Epitaxial Growth and Electronic States of Ultrathin Bi (0001) Films on InSb (111)B

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Synthesizing high-quality single-domain bismuth films with controlled strain, interface chemistries, and ultrathin thickness remains a fundamental materials science challenge with significant consequences such as obtaining high spin-polarization in spintronic devices[1] and 1D-edge transport in the theortical quantum spin Hall phase[2]. Tensile strain in quantum-confined (0001) bismuth films is predicted to lead to a band gap opening[3], allowing surface states to contribute mainly to transport signatures. Yet, only small-area planar Bismuthene wetting layers[4] or thick (>6 bilayers, BL) compressively strained films were studied on semiconducting substrates[5].

In this work, ultrathin large-area Bi (0001) layers were grown on InSb (111)B substrates by molecular beam epitaxy and *in vacuo* transferred for synchrotron-based angle-resolved photoemission spectroscopy (ARPES). *In vacuo* scanning tunneling microscopy (STM) and *ex situ* x-ray diffraction confirm the successful synthesis of single-crystal Bi epitaxial films. Through a core-level photoemission study we show that large-area single-domain ultrathin Bi films can be stabilized through strong film-substrate interactions. With decreasing film thickness from 200 BL to 1 BL, we quantify the confinement-induced shifts in the bulk band structure and trace the quantum well energy levels with a phase accumulation model. Significant film-substrate bonding breaking inversion symmetry affect the surface state dispersion leading to a surface state degeneracy which allows us to assign the topological order in Bi (0001) thin films. The findings of this study offer a new route for epitaxial growth and integration of band-engineered Bi films with III-V substrates.

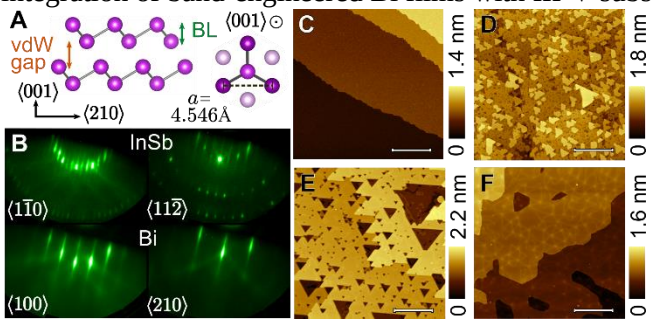

Figure 1: (A) Side- and top-view of the Bi (0001) surface model showing intra-layer covalent-like bonds and inter-layer van der Waals (vdW) stacking. (B) Reflection high energy diffraction patterns and epitaxial alignment of InSb (111)B and Bi (0001). (C) STM images (scale bar 100 nm) of InSb (111)B substrate and (D) 1.3 BL (E) 2.6 BL (F) 5.4 BL bismuth films.

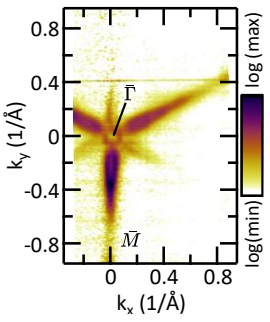

Figure 2: (0001) Fermi surface of a 2nm thick (5.4 BL) bismuth film, showing the 3-fold symmetry of the bismuth layer indicating a single domain is present. Collected at $h\nu = 37.5eV$.

Reference

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Supplemental Information

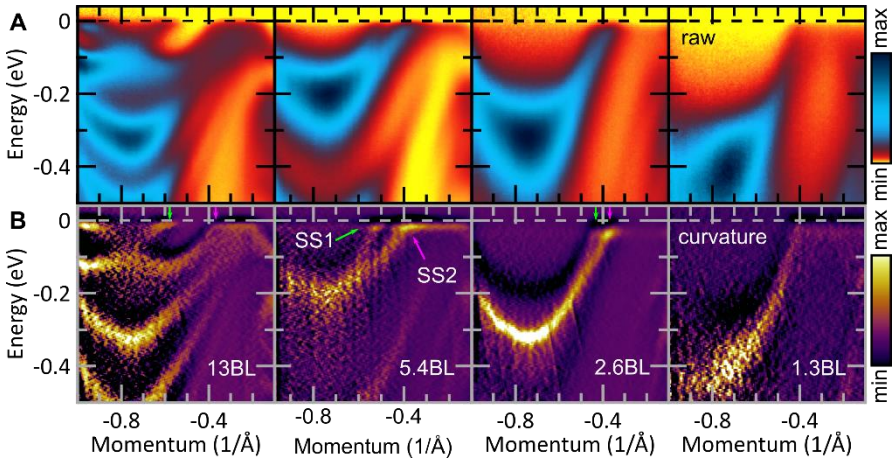

Figure S1: (A) ARPES raw images and (B) curvature plots of the surface state band dispersion along $\bar{M} - \bar{\Gamma}$ for varying film thickness, collected at $h\nu = 37.5eV$. The evolution of the surface state dispersion as a function of film thickness. Inversion symmetry breaking leads to a crossing of the Rashba surface states at $\bar{M}$ (k=0.79Å^{-1}), which would result in surface state spin-polarization maintained down to the ultrathin limit.

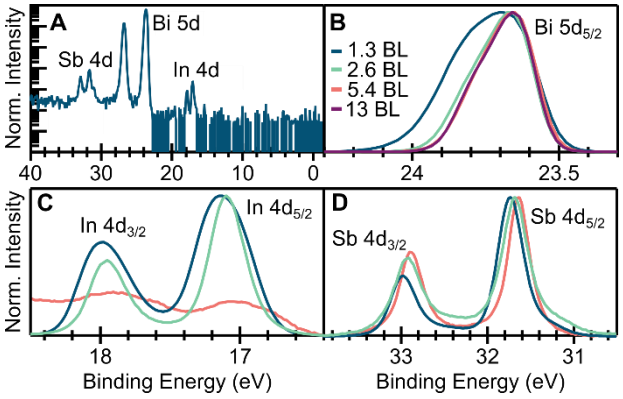

Figure S2: Changes in core level photoemission spectra upon Bi deposition on InSb(111)B. (A) log-scale survey spectrum of the 1.25BL collected at 80eV. Photoemission spectra of (B) Bi $5d_{5/2}$ measured at 80eV (C) In 4d at 50eV and (D) Sb 4d at 80 eV.Photoemission experiments provide evidence for the strong interface bonding, showing signatures of Bi-Sb and Bi-In bonding for the thinnest bismuth films, followed by a core level shift as the film thickness increases.