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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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