Observation of quantum-tunnelling-modulated spin texture in ultrathin topological insulator Bi$_2$Se$_3$ films

Madhab Neupane$^1$, Anthony Richardella$^2$, Jaime Sánchez-Barriga$^3$, SuYang Xu$^1$, Nasser Alidoust$^1$, Ilya Belopolski$^1$, Chang Liu$^1$, Guang Bian$^1$, Duming Zhang$^2$, Dmitry Marchenko$^{3,4}$, Andrei Varykhalov$^3$, Oliver Rader$^3$, Mats Leandersson$^5$, Thiagarajan Balasubramanian$^5$, Tay-Rong Chang$^6$, Horng-Tay Jeng$^{6,7}$, Susmita Basak$^8$, Hsin Lin$^9$, Arun Bansil$^8$, Nitin Samarth$^2$ & M. Zahid Hasan$^{1,10}$

Understanding the spin-texture behaviour of boundary modes in ultrathin topological insulator films is critically essential for the design and fabrication of functional nanodevices. Here, by using spin-resolved photoemission spectroscopy with p-polarized light in topological insulator Bi$_2$Se$_3$ thin films, we report tunnelling-dependent evolution of spin configuration in topological insulator thin films across the metal-to-insulator transition. We report a systematic binding energy- and wavevector-dependent spin polarization for the topological surface electrons in the ultrathin gapped-Dirac-cone limit. The polarization decreases significantly with enhanced tunnelling realized systematically in thin insulating films, whereas magnitude of the polarization saturates to the bulk limit faster at larger wavevectors in thicker metallic films. We present a theoretical model that captures this delicate relationship between quantum tunnelling and Fermi surface spin polarization. Our high-resolution spin-based spectroscopic results suggest that the polarization current can be tuned to zero in thin insulating films forming the basis for a future spin-switch nanodevice.

1 Joseph Henry Laboratory, Department of Physics, Princeton University, Princeton, New Jersey 08544, USA. 2 Department of Physics, The Pennsylvania State University, University Park, Pennsylvania 16802, USA. 3 Helmholtz-Zentrum Berlin für Materialien und Energie, Elektronenspeicherring BESSY II, Albert-Einstein-Strasse 15, D-12489 Berlin, Germany. 4 Physikalische und Theoretische Chemie, Freie Universität Berlin, Takustraße 3, 14195 Berlin, Germany. 5 MAX-lab, PO Box 118, S-22100 Lund, Sweden. 6 Department of Physics, National Tsing Hua University, Hsinchu 30013, Taiwan. 7 Institute of Physics, Academia Sinica, Taipei 11529, Taiwan. 8 Department of Physics, Northeastern University, Boston, Massachusetts 02115, USA. 9 Graphene Research Centre, Department of Physics, National University of Singapore, Singapore 117542, Singapore. 10 Princeton Center for Complex Materials, Princeton University, Princeton, New Jersey 08544, USA. Correspondence and requests for materials should be addressed to M.Z.H. (email: mzhasan@princeton.edu).
A three-dimensional (3D) topological insulator (TI) is a non-trivial phase of matter that acts as an electrical insulator in the bulk but can conduct a spin-polarized current on the surface\(^1\)–\(^{19}\). These topological surface states are characterized by a Dirac-cone-like energy–momentum dispersion relation. The novel electronic structure of TIs can be manipulated to realize various novel quantum phenomena such as spin-galvanic effects, dissipationless spin currents or neutral half-fermions for quantum information storage devices\(^{20}–^{25}\). The magnitude and wavevector dependence of the spin polarization of electrons and holes are among the most important key ingredients in considerations for the design of any functional device. However, such developments have been limited because of the residual bulk conductance in currently available materials, which overwhelms the surface contribution. In addition, scattering from the extrinsic bulk states leads to the reduction of spin polarization of the surface states. One promising route to minimize bulk conductance and thus improve effective spin polarization is to work with ultrathin films where the surface-to-volume ratio is significantly enhanced\(^{26,27}\) and surface current can potentially dominate. On the other hand, in this limit the desired spin polarization of the surface states is kinematically reduced near the metal-to-insulator transition in the ultrathin films where the spin behaviour is not known to this date\(^{10,11,15,18,19}\).

