Electronic Supplementary Material (ESI) for CrystEngComm.

Supporting Information:
Chemical epitaxy of π-phase cubic tin monosulphide

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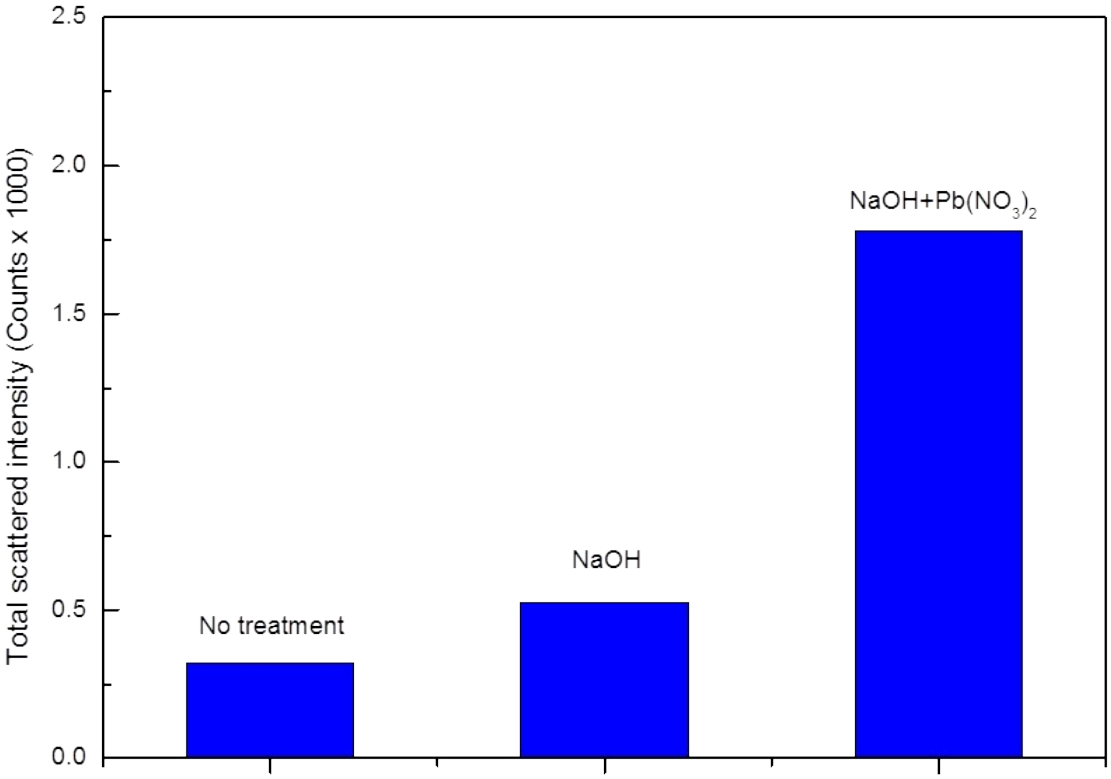

Figure S1. Total integrated intensity (TII) of all π-SnS Bragg reflections obtained in XRD, plotted against surface treatment of the GaAs substrates. Scattered X-ray intensity is highest for films obtained on substrates treated with NaOH / $Pb(NO_3)_2$ solution, indicating considerably thicker films obtained following this treatment.

