Table 1: Operating parameters for CVD growth of In₂Se₃

Temperature (°C)	Flow rate (ml/min)	Pressure (Torr)	Time (mins)
800	200	5	30
			10
			5
	200		30
	100		
	50		



Figure S1: Stability analysis of 2D-In₂Se₃. Optical image after (a) one day of CVD growth (b) after one month placed in the ambient.



Figure S2: Raman spectra of sample grown at 50 ml/min flow rate (a) β -In₂Se₃ thick flake (thickness>20 nm, lateral size>10 um) (b) Thin circular flake (~1.2 nm) showed α phase of Indium Selenide. This is indicating the mixed growth of α and β phase of In₂Se₃ at low gas flow rate.



Figure S3: (a) Raman spectra of precursor powder used for evaporation. **(b)** Raman spectra of mica $[KMg3 (AlSi_3O_{10})F_2]$ substrate



Figure S4: PFM measurement in CVD grown α -In₂Se₃ at 50 ml/min flow rate, white dotted line indicated the line profile position, scale bar 2um **(a&b)** topography and height profile **(c&d)** OOP-PFM amplitude **(e&f)** OOP-PFM phase.



Figure S5: Optical image of the flake used for SHG measurement. P \sim 10 mW power is found to be the damage threshold for our sample in SHG measurement



Figure S6: schematic of the angle resolved SHG measurement system.



Figure S7: Optical images of CVD grown In_2Se_3 (a-c) at different growth time (d-f) at different flow rate



Figure S8: (a) SEM image of the nucleation site observed in AFM image of samples grown at 200 ml/mins flow rate (b) corresponding EXD spectra indicating In:Se=38:62, which is consistent with the formation of In₂Se₃



Figure S9: (a) EDX analysis of the sample (similar to Figure 1c), Inset is the SEM image of same circular flake. The EDX confirmed the atomic ratio of In:Se= 39:62, this indicated that circular flakes consist on In_2Se_3 phase (b) Raman result of similar circular flake of CVD grown β phase In_2Se_3 grown at 200 ml/min Ar gas flow rate for 5 mins.



Figure S10: OOP-PFM measurement performed at zero drive voltage indicating the zero ferroelectric response of In_2Se_3 at zero V_{ac} while the topography remains the same.



Figure S11: PFM measurement at different tip scan angle. (a-c) Topography, OOP and IP PFM amplitude at 0° (d-f) at 45° (g-i) at 135° (j-l) at 180° respectively.



Figure S12: Optical images of the samples grown at (a) low flow rate 30 ml/min (b) high flow rate of 400 ml/min.