To date, no systematic spin-sensitive spectroscopic experimental study has been reported in the ultrathin limit across the metal-to-insulator transition, despite the direct relevance of spins in ultrathin film limits for nanodevice fabrication as well as the potential discovery of novel topological phenomena. Studying the spin polarization in the ultrathin limit is further important to experimentally demonstrate the theoretically predicted tunable Berry’s phase in TI thin films\(^{28}\). Systematic mapping of the surface spin texture in energy, momentum space and its thickness dependence is essential to understand and interpret many transport experiments on thin film TIs that are of core interest in the current TI research. We report a systematic spin-resolved, angle-resolved photoemission spectroscopy (SR-ARPES) and spin-integrated ARPES measurements on ultrathin Bi\(_2\)Se\(_3\) thin films for the first time. Our measurements reveal that the spin polarization is large for larger wavevectors, and the polarization magnitude increases with reduced tunnelling, and its magnitude saturates to the bulk limit at a faster rate at large electron momenta.

We observe strongly binding energy- and wavevector-dependent spin polarization for the topological surface electrons in the ultrathin gapped-Dirac-cone limit, which experimentally shows that the Dirac gap opening and the thickness-dependent topological phase transition are a result of the quantum tunnelling between the two oppositely spin-textured topological surface states. These unique spin features of ultrathin films, evidently distinct from the 3D TI, open up new possibilities for devices not possible with bulk TIs.

**Results**

**Sample characterization.** Spectroscopic measurements were performed on large ultrathin Bi\(_2\)Se\(_3\) films prepared using the Molecular Beam Epitaxy (MBE) method on GaAs(111)A substrates (Fig. 1a). Each crystal layer of Bi\(_2\)Se\(_3\) is constituted of five atomic layers (QL), namely Se-Bi-Se-Bi-Se, which is called quintuple layer (QL) with the thickness of \(\sim 1\) nm (ref. 6). Our MBE films grow in a self-organized quintuple layer by quintuple-layer mode, and high-quality atomically smooth films can be obtained with the desired thickness (also see Supplementary Figs 1–7 and Supplementary Notes 1–3). A compositional layout of the film used in our measurements is shown in Fig. 1b. To protect the surface from contamination, about 40-nm Se capping is used on the top of ultrathin Bi\(_2\)Se\(_3\) films. To expose the surface, the films were transferred into the ARPES chamber and heated to 250 °C at pressures lower than 1 × 10\(^{-9}\) Torr about an hour, which blows off the Se-capping layer. Figure 1c shows the ARPES core level spectroscopy measurement of the ultrathin film before and after decapping of the Se layer. Before decapping, only Se peaks are visible while both Se and Bi peaks are observed after the decapping process, which proves that the Se capping works well in the ultrathin film TI system. Thin films are characterized with atomic force microscopy (see Fig. 1d) and show that the root mean square (r.m.s.) surface roughness on these films is in the order of \(\sim 0.2\) nm, which confirms the high quality of the films used in our measurements. The transport measurements of the Se-capped ultrathin films result in carrier concentration, mobility and resistivity in the order of \(1 \times 10^{19} \text{cm}^{-3}\), \(1.270 \text{cm}^{2}\text{V}^{-1}\text{s}^{-1}\) and 0.30 mOhm cm, respectively (also see Supplementary Fig. 1 and Supplementary Note 1).

