A novel artificial condensed matter lattice and a new platform for one-dimensional topological phases

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Engineered lattices in condensed matter physics, such as cold-atom optical lattices or photonic crystals, can have properties that are fundamentally different from those of naturally occurring electronic crystals. We report a novel type of artificial quantum matter lattice. Our lattice is a multilayer heterostructure built from alternating thin films of topological and trivial insulators. Each interface within the heterostructure hosts a set of topologically protected interface states, and by making the layers sufficiently thin, we demonstrate for the first time a hybridization of interface states across layers. In this way, our heterostructure forms an emergent atomic chain, where the interfaces act as lattice sites and the interface states act as atomic orbitals, as seen from our measurements by angle-resolved photoemission spectroscopy. By changing the composition of the heterostructure, we can directly control hopping between lattice sites. We realize a topological and a trivial phase in our superlattice band structure. We argue that the superlattice may be characterized in a significant way by a one-dimensional topological invariant, closely related to the invariant of the Su-Schrieffer-Heeger model. Our topological insulator heterostructure demonstrates a novel experimental platform where we can engineer band structures by directly controlling how electrons hop between lattice sites.

INTRODUCTION

While crystals found in nature offer a great richness of phenomena, recent progress in physics has also been driven by the study of engineered systems, where key material parameters or Hamiltonian matrix elements can be directly controlled in experiments to give rise to novel emergent properties. For example, the Haldane model for a Chern insulator was recently realized in an optical lattice of ultracold atoms (1), and a Weyl semimetal was observed in a photonic crystal with a double-gyroid lattice structure (2). Each advance required engineering a specific ultracold-atom optical lattice or photonic crystal. In both cases, these engineered lattices gave rise to an emergent band structure, analogous to the band structure formed by electrons in a crystal lattice, but with distinct properties and highly tuned parameters that gave rise to a novel phase of matter.

Here, we demonstrate an emergent band structure in a novel type of condensed matter lattice based on topological insulators (3, 4). Specifically, we stack together layers of topological and trivial insulators to create a one-dimensional topological insulator heterostructure. Each interface in the heterostructure hosts a set of topologically protected interface states that hybridize with each other across the layers, giving rise to a superlattice band structure where the interfaces play the role of lattice sites, the topological surface states play the role of atomic orbitals, and the heterostructure acts as a one-dimensional atomic chain. Such a superlattice band structure is emergent in the sense that it arises only when many layers are stacked together, in the same way that an ordinary band structure arises as an emergent property of a crystal lattice. A similar heterostructure has previously been proposed as a simple theoretical model for a Weyl semimetal (5). Here, we experimentally realize and directly observe the first superlattice band structure of this type. We use molecular beam epitaxy to build the topological insulator heterostructure, and we use angle-resolved photoemission spectroscopy (ARPES) to study its band structure. By adjusting the pattern of layers in the heterostructure, we realize both a topological and a trivial phase, demonstrating that we can tune our system through a topological phase transition. Our work may lead to the realization of novel three-dimensional and one-dimensional symmetry-protected topological phases. Moreover, our topological insulator heterostructure provides a highly tunable emergent band structure in a true electron system, relevant for transport experiments and device applications.

