{R_{bulk}}{R_{bulk} + R_{contact}} I_{pv} \qquad (4)$$

Eq. 3 indicates that V_{oc} is proportional to R_{bulk}. Suppose I_{pv} is temperature independent, V_{oc} of the device will increase with decreasing temperature. Suppose $\frac{R_{contact}}{R_{bulk}} = 1$, the I_{sc} is temperature-independent from Eq.4, which aligns well with the experimental results.

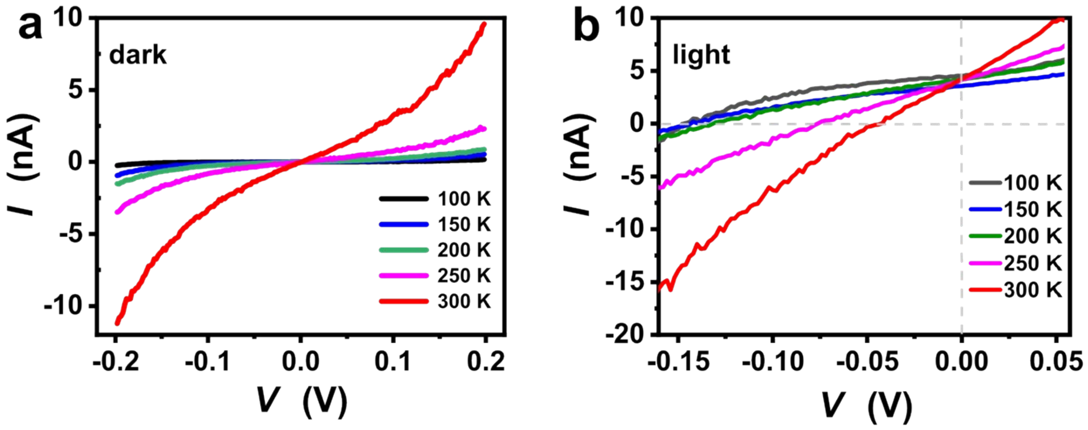

Figure S5 | Current-voltage *I-V* curves from a thin graphite/α-In_2Se_3/thin graphite device under dark (a) and light illumination (b) conditions at different temperatures.

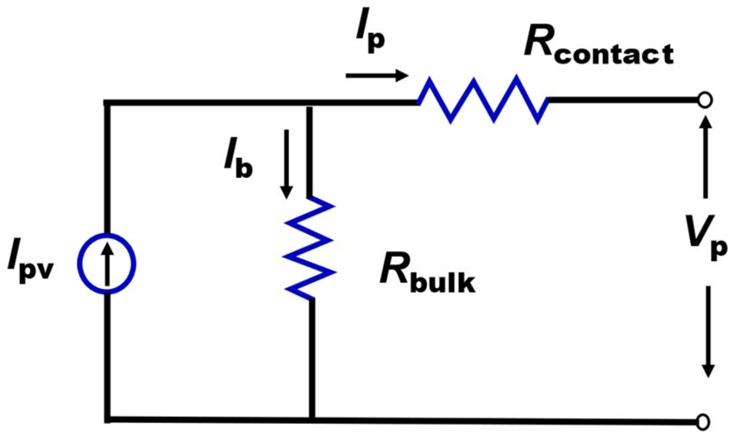

Figure S6 |An equivalent circuit mode for BPV.

Section 6: 2D In_2Se_3 BPV device under three different light excitations

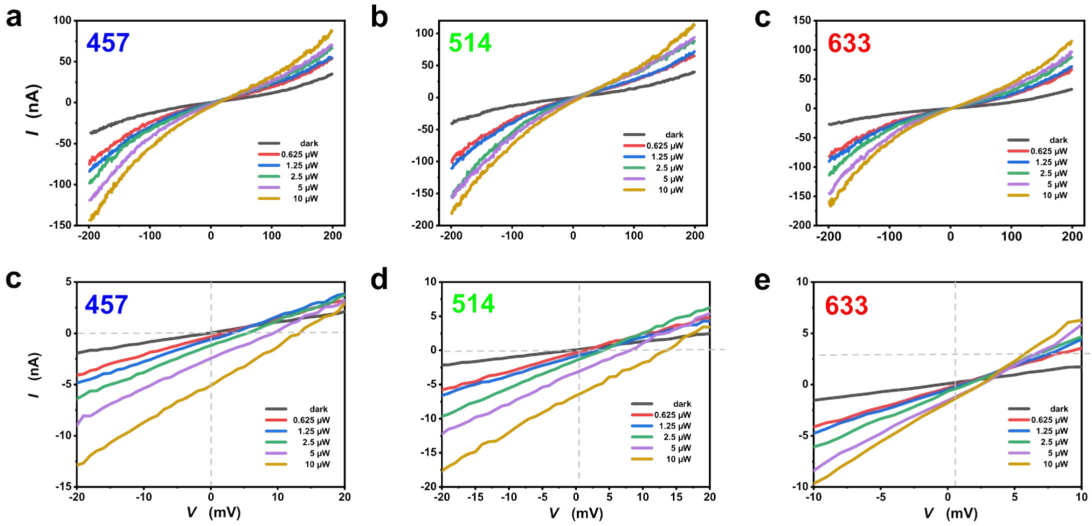

Figure S7 |Current-voltage *I-V* curves for thin graphite/In_2Se_3/thin graphite device under three light wavelengths and their corresponding zoom-in images.