**Geometry of the spectroscopic measurements.** We used p-polarized light for our ARPES measurements. The photons approach the sample surface with an angle of incident (\(\theta\)) 45 degree with the sample normal and our samples are aligned along \(\Gamma - \text{K}\) momentum space cut (Fig. 2a) for spin–ARPES measurements. The surface wavevector-dependent spin polarization is obtained using a Mott polarimeter (Fig. 2a), which measures two orthogonal spin components of a photoemitted electron\(^{29,30}\). In the polarimeter, a gold foil was used as a scattering source to generate an asymmetry of high energy photoelectrons into different divergent spin states (see refs 29,30 for details). Each orthogonal spin-polarization component is selected by the orientation of a scattering plane defined by the incident beam direction of the photoelectron on the gold foil and the orientation of two electron detectors mounted on each side of the foil. For this experiment, the detector configuration was set in a way that the two spin components correspond to the in-plane and out-of-plane directions of the (111) plane of the sample.

**Thickness- and wavevector-dependent spin polarization.** We present high-resolution spin-integrated ARPES data and corresponding energy distribution curves (EDCs) along the high-symmetry line \(\Gamma - \text{K}\) for 1QL, 3QL, 4QL, 6QL and 7QL Bi\(_2\)Se\(_3\) films in Fig. 2c,d. As long as the thickness of the film is comparable to the decay length of the surface states into the bulk, there is a spatial overlap between the top and bottom surface states resulting in an energy gap at the time-reversal invariant point (\(\Gamma\) point). As expected theoretically\(^{26,27}\), the energy gap decreases and eventually vanishes for sufficiently thick films, corresponding to the transition from a two-dimensional (2D) gapped system (insulator) to a 3D gapless system (metal)\(^{10,11}\). In particular, the gapless dispersion relation observed in the 7QL film from ARPES measurement indicates that this thickness is above the quantum tunnelling limit. On the basis of experimental observations, we present an illustration of the spin configuration for 3QL (insulator; see Supplementary Figs 2 and 3 and Supplementary Note 2) and 7QL (metal) ultrathin Bi\(_2\)Se\(_3\) films in Fig. 2b. It is important to note that a wide range of electronic structures have been reported in ultrathin TI films grown by MBE depending on the nature of the substrates used\(^{10,11,18}\). Different substrates result in different potential jumps from the substrate (the bottom surface of the ultrathin film) to the vacuum (the top surface of the film). When the potential is large, the Dirac point energy of the bottom surface is offset with respect to that of the top surface. Such Dirac point energy offset is observed to cause
The right-handed contribution of the bottom surface to the spin polarization must increase to effectively cancel the left-handed helical spin texture of the top surface. The calculation suggests that the relative contribution from the top surface systematically increases with film thickness.

Model calculation. First-principles theoretical modelling of the spin-polarization behaviour in thin films is presented in Fig. 4b,c. In the calculations, symmetric slabs are used to simulate the thickness of films. While a gapless spin-polarized Dirac-cone is seen on the surface of a semi-infinite crystal of the TI Bi$_2$Se$_3$, a gap is found to open at the Dirac point for thin films because of a finite tunnelling amplitude between the two sides of the slab surface in our calculations. The tunnelling amplitude increases as the thickness decreases, causing the gap to increase and spin polarization to decrease in the gap region. In the calculation we consider the electron attenuation length ($\lambda$) because of the scattering processes since only electrons near the surface are able to reach the top of the surface and escape into the vacuum as in the measurement condition. Indeed, the spin polarization obtained by ARPES reflects the spin texture of the states associated with top surface rather than the bottom surface. The calculated spin expectation value for the electrons that can escape from the sample is $\langle S \rangle_{\text{atom}} \times \exp(-d_{\text{atom}}/\lambda)$, where $d_{\text{atom}}$ is the
distance of an atom to the top surface, and the \( \langle S \rangle_{\text{atom}} \) is the spin expectation value for each atom. The contribution from each atom is weighted by \( \exp \left( -\frac{1}{C_0 d_{\text{atom}}} \right) \), which reduces the contribution from the bottom layer. Figure 4b,c shows the calculated results with \( l = 8 \, \text{Å} \), which agrees excellently with our experimental observation (see Supplementary Methods for details related to calculations).