We first provide a more detailed introduction to the topological insulator heterostructure and argue that our system forms a novel type of condensed matter lattice. A topological insulator heterostructure is built up of alternating layers of topologically trivial and nontrivial insulators. Here, we use Bi$_x$Se$_{3-x}$ as the topological insulator and In$_x$Bi$_{2-x}$Se$_3$ as the trivial insulator (see Fig. 1A). We note that heterostructures of Bi$_x$Se$_3$ and In$_x$Se$_3$ have already been successfully synthesized (6–8). Bulk Bi$_x$Se$_3$ has a nontrivial $Z_2$ invariant, $v_0 = 1$ (9, 10). Under doping by In, bulk Bi$_x$Se$_3$ undergoes a topological phase transition to a topologically trivial phase with $Z_2$ invariant, $v_0 = 0$ (11). In all samples considered here, all In$_x$Bi$_{2-x}$Se$_3$ layers are well into the trivial phase. As a result, the topological invariant flips back and forth from layer to layer in the heterostructure, so that each interface hosts a set of topological interface states (3, 4, 12–15). Next, we note that these interface states have a finite penetration depth into the bulk of the crystal, so that for sufficiently thin
layers, the topological interface states hybridize with each other across the layers and will be subject to an energy level repulsion \((16,\ 17)\). This hybridization can be captured by a hopping amplitude \(t\) across the topological layer and a hopping amplitude \(t'\) across the trivial layer. In this way, the topological insulator heterostructure can be viewed as a toy model in the study of one-dimensional topological phases. The model has two carbon atoms per unit cell, with one orbital each and with hopping amplitudes \(t\) and \(t'\) associated with the double and single carbon bonds. In the topological insulator heterostructure, the topological and trivial layers play the role of the double and single carbon bonds. (C) A conventional semiconductor heterostructure consists of an alternating pattern of materials with different band gaps. (D) In a topological insulator heterostructure, the band gaps in adjacent layers are inverted, giving rise to topologically protected Dirac cone interface states between layers. If the layers are thin, then adjacent Dirac cones may hybridize. This hybridization can be described by a hopping amplitude \(t\) across the topological layer and a hopping amplitude \(t'\) across the trivial layer. (E) Illustration of the Dirac cone surface states at each interface in the heterostructure, assuming no hybridization. With a hybridization \(t\), the topological interface states form a superlattice band structure. A standard low-energy electron diffraction pattern (F) and a core-level photoemission spectrum (G) show that the samples are of high quality and that the Se capping layer was successfully removed by in situ heating, exposing a clean sample surface in vacuum.

RESULTS

We next demonstrate that we have observed an emergent superlattice dispersion in our topological insulator heterostructure. We present a systematic study of four different \(\text{Bi}_2\text{Se}_3/\text{In}_{x}\text{Bi}_{2-2x}\text{Se}_3\) superlattices: \(4\text{QL}/4\text{QL}\) 20%, \(4\text{QL}/2\text{QL}\) 25%, \(4\text{QL}/2\text{QL}\) 15%, and \(4\text{QL}/1\text{QL}\) 20% (see Fig. 2, A to D). In this notation, the first parameter refers to the thickness of the \(\text{Bi}_2\text{Se}_3\) layer, the second parameter refers to the thickness of the \(\text{In}_{x}\text{Bi}_{2-2x}\text{Se}_3\) layer; and the percentage refers to the quality of our samples (see Fig. 1, F and G).
energy at $k_{\parallel} = 0$. These curves correspond to a vertical line on the image plot, indicated by the green arrowheads in Fig. 2 (I to L). The gapless surface state, labeled 1 in 4QL/2QL 15% and 4QL/1QL 20%, is entirely inconsistent with a 4QL-thick film of Bi$_2$Se$_3$. Instead, it can only be explained by considering hybridization across the In$_x$Bi$_{2-x}$Se$_3$ layer, demonstrating an emergent superlattice band structure arising from the hopping of Dirac cone interface states within the heterostructure. In addition, a gap opens from 4QL/2QL 15% to 4QL/2QL 25% without any observable bulk band inversion and without time-reversal symmetry breaking. This is an apparent contradiction with the basic theory of $\mathbb{Z}_2$ topological insulators. Again, this result demonstrates a superlattice dispersion. As we increase $t/t'$, the top two lattice sites show larger hybridization, and a gap opens in the Dirac cone on the last site. Because ARPES is only sensitive to the top surface of the heterostructure, we can only observe the increased coupling to the top surface, and not the inversion of the superlattice bands. Our observation of (i) a gapless surface state on a 4QL-thick film of Bi$_2$Se$_3$ and (ii) a gap opening in a Bi$_2$Se$_3$ surface state without apparent band inversion demonstrates that we have observed a superlattice dispersions in our system. In this way, we have shown a novel type of electronic band structure, which arises from a lattice of topological insulator Dirac cones that are allowed to hybridize.