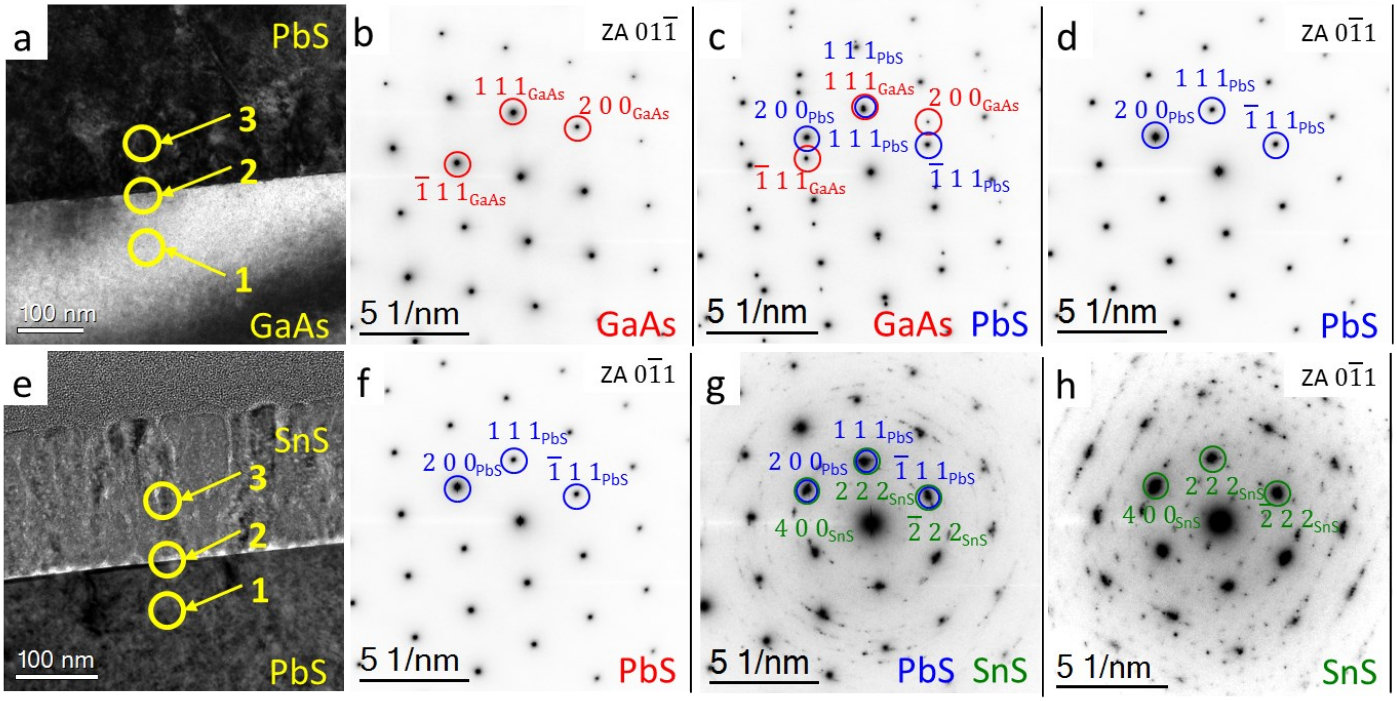

Figure S2. Cross sectional TEM analysis of π- SnS (111) thin films deposited onto GaAs(111)/PbS(111). (a) TEM micrograph showing the GaAs substrate + PbS intermediate layer (b) SAED pattern taken from circle a1 which correspond to the GaAs substrate. (c) SAED pattern taken from the area marked by circle a2 which correspond to the GaAs substrate + PbS intermediate layer. (d) SAED pattern taken from the area marked by circle a3 which correspond to the PbS intermediate layer. (e) TEM micrograph showing the PbS intermediate layer + π-SnS layer. Bright triangular shapes near the interface of SnS and PbS are likely to be voids in the film. (f) SAED pattern taken from the area marked by circle e1 which corresponds to the PbS intermediate layer. (g) SAED pattern taken from the area marked by circle e2 which corresponds to the PbS intermediate layer + π-SnS film. (h) SAED pattern taken from the area marked by circle e3 which corresponds to the π-SnS film.