**Discussion**

It is important to note that the maximum spin polarization observed in the bulk limit is only \( \sim 40\% \) (Fig. 4c), whereas the original ideal theoretical limit is that of nearly 100\% without considering any specific material system\(^{31}\). In real TI materials, the strong spin–orbit interaction entangles the spin and orbital momenta of different atomic types, resulting in the reduction of spin polarization\(^{32}\). Specifically, the low-energy states in Bi\(_2\)Se\(_3\) arise from \( p \)-orbitals of Bi (6\( p^3 \)) and Se (4\( p^4 \)), mostly \( p_z \) levels of Bi and Se\(^{6,12}\). The dominance of the \( p_z \) orbitals in the topological surface states is further suggested by our circular dichroism measurements (see Supplementary Fig. 4; Supplementary Note 2 and Supplementary Methods). The spin–orbit coupling mixes spin and orbital angular momenta while preserving the total

---

**Figure 2 | Spin-texture versus quantum tunneling in ultrathin Bi\(_2\)Se\(_3\)**

(a) Experimental geometry used in our measurements. (b) Visualization of the contrasting spin configurations in 3QL (insulator) and 7QL (metal) thin films. The dumbbell signs indicate that the current experimental geometry mainly probes the \( p_z \) orbitals of Bi and Se. (c) High-resolution ARPES measurements on ultrathin films of Bi\(_2\)Se\(_3\): \( E - k \) band dispersion images for 1QL, 3QL, 4QL, 6QL and 7QL of Bi\(_2\)Se\(_3\) films taken near the \( \Gamma \) point along \( \Gamma - K \) high-symmetry direction. The spin configuration is noted on the plots. These spectra are measured with photon energy of 60 eV. (d) The corresponding EDCs. The EDC through the \( T \) point (solid black curve) is highlighted.
angular momentum\textsuperscript{6,12}. The hybridization of orbitals in Bi and Se together with the entanglement of their spins contribute to the reduction of net spin polarization in real materials. Moreover, under the experimental geometry used in our measurement with p-polarized light (which is most sensitive to $p_z$ orbitals and most reflective of initial ground state of the wavefunction), the penetration depth of the ARPES experiment (three to five atomic layers maximum), the experimental observation of spin polarization is well agreed with recent theoretical calculations\textsuperscript{33–35}.

Most importantly, our systematic spin spectroscopy results suggest that ultrathin films can serve as the basis for making qualitatively new devices, not possible with much-studied conventional 3D bulk TIs, despite the reduction of the polarisation magnitude or forms the physical basis for a spin device, among many other new application possibilities suggested by our observations of the fundamental spin modulation behaviour in ultrathin films.

**Methods**

**Sample growth and transport measurements.** Ultrathin Bi$_2$Se$_3$ films used for this study were synthesized by MBE on a GaAs(111)A substrate with a ZnSe buffer layer. Details of sample preparation are described elsewhere\textsuperscript{36,37}. To protect the surface from oxidation, a thick Se-capping layer was deposited on the Bi$_2$Se$_3$ thin film immediately after growth. To achieve the clean Bi$_2$Se$_3$ surface required for photoemission measurements, thin films were heated up inside the ARPES chamber to 250 °C under vacuum better than 1 Torr to remove the Se-capping layer on top of ultrathin Bi$_2$Se$_3$. Hall bars fabricated from the Se-capped samples had carrier concentrations in the 1–2 × 10$^{19}$ cm$^{-3}$ range. Transport measurements were carried out on Hall bars of the 5 and 6QL thick films with the
Se-capping layer in place by standard photolithography and dry etching and were measured at 4.2 K.

