Growth and angle-resolved photoemission of strain- and thickness- tuned epitaxial α-Sn thin films

A.N. Engel[1], H.S. Inbar[1], P.J. Corbae[2], C.P. Dempsey[2], S. Nishihaya[2], Y.-H. Chang[1], J. T. Dong[1], A.V. Fedorov[3], M. Hashimoto[4], D. Lu[4], C. J. Palmstrøm[1,2]

[1]*Materials Department, University of California Santa Barbara, CA, USA*

[2]*Dept. of Elec. and Comp. Eng., University of California Santa Barbara, CA, USA*

[3]*Advanced Light Source, Lawrence Berkeley National Lab, CA, USA*

[4]*Stanford Synchrotron Radiation Lightsource, SLAC National Accelerator Lab, CA, USA*

α-Sn, the diamond structure allotrope of Sn, is a zero-gap semiconductor with band inversion. Calculations suggest that epitaxial tensile strain induces a 3D topological insulator (TI) phase, while epitaxial compressive strain induces a 3D Dirac semimetal (DSM) phase [1,2]. When this DSM phase is confined in a thin film, it has been suggested to form a quasi-3D TI phase [3]. Transitions to other phases instead, such as 2D TI, have been suggested as well [4].

We first explore the topological phase of ultrathin unintentionally doped α-Sn thin films. Using spin- and angle-resolved photoemission spectroscopy (ARPES), we study compressively strained α-Sn films on InSb(001). We find clear evidence of the confinement-induced quasi-3D TI phase in compressively strained α-Sn. We also find that the spin-polarization of the topological surface states differs markedly from reports in the literature where the films are intentionally doped with Te [5], indicating this intentional doping (a frequently used procedure) could have a significant effect on the electronic structure of α-Sn.

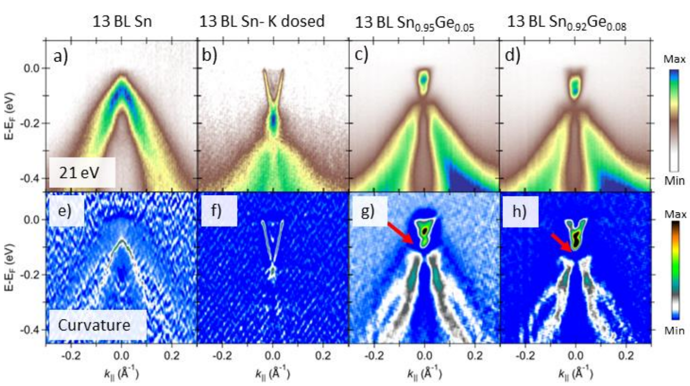

Figure 1. Topological phase transition upon Ge alloying as observed in ARPES (h*ν*=21 eV) spectra for 13 bilayers a) Pristine α-Sn (-0.15% strain) b) Potassium dosed (electron doping) of the same film c) 5% Ge alloying (+0.5%) d) 8% Ge alloying (+0.9%). A small gap opens at the surface Dirac node with Ge alloying, indicating a topological phase transition. The size of this gap increases with Ge content e)-h) Curvature of a)-d) to enhance band features.

With the previous behavior benchmarked, we then alloy the α-Sn films with Ge to decrease the bulk lattice constant and switch from compressive to tensile strain when grown on InSb(001). Morphology changes as a function of Ge alloying were studied with *in-situ* scanning tunneling microscopy, and strain was confirmed through X-ray diffraction. Finally, the presence of a topological phase transition induced by tensile strain *away* from the expected 3D TI phase is found in ARPES (Fig. 1). Our results pave the way for a better understanding of the effect of strain and confinement on α-Sn's band structure.

[1] Phys Rev B **97**, 195139 (2018).
[2] Phys Rev B **90**, 125312 (2014).
[3] Phys Rev Lett **111**, 216401 (2013).
[4] Advanced Materials **33**, 2104645 (2021).
[5] Phys Rev B **97**, 75101 (2018).