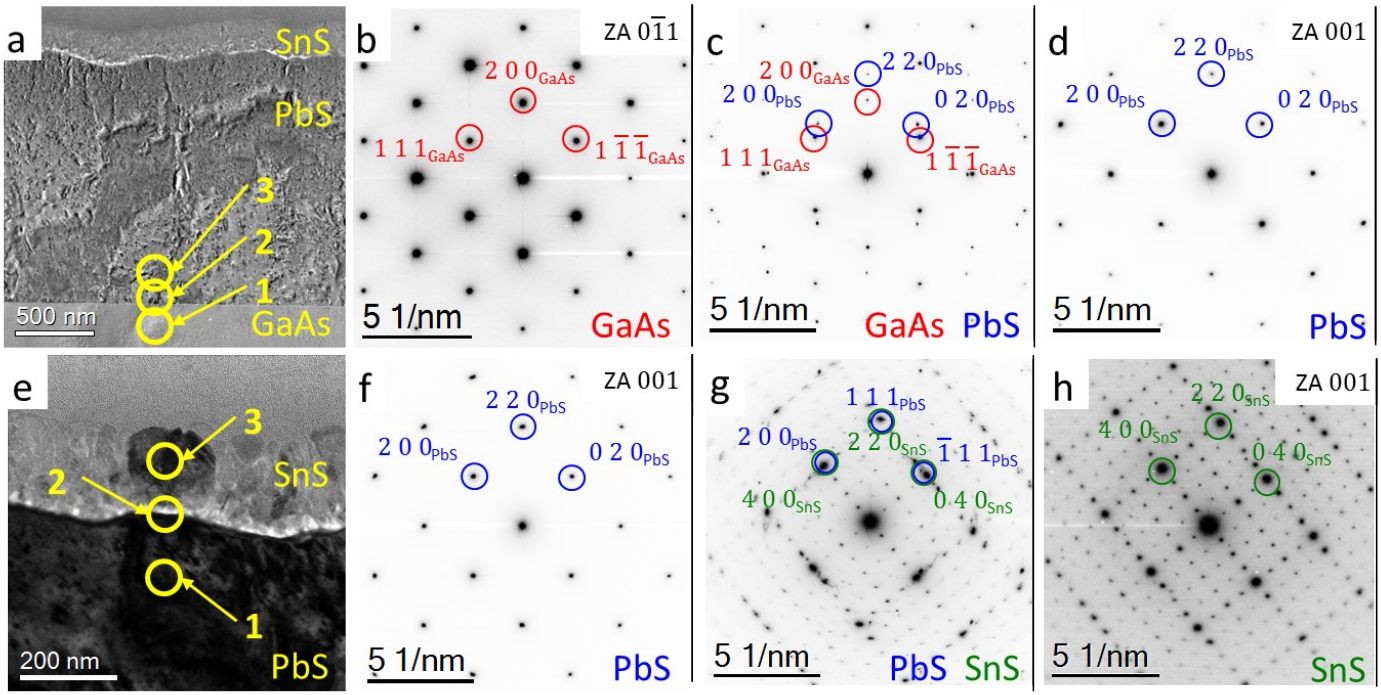

Figure S3. Cross sectional TEM analysis of π-SnS(110) thin films deposited onto GaAs(100)/PbS(110). (a) TEM micrograph showing the GaAs substrate + PbS intermediate layer (b) ED pattern taken from the area marked by circle a1 which correspond to the GaAs substrate. (c) ED pattern taken from the area marked by circle a2 which correspond to the GaAs substrate + PbS intermediate layer. (d) ED pattern taken from the area marked by circle a3 which corresponds to the PbS intermediate layer. (e) TEM micrograph showing the PbS intermediate layer + π-SnS layer. Bright shapes near the interface of SnS and PbS are likely to be voids in the film. (f) ED pattern taken from the area marked by circle e1 which correspond to the PbS intermediate layer. (g) ED pattern taken from the area marked by circle e2 which includes parts of the PbS intermediate layer + π-SnS film. (h) ED pattern taken from the area marked by circle e3 which corresponds to the π-SnS film.

Mosaic spread:

SAED patterns were projected azimuthally in order to allow line scan analysis along the diffractions arcs. the broadening and splitting of the diffraction reflections correspond to the mosaic spread in the films and orientation variation, respectively. For SAED taken from the monocrystalline PbS intermediate layer, a single reflection typically appears. Pseudo-vogit function was fitted to the analyzed reflections in order to measure the full width at half maximum (FWHM). For π-SnS films typically more than one reflection appears. In this case, pseudo-vogit function was fitted to each reflection and the mosaic spread was calculated as the width of both reflections at half the peak intensity. The results are presented in Figs. S4 and S5 below.