High-resolution ARPES measurements. High-resolution spin-integrated ARPES measurements were performed with 29–64 eV photon energy on beamlines 10.0.1 and 12.0.1 at the Advanced Light Source in Lawrence Berkeley National Laboratory. Both endstations were equipped with a Scienta hemispherical electron analyser (see VG Scienta manufacturer website (http://www.vgscienta.com/) for instrument specifications). The typical energy and momentum resolutions were 15 meV and 1% of the surface Brillouin zone, respectively, for spin-integrated measurements.

Spin-resolved ARPES measurements. Spin-resolved ARPES measurements were performed at the UE112-PGM1 beamline at Bessy II in Berlin, Germany, and the I3 beamline at Maxlab in Lund, Sweden, using classical Mott detectors and photon energies 55–60 and 8–20 eV, respectively. The typical energy and momentum resolutions were 100 meV and 3% of the surface Brillouin zone for spin-resolved measurements. All the SR spectra presented are measured in BESSY II unless it is specified.

Theoretical calculations. The first-principles calculations for spin polarizations of ultrathin films are based on the generalized gradient approximation using the full-potential projected augmented wave method as implemented in the VASP package. The 1QL, 3QL, 4QL, 6QL, and 12QL slab models with a vacuum thickness larger than 10 Å are used in this work. The electronic structure calculations were performed over 11 × 11 × 1 Monkhorst-Pack k-mesh with the spin–orbit coupling included self-consistently.

References
1. Hasan, M. Z. & Kane, C. L. Colloquium: topological insulators. Rev. Mod. Phys. 82, 3045–3067 (2010).
2. Moore, J. E. The birth of topological insulators. Nature 464, 194–198 (2010).
3. Qi, X.-L. & Zhang, S.-C. Topological insulators and superconductors. Rev. Mod. Phys. 83, 1057–1110 (2011).
4. Fu, L. & Kane, C. L. Topological insulators with inversion symmetry. Phys. Rev. B 76, 045302 (2007).
5. Hsieh, D. et al. A topological Dirac insulator in a quantum spin Hall phase. Nature 452, 970–974 (2008).
6. Xia, X. et al. Observation of a large-gap topological-insulator class with a single Dirac cone on the surface. Nat. Phys. 5, 398–402 (2009).
7. Chen, Y.-L. et al. Massive Dirac Fermion on the surface of a magnetically doped topological insulator. Science 329, 659–662 (2010).
8. Xu, S.-Y. et al. Observation of a topological crystalline insulator phase and topological phase transition in Pb1–xSnxTe. Nat. Commun. 3, 1192 (2012).
9. Neupane, M. et al. Surface electronic structure of the topological Kondo-insulator candidate correlated electron system SmB6. Nat. Commun. 4, 2991 (2013).
10. Zhang, Y. et al. Crossover of the three-dimensional topological insulator Bi2Se3 to the two-dimensional limit. *Nat. Phys.*, 6, 584–588 (2010).

11. Sakamoto, Y. et al. Spectroscopic evidence of a topological quantum phase transition in ultrathin Bi2Se3 films. *Phys. Rev. B*, 81, 165432 (2010).

12. Zhang, H. et al. Topological insulators in Bi2Se3, Bi2Te3, and Sb2Te3 with a single Dirac cone on the surface. *Nat. Phys.*, 5, 438–442 (2009).

13. Hsieh, D. et al. A tunable topological insulator in the spin helical Dirac transport regime. *Nature* **460**, 1101–1105 (2009).

14. Liu, C.-X. et al. Oscillatory crossover from two-dimensional to three-dimensional topological insulators. *Phys. Rev. B*, 81, 041307(R) (2010).

15. Jiang, Y. et al. Landau quantization and the thickness limit of topological insulator thin films of Sb2Te3. *Phys. Rev. Lett.* **108**, 016401 (2012).

