

Figure 1 Control of doping in α -Sn films via surface termination of the InSb(001) substrate. a) ARPES measurement of 13 BL α -Sn grown in the In-rich c(8x2) reconstruction. b) ARPES measurement of 13 BL α -Sn grown in the Sb-rich c(4x4) reconstruction. A photon energy of 21 eV is chosen to accentuate the surface states. A clear shift in chemical potential from below the Dirac node to at the Dirac node is evident from a) to b). c) shows R_{xx} and d) shows R_{xy} measurements of 56 BL α -Sn grown on the same InSb surface terminations. Conductive InSb substrates make quantification difficult, but a clear change in carrier type is evident, showing the change in doping in a), b) is carried through to the bulk α -Sn.



Figure 2 Confirmation of 3D topological insulator-like phase. ARPES measurement at 21 eV before a) and after b) potassium dosing the surface. Potassium dosing effectively electron dopes the surface, allowing to see further above the surface Dirac node. The surface Dirac node is preserved without a gap opening. c) ARPES measurement at 127 eV after potassium dosing the surface. At this photon energy the bulk states at the bulk Γ point are observed. There is clear evidence of the valence band, but no conduction band evident below the Fermi level. This indicates at least a 200 meV bulk band gap. This film is thus 3D topological insulator-like.





Figure 3 Growth of α -SnGe thin films. a) RHEED oscillations showing preservation of layer by layer growth mechanism when the Sn films are alloyed with Ge. b) RHEED pattern of (2x1)/(1x2) in 50 BL α -Sn. c) The same as b) in 50 BL α -SnGe. d) High resolution XRD reciprocal space map around the InSb(115) peak showing the α -SnGe is fully strained. Pendellosung fringes in the (001) direction are clear, indicative of highquality interfaces.

Figure 4 Topological phase transition upon Ge alloying α -Sn films studied via ARPES measurements at 21 eV to accentuate the surface states. a) Pristine 13 BL Sn, b) Potassium dosing the film in a). c) 5% Ge alloying, resulting in about +0.5% strain. d) 8% Ge alloying, resulting in about +0.9% strain. A small gap opens at the surface Dirac node. A double peak is evident just above the gapped Dirac node. The exact cause is unclear, but is likely an additional, unpredicted surface state. e)-h) shows the curvature of a)-d), enhancing the contrast of the bands.