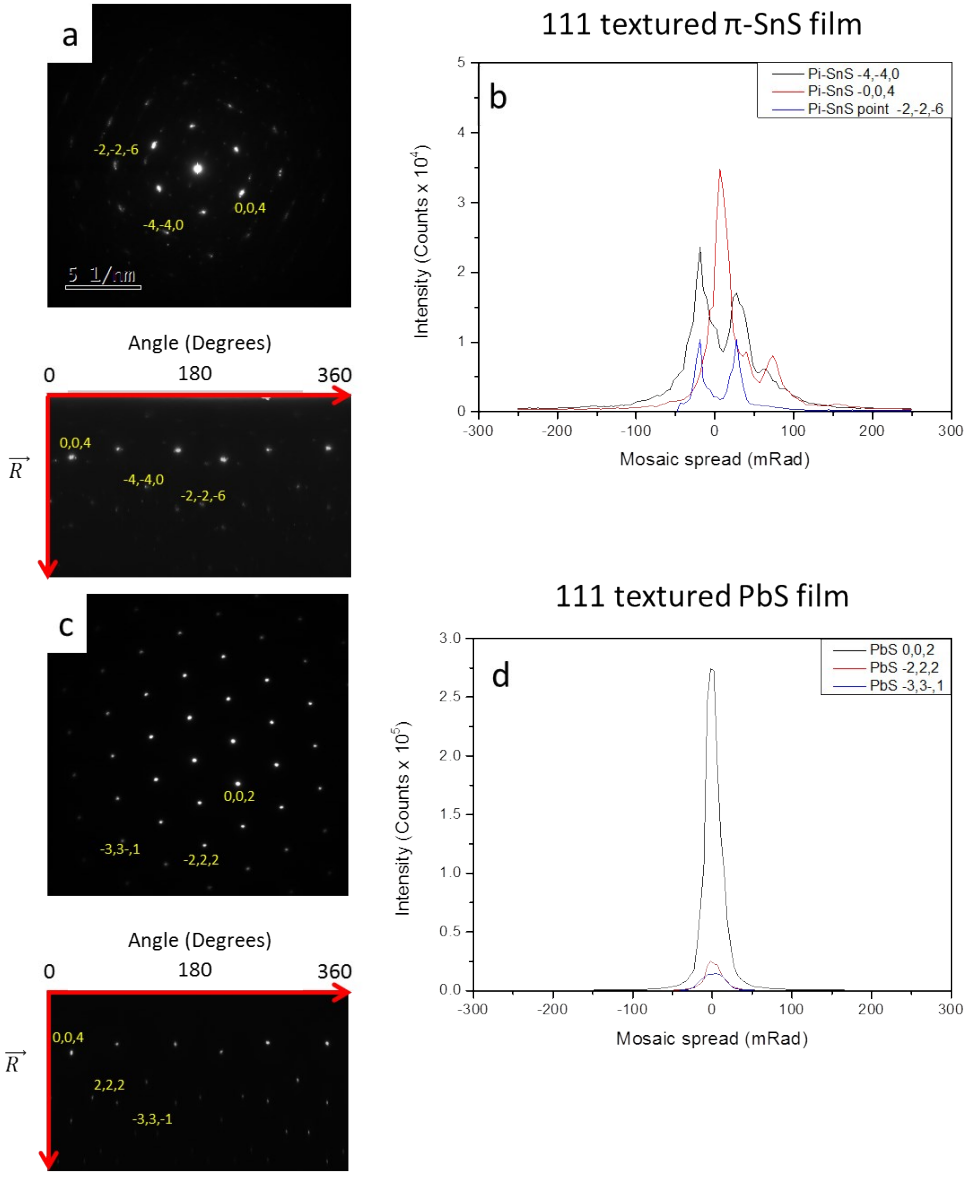

Figure S4. SAED patterns, azimutal projection of the SAED patterns and corresponding line scans of selected diffraction spots aquired from figure 4c and figure 4f in main text (the PbS(111)/SnS(111) interface). Results are presented for each of the two layers: (a) SAED patterns and corresponding azimuthal projection of SAED pattern obtained for π-SnS film with a very strong [111] texture. Miller indices of the reflections analyzed are denoted in yellow in the figure. (b) Line scan across the three reflections marked in a. The appearance of several peaks for a single reflection indicate on multiple crystallographic domains which are slightly mis-oriented (c) SAED patterns and corresponding azimuthal projection obtained for a (111) oriented PbS film. Miller indices of the reflections analyzed are denoted in yellow in the figure. (d) Line scan across the three reflections marked in c. In the case of PbS, a single peak appears for each reflection, indicating a single crystallographic orientation.