16. Taskin, A. A. et al. Manifestation of topological protection in transport properties of epitaxial Bi2Se3 thin films. *Phys. Rev. Lett.* **109**, 066803 (2012).

17. Neupane, M. et al. Topological surface states and Dirac point tuning in ternary topological insulators. *Phys. Rev. B*, 85, 235406 (2012).

18. Berntsen, M. H. et al. Direct observation of decoupled Dirac states at the interface between topological and normal insulators. *Phys. Rev. B*, 88, 195132 (2013).

19. Chang, C.-Z. et al. Growth of topological insulator Bi2Se3 on insulating substrate. *Spin* **01**, 21–25 (2011).

20. Garate, I. & Franz, M. Inverse spin-galvanic effect in the interface between a topological insulator and a ferromagnet. *Phys. Rev. Lett.* **104**, 146802 (2010).

21. Qi, X.-L. et al. Inducing a magnetic monopole with topological surface states. *Science* **323**, 1184–1187 (2009).

22. Essin, A. et al. D. Moel, F. D. M. and B. V. D. M. Magnetoelectric polarizability and axion electrodynamics in crystalline insulators. *Phys. Rev. Lett.* **102**, 146805 (2009).

23. Yu, R. et al. Quantized anomalous Hall effect in magnetic topological insulators. *Science* **329**, 61–64 (2010).

24. Linder, J. et al. Unconventional superconductivity on a topological insulator. *Phys. Rev. Lett.* **104**, 067001 (2010).

25. Fu, L. & C. L. Majorana fermion edge modes with charge transport. *Phys. Rev. Lett.* **102**, 216403 (2009).

26. Linder, J., Yokoyama, T. & Sudbo, A. Anomalous finite size effects on surface states in the topological insulator Bi2Se3. *Phys. Rev. B* **80**, 205401 (2009).

27. Lu, H.-Z. et al. Massive Dirac fermions and spin physics in an ultrathin film of topological insulator. *Phys. Rev. B*, 81, 115407 (2010).

28. Lu, H.-Z. et al. Competition between weak localization and antilocalization in topological surface states. *Phys. Rev. Lett.* **107**, 076801 (2011).

29. Berntsen, M. H. A spin- and angle-resolving photoelectron spectrometer. *Rev. Sci. Instrum.* **81**, 035104 (2010).

30. Díl, J. H. Spin and angle resolved photoemission on non-magnetic low-dimensional systems. *J. Phys. Condens. Matter.* **21**, 403001 (2009).

31. Kane, C. L. & Mele, E. J. Z. Topological order and the quantum spin Hall effect. *Phys. Rev. Lett.* **95**, 146802 (2005).

32. Yazeyev, O. V., Moore, J. E. & Louie, S. G. Spin polarization and transport of surface states in the topological insulators Bi2Se3 and Bi2Te3 from first principles. *Phys. Rev. Lett.* **105**, 266806 (2010).

33. Zhou, H. Z. et al. Layer-by-layer entangled spin-orbital texture of the topological surface state in Bi2Se3. *Phys. Rev. Lett.* **110**, 216401 (2013).

34. Park, S. R. et al. Chiral orbital-angular momentum in the surface states of Bi2Se3. *Phys. Rev. Lett.* **108**, 046805 (2012).

35. Sánchez-Barriaga, J. et al. Photoemission of Bi2Se3 with circularly polarized light: probe of spin polarization or means for spin manipulation? *Phys. Rev. X* **4**, 011046 (2014).

36. Xu, S.-Y. et al. Hedgehog spin texture and Berry’s phase tuning in a magnetic topological insulator. *Nat. Phys.* **8**, 616–622 (2012).

37. Zhang, D. et al. Interplay between ferromagnetism, surface states, and quantum corrections in a magnetically doped topological insulator. *Phys. Rev. B* **86**, 205127 (2012).

38. Perdew, J. P., Burke, K. & Ernzerhof, M. Generalized gradient approximation made simple. *Phys. Rev. Lett.* **77**, 3865 (1996).