Author for correspondence: aengel@ucsb.edu

Supplementary Information

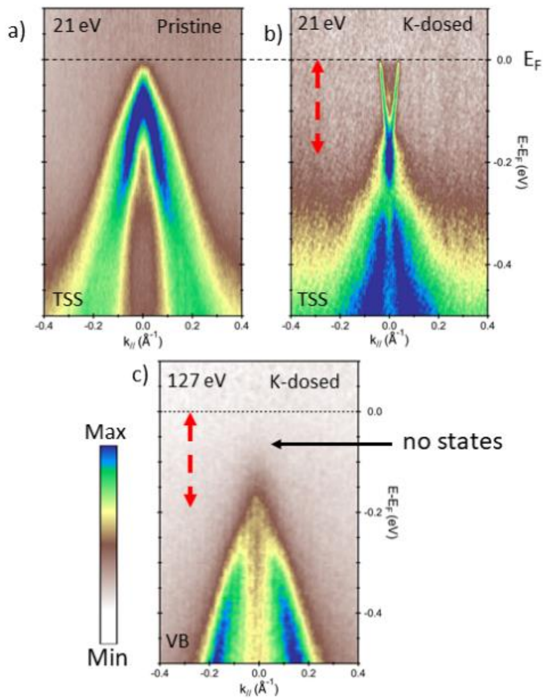

Figure 2. Determination of the 3D topological insulator-like state in 13 BL α-Sn. ARPES measurement at 21 eV before a) and after b) potassium dosing the surface. Potassium dosing effectively electron dopes the surface, allowing to see further above the surface Dirac node. The surface Dirac node is preserved. c) ARPES measurement at 127 eV photon energy after potassium dosing the surface. At this photon energy the bulk states at the bulk Γ are observed. There is clear evidence of the valence band, but no conduction band below the Fermi level. This indicates at least a 200 meV bulk band gap. This film is thus 3D topological insulator-like.

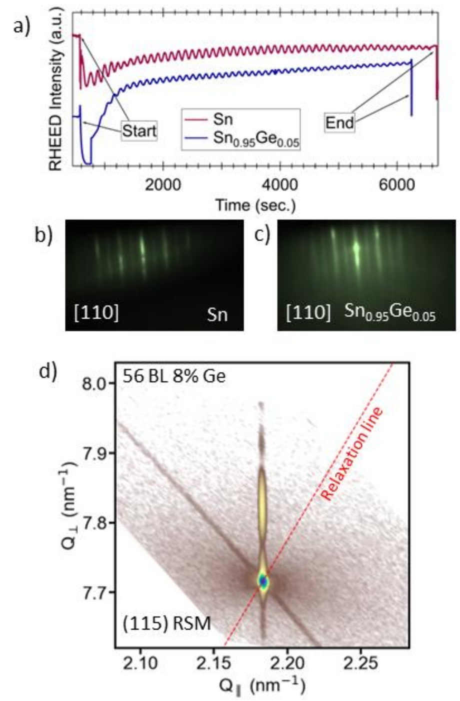

Figure 3. Growth of α-SnGe thin films. a) RHEED oscillations showing preservation of layer by layer growth mechanism when the Sn films are alloyed with Ge. b) RHEED pattern of (2x1)/(1x2) in 50 BL α-Sn. c) The same as b) in 50 BL α-SnGe. d) High resolution XRD reciprocal space map around the InSb(115) peak showing the α-SnGe is fully strained. Pendellosung fringes in the (00l) direction are clear, indicative of high quality interfaces.

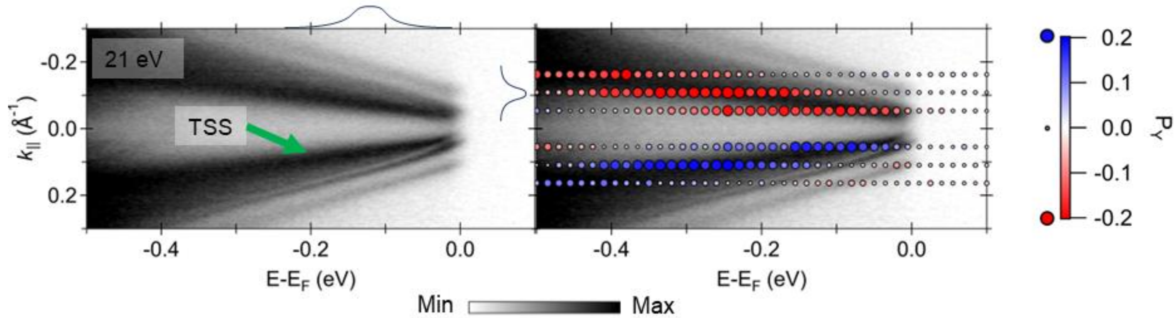

Figure 4. Spin polarization of the surface states of a 400 BL α-Sn film. Left: Measurement of the surface states of 400 BL α-Sn. Right: Spin polarization in the direction perpendicular to the momentum, measured only at the specified $k_{\parallel}$ values and overlaid on the full ARPES spectrum. Spin polarization is only observed in the direction perpendicular to the momentum, thus the Dirac surface states are truly spin-momentum locked. This contrasts with a previous report where spin-momentum locking was not enforced in Te doped α-Sn films [5]. The spin polarization measurements are at a lower resolution than the E-*k* measurement. The resolution for the spin-polarized measurement is indicated with the corresponding Gaussian on the E-*k* measurement.