We provide a one-dimensional picture of the topological and trivial phases of the superlattice. We again consider 4QL/1QL 20%, as shown in Fig. 3 (A to D). Above, we argued that 4QL/1QL 20% has a gapless Dirac cone because $t < t'$. We note that the different hopping amplitude arises from the large bulk band gap and large thickness of the Bi$_2$Se$_3$ layer relative to the In$_x$Bi$_{2-x}$Se$_3$ layer, as illustrated in Fig. 3E. Alternatively, we can consider how orbitals pair up and gap out in real space. We see that, in the topological phase, there are two end modes left without a pairing partner (see Fig. 3F). We contrast the topological...
phase with the trivial phase observed in 4QL/1QL 20%, as shown in Fig. 3 (G to J). In this case, $t > t'$, because the trivial phase has a larger band gap than the topological phase, as illustrated in Fig. 3K. Alternatively, the real space pairing leaves no lattice site without a pairing partner (see Fig. 3L). We see that our observation of a topological and trivial phase in a topological insulator heterostructure can be understood in terms of an emergent one-dimensional atomic chain where the termination of the chain is either on a strong or on a weak bond.

We present the ARPES spectra of other compositions in the topological and trivial phase to provide a systematic check of our results. First, we compare 4QL/1QL 10% and 4QL/1QL 15% with 4QL/1QL 20%. The unit cells of these lattices are illustrated in Fig. 4 (A to C). The ARPES spectra are shown in Fig. 4 (D to F). We also show an EDC at $k_x = 0$ in Fig. 4 (G to I), as indicated by the green arrowheads in Fig. 4 (D to F). We find that 4QL/1QL 10% and 4QL/1QL 15% also host a gapless surface state and are also topological. This is expected because we have further decreased the In concentration in the trivial layer, increasing the hybridization $t$ and pushing the sample further into the topological phase. Next, we compare 3QL/3QL 20% with 4QL/4QL 20%. The unit cells are illustrated in Fig. 4 (J to L). The ARPES spectra...
are shown in Fig. 4 (M to O), and an EDC at $k_z = 0$ is shown in Fig. 4 (P to R). We find that 3QL/3QL 20% is also in the trivial phase, with a gap in the surface states larger than that in 4QL/4QL 20%. This is consistent with earlier ARPES studies of single thin films of Bi$_2$Se$_3$ on a topologically trivial substrate (16, 17). For 3QL/3QL 20%, we further observe the same bands at two different photon energies, providing a check that the gapped surface state is not an artifact of low photoemission cross section at a special photon energy. Our results on 4QL/1QL 10%, 4QL/1QL 15%, and 3QL/3QL 20% provide a systematic check of our results.

**DISCUSSION**

Our ARPES spectra show that we have realized the first emergent band structure in a lattice of topological interface states. We further demonstrate that we can tune this band structure through a topological phase transition. We summarize our results by plotting the compositions of Fig. 2 on a phase diagram as a function of $t'/t$ (Fig. 5A). Our results can also be understood in terms of a one-dimensional atomic chain terminated either on a strong or on a weak bond. It is natural to ask whether any rigorous analogy exists between our topological insulator heterostructure and a one-dimensional topological phase (22). An interesting property of our heterostructure is that it naturally gives rise to an approximate chiral symmetry along the stacking direction. In particular, because
the wave function of a topological insulator surface state decays rapidly into the bulk, we expect that the only relevant hybridization in our heterostructure takes place between adjacent interface states. Further, along the stacking direction, the lattice is bipartite. The nearest-neighbor hopping on a bipartite lattice gives rise to an emergent chiral symmetry along the stacking direction. We note that chiral symmetry is required for a number of topological phases in one dimension, including the BDI, CII, DIII, and CI classes, as well as the well-known Su-Schrieffer-Heeger model (20–22). We