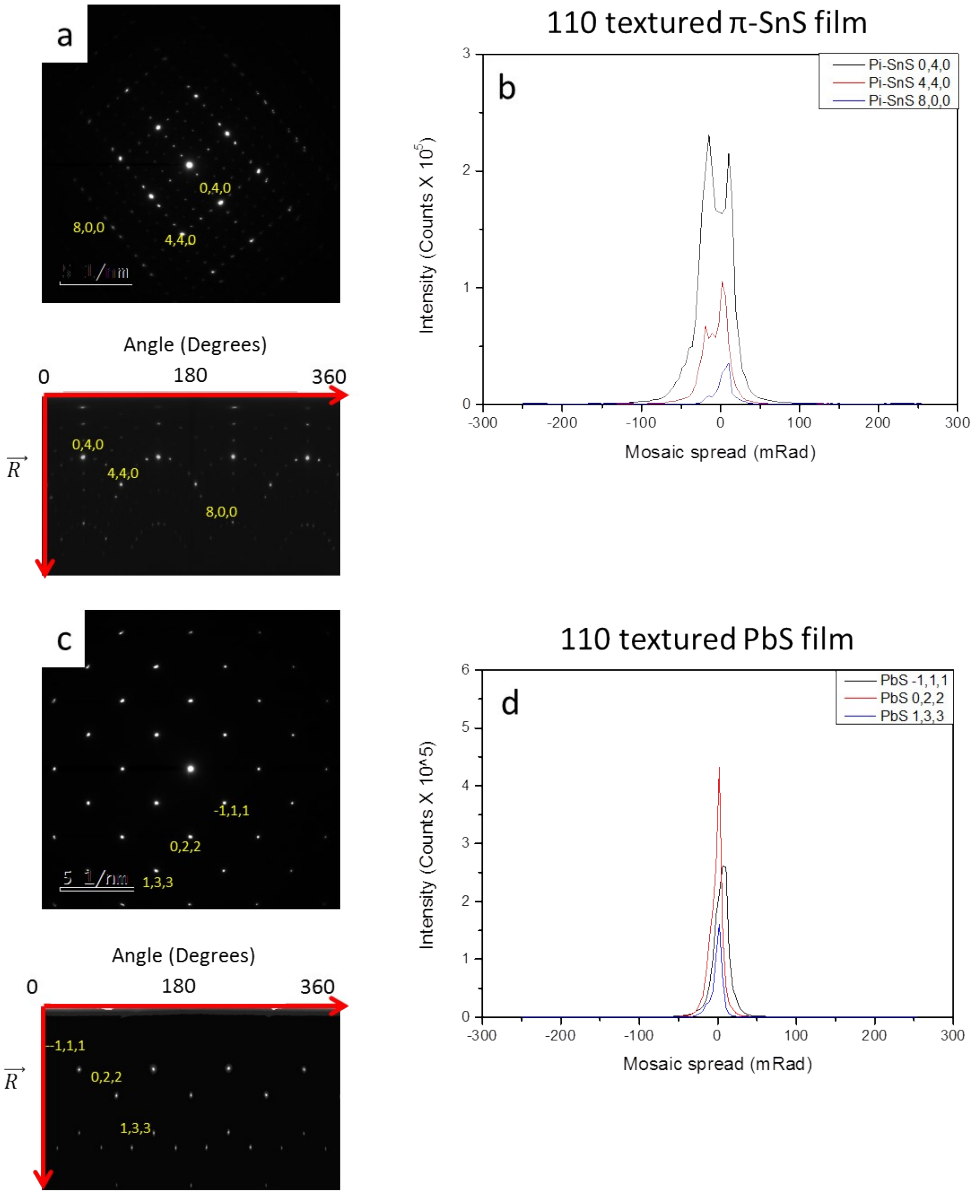

Figure S5. SAED patterns, azimutal projection of the SAED patterns and corresponding line scans of selected diffraction spots aquired from SAED patterns in figures 5c and 5f in main text (the PbS(110)/SnS(110) interface). Results are presented for each of the two layers: (a) SAED patterns and corresponding azimuthal projection of SAED pattern obtained for π-SnS film with a strong [110] texture. Miller indices of the reflections analyzed are denoted in yellow in the figure. (b) Line scan across the three reflections marked in a. The appearance of several peaks for a single reflection indicate on multiple crystallographic domains which are slightly mis-oriented (c) SAED patterns and corresponding