Supporting Information

Evolution of the Electronic Band Structure and Efficient Photo-Detection in Atomic Layers of InSe

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SEM and EDX characterization on InSe grown by non-stoichiometric melt.

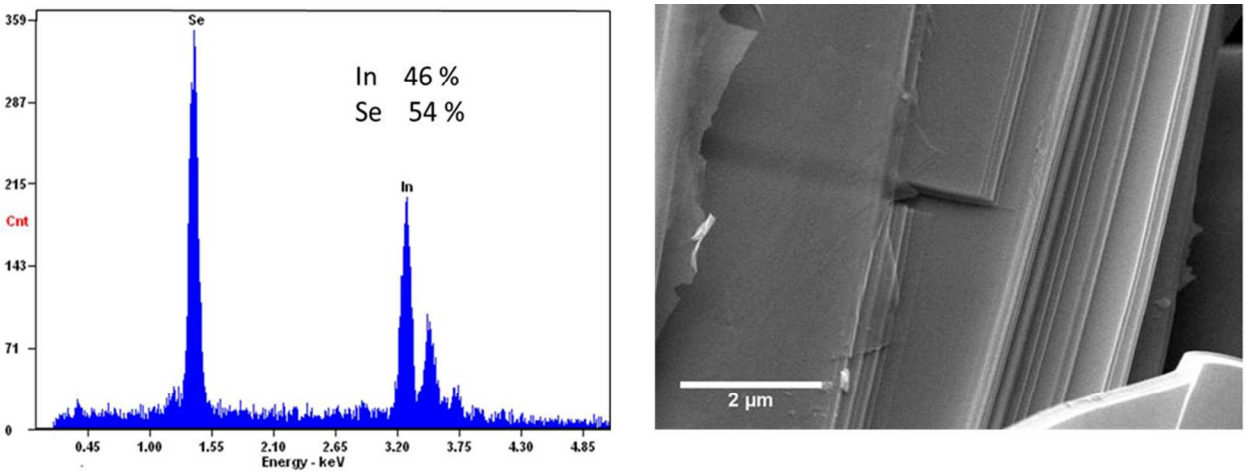

Figure S1. EDX spectra (left) and SEM image (right) of InSe crystal synthesized by non-stoichiometric melt method. EDX shows an elemental ratio of 1 :1 between In and Se. SEM image shows an obvious layered structure.

XRD Characterization

XRD is performed to study the crystal quality and yield. Figure S2 show the XRD spectrum.

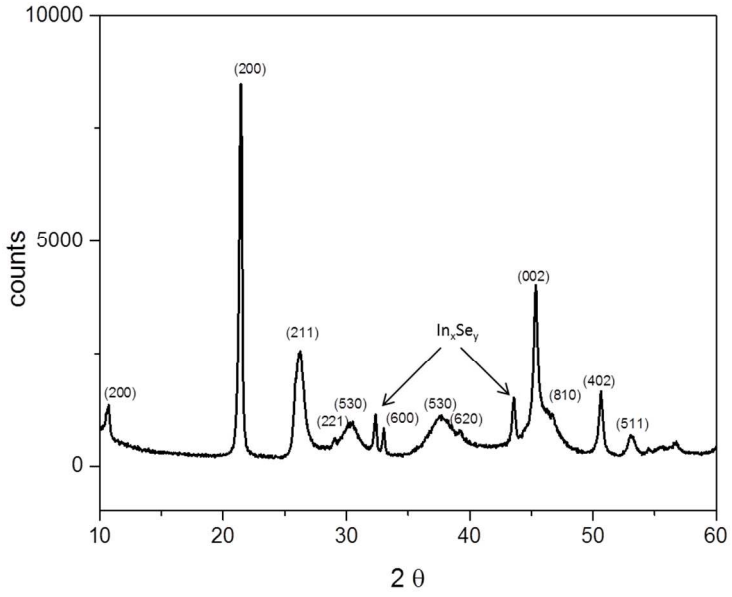

Figure S2. XRD spectrum of crystal synthesized by non-stoichiometric melt

With non-stoichiometric melt method, one will have a hard crust cover the layered texture, which is no in layered InSe phase.[1] As-grown crystal is ground into powder with the hard crust (In_xSe_y phase). The XRD shows good layered InSe phase.[2] Two extra peaks come from In_xSe_y phase.[3]

The non InSe phase will not affect the measurement in our report. Because we removed the crust and select the layered phase carefully. Meanwhile, the crust is not a layered structure, so it will not be exfoliated into thin flakes.

Reason for limited observed Raman modes

Considering three modes with frequencies out of the ability of our instrument, it is only possible to observe ten modes. The reason comes from two factors. First, most of the vibration modes are degenerated or near degenerated, because the inter-layer interaction is mainly van de Waals force and the vibration in each layer is nearly independent. So if the difference among different vibrational modes is only relative vibrational phase, they will have the same or similar eigen-energy. Second, lots of Raman active polar optical modes do not generate actual dipole momentum will not split into transverse optical mode (TO) and longitude optical mode (LO). This is supported by reported infrared (IR) observation[4, 5] that only modes A_2" (Γ_1^1)-TO mode and one E' (Γ_3^1)-TO modes are observable in IR spectra.)

Reference

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