Electronic Supplementary Information: Emergence of quasi-1D spin-polarized states in ultrathin Bi films on InAs(111)A for spintronics applications

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S1. X-RAY PHOTOELECTRON SPECTROSCOPY (XPS) MEASUREMENTS ON STABILITY OF BI-DIMER/INAS(111)-$2\sqrt{3}\times 3$ AND BI-BL/INAS(111)-2×2 PHASES

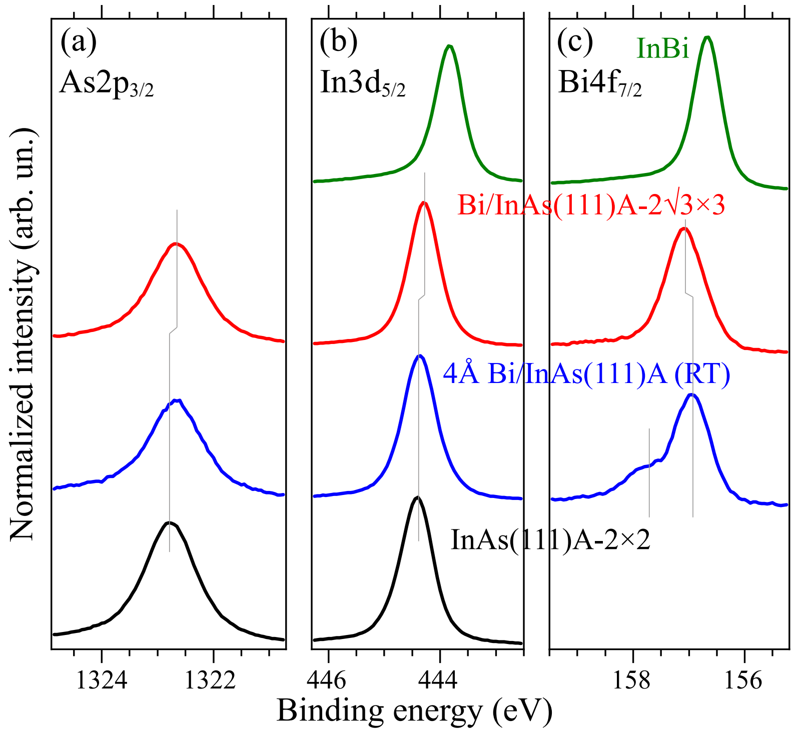

FIG. S1. XPS spectra ($h\nu = 1486.7$ eV) of the (a) As-2$p_{3/2}$, (b) In-3$d_{5/2}$ and (c) Bi-4$f_{7/2}$ core levels. Black, blue, red and green lines correspond to clean InAs(111)-2×2, 4 Å thick Bi film, grown on InAs(111)-2×2 at room temperature, Bi-dimer/InAs(111)-$2\sqrt{3}\times 3$ and InBi (001) surfaces, respectively.

