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Selective hydrogen etching leads to 2D Bi (111) bilayers on Bi2Se3 : Large Rashba splitting in topological insulator heterostructure

Ultrathin bilayers (BLs) of bismuth have been predicated to be a two-dimensional (2D) topological insulator. Here we report on the new route to manufacture the high quality Bi bilayers from 3D topological insulator, a top-down approach to prepare large-area and well-ordered Bi (111) BL with deliberate hydrogen etching on epitaxial Bi2Se3 films. With scanning tunneling microscopy (STM) and X-ray photoelectron spectra (XPS) in-situ, we confirm that the removal of Se from the top of a quintuple layer (QL) is the key factor, leading to a uniform formation of Bi (111) BL in the van der Waals gap between the first and second QL of Bi2Se3. The angle resolved photoemission spectroscopy (ARPES) in situ and complementary density functional theory (DFT) calculations show that a giant Rashba splitting with a coupling constant of 4.5 eV． Å in the Bi (111) BL on Bi2Se3. Moreover, the thickness of Bi BLs can be tuned by the amount of hydrogen exposure. Our ARPES and DFT study indicated that the Bi hole-like bands increase with increasing the Bi BL thickness. The selective hydrogen etching is a promising route to produce a uniform ultrathin 2D TI that is useful for fundamental investigations and applications in spintronics and valleytronics.

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