39. Blochl, P. E. Projector augmented-wave method. *Phys. Rev. B* **50**, 17953 (1994).

40. Kresse, G. & Joubert, J. From ultrasoft pseudopotentials to the projector augmented-wave method. *Phys. Rev. B* **59**, 1758 (1999).

41. Kress, G. & Hafner, J. Ab-initio molecular dynamics for open-shell transition metals. *Phys. Rev. B* **48**, 13115 (1993).

Acknowledgements

Sample growth and ARPES characterization are supported by US DARPA (N66001-11-1-4110). The work at the Princeton University and Princeton-led synchrotron X-ray-based measurements and the related theory at the Northeastern University are supported by the Office of Basic Energy Science, US Department of Energy (grants DE-FG02-05ER46200, AC03-76SF00098 and DE-FG02-07ER46352). M.Z.H. acknowledges visiting-scientist support from the Lawrence Berkeley National Laboratory and additional support from the A.P. Sloan Foundation. The spin-resolved and spin-integrated photoemission measurements using synchrotron X-ray facilities are supported by the Swedish Research Council, the Knut and Alice Wallenberg Foundation, the German Federal Ministry of Education and Research, and the Basic Energy Sciences of the US Department of Energy. Theoretical computations are supported by the US Department of Energy (DE-FG02-07ER46352 and AC03-76SF00098) as well as the National Science Council and Academia Sinica in Taiwan, and benefited from the allocation of supercomputer time at NERSC and Northeastern University’s Advanced Scientific Computation Center. H.L. acknowledges the Singapore National Research Foundation for the support under NRF Award No. NRF-NRFIF2013-03. T.R.C. and H.T.I. are supported by the National Science Council, Taiwan. H.T.I. also thanks NCHC, CINC-NTU and NCTS, Taiwan, for technical support. We also thank S.-K. Mo and A. Fedorov for beamline assistance on spin-integrated photoemission measurements (supported by DE-FG02-05ER46200) at the Lawrence Berkeley National Laboratory (The synchrotron facility is supported by the US DOE).

Author contributions

M.N., J.S.-B., and S.K. performed the experiments with assistance from N.A., I.B., C.L., G.B., D.M., A.V., O.R., M.L., T.B. and M.Z.H.; A.R., D.Z. and N.S. provided thin film assistance on spin-integrated photoemission measurements using synchrotron X-ray facilities are supported by the Swedish Research Council, the Knut and Alice Wallenberg Foundation, the German Federal Ministry of Education and Research, and the Basic Energy Sciences of the US Department of Energy. Theoretical computations are supported by the US Department of Energy (DE-FG02-07ER46352 and AC03-76SF00098) as well as the National Science Council and Academia Sinica in Taiwan, and benefited from the allocation of supercomputer time at NERSC and Northeastern University’s Advanced Scientific Computation Center. H.L. acknowledges the Singapore National Research Foundation for the support under NRF Award No. NRF-NRFIF2013-03. T.R.C. and H.T.I. are supported by the National Science Council, Taiwan. H.T.I. also thanks NCHC, CINC-NTU and NCTS, Taiwan, for technical support. We also thank S.-K. Mo and A. Fedorov for beamline assistance on spin-integrated photoemission measurements (supported by DE-FG02-05ER46200) at the Lawrence Berkeley National Laboratory (The synchrotron facility is supported by the US DOE).

Additional information

Supplementary Information accompanies this paper at http://www.nature.com/naturecommunications

Competition of financial interests: The authors declare no competing financial interests.

Reprints and permission information is available online at http://npg.nature.com/reprintsandpermissions/

How to cite this article: Neupane, M. et al. Observation of quantum-tunnelling-modulated spin texture in ultrathin topological insulator Bi2Se3 films. *Nat. Commun.* 5:3841 doi: 10.1038/ncomms4841 (2014).