azimuthal projection obtained for a (110) oriented PbS film. Miller indices of the reflections analyzed are denoted in yellow in the figure. (d) Line scan across the three reflections marked in c. In the case of PbS, a single peak appears for each reflection, indicating a single crystallographic orientation.

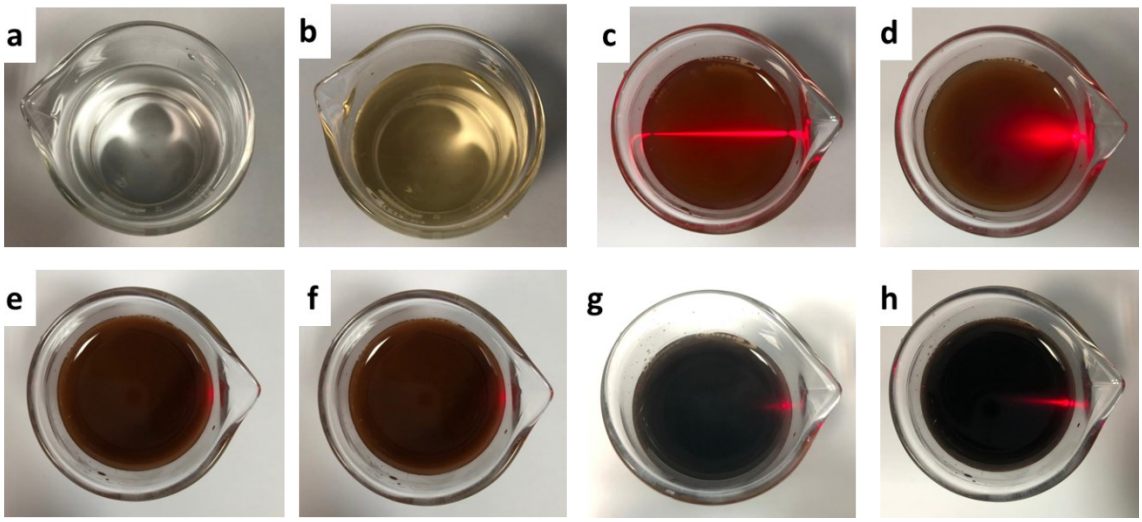

Figure S6. Time resolved light scattering from the SnS deposition solution. A red laser beam was passed through the deposition bath from right to left, and photos were taken at predefined times. (a) Prior to reaction initiation; (b) Immediately after addition of the sulfide precursor and reaction initiation; (c) after 1 min. (d) 5 min. (e) 10 min. (f) 60 min. (g) 180 min. (h) 240 min.