Chemical stability is a vital characteristic for a material in nanotechnology applications, especially for thin 2D materials. To investigate the interaction between the Bi adlayer and the substrate experimentally, the chemical composition of the Bi-dimer/InAs(111)-$2\sqrt{3}\times 3$ system was studied using X-ray photoelectron spectroscopy (XPS). Figures S1a,b,c show the As-2$p_{3/2}$, In-3$d_{5/2}$ and Bi-4$f_{7/2}$ core levels XPS spectra for the pristine InAs(111)2×2 surface (black lines), surface of 4 Å thick Bi film, grown on InAs(111)-2×2 at room temperature (blue lines), Bi-dimer/InAs(111)-$2\sqrt{3}\times 3$ surface, formed by deposition of a 4 Å thick Bi film onto InAs(111)-2×2 surface at 300 °C (red lines) and the vacuum-cleaved InBi (001) surface (green lines). The As-2$p_{3/2}$ and In-3$d_{5/2}$ lines XPS spectra for the pristine InAs(111) surface (black curve in Figures S1a and S1b) have binding energies of 1322.75 eV and 444.4 eV, respectively. The deposition of 4 Å bismuth onto the InAs(111)2×2 surface both at room temperature and 300 °C (blue and red curves in Figures S1a and S1b) leads to the shift of these two peaks to lower binding energies by ~ 0.1 eV due to a change of the InAs surface band bending. There is no evident change in the As and In photoemission lines shape and FWHM, indicating no obvious difference in the chemical state of the substrate surface before and after the formation of the Bi adlayers. However, it should be noted that the electron mean free path for the most surface sensitive In3d line at given X-ray photon energy of 1486.71 eV is about 2 nm, and the contribution of the surface component in the observed spectra may be negligible. The binding energy of Bi-4$f_{7/2}$ line (figure S1c) of 156,95 eV is close to the value for metallic Bi. According to Ref. [1] bismuth deposition on the InAs(111)A surface at room temperature induces the formation of Bi-In and Bi-As chemical bonds at the interface. The formation of Bi-As chemical bonds usually leads to a shift of the Bi XPS lines to higher binding energies, while the formation of Bi-In bonds shifts the Bi lines to lower binding energies. Indeed, as seen in figure S1c there is an additional component with a binding energy of 157.8 eV in the Bi-4$f_{7/2}$ XPS spectra for the room-temperature grown Bi film, which can be attributed to the formation of Bi-As bonds [1]. The position of the Bi-In component is close to that of the metallic Bi-Bi, and it is difficult to separate their contributions. The Bi-4$f_{7/2}$ XPS spectra for Bi-dimer/InAs(111)-$2\sqrt{3} \times 3$ surface has symmetric shape and peak position of 157.1 eV. The observed 0.15 eV shift of the Bi-4$f_{7/2}$ line to higher binding energies is explained neither by the formation of additional Bi-In bonds nor by the formation of Bi-As bonds since its value is significantly less than the chemical shift due to the formation of Bi-As bonds and its direction is opposite to the shift expected for the formation of Bi-In bonds. The formation of InBi alloy at the surface is also excluded, since it should induce large chemical shift of both In and Bi XPS lines to lower binding energies as is shown by the green lines in Figures S1b and S1c for the InBi single crystal (001) surface. The formation of the InBi Figure S1b,c shows in green the position of the XPS In-3$d_{5/2}$ and Bi-4$f_{7/2}$ lines measured on the surface of the InBi. Conversely, one can suggest the formation of the weakly bonded Bi film on InAs surface in the case of the Bi-dimer/InAs(111)-$2\sqrt{3} \times 3$ system. The effective thickness of the Bi film in the Bi-dimer/InAs(111)-$2\sqrt{3} \times 3$ system was estimated from the relative intensity of the Bi-4f XPS line and the attenuation of the InAs substrate XPS lines to be ~ 3.5 Å. We found no obvious difference in the XPS spectra measured on the surface of the type A and type B samples.

S2. ANGLE-RESOLVED PHOTOELECTRON SPECTROSCOPY (ARPES) MEASUREMENTS OF PRISTINE INAS(111)-2×2 PHASE

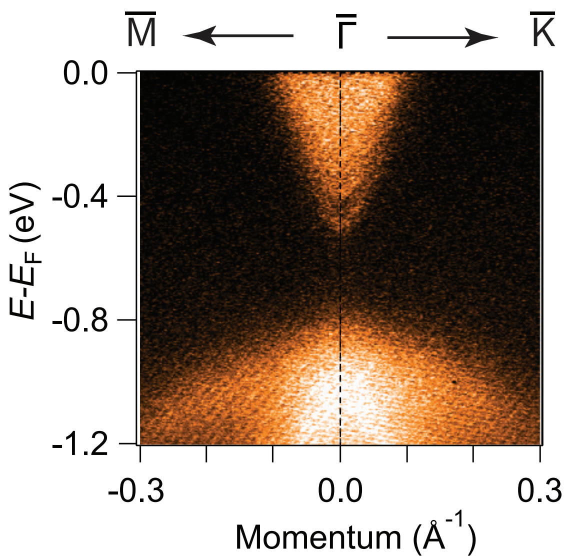

FIG. S2. The ARPES spectrum of pristine InAs(111)-2×2 phase measured at 78 K in the vicinity of the $\bar{\Gamma}$ point.

[1] L. Nicolai, J.-M. Mariot, U. Djukic, W. Wang, O. Heckmann, M. C. Richter, J. Kanski, M. Leandersson, T. Balasubramanian, J. Sadowski, J. Braun, H. Ebert, I. Vobornik, J. Fujii, J. Minar, and K. Hricovini, Bi ultra-thin crystalline films on InAs(111)A and B substrates: a combined core-level and valence-band angle-resolved and dichroic photoemission study, New Journal of Physics **21**, 123012 (2019).