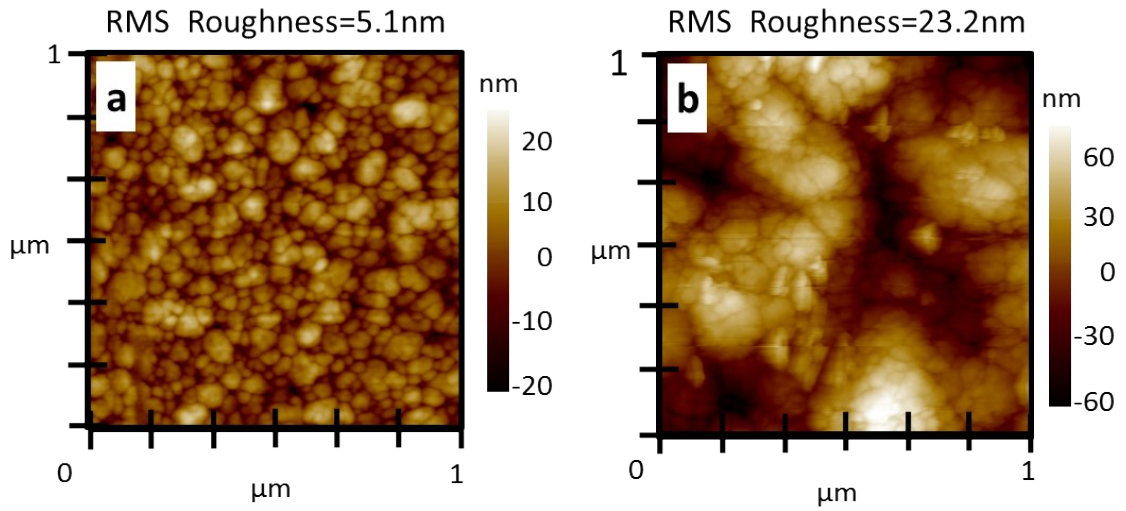

Figure S7. AFM topography images of SnS film deposited on (a) GaAs(111)/PbS(111) (b) GaAs(100)/PbS(110).

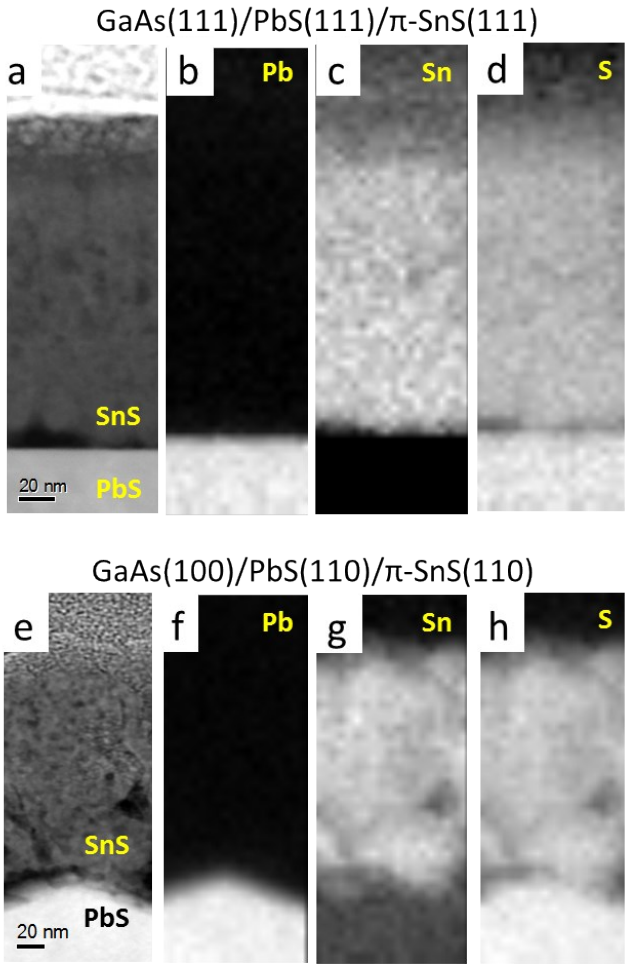

Figure S8. (a) Cross-section STEM micrograph of PbS(111)/π-SnS(111) and corresponding elemental maps of (b) Pb (c) Sn (d) S (e) HRTEM micrograph of PbS(110)/π-SnS(110) and corresponding elemental maps of (f) Pb (g) Sn (h) S. Magnification is the same for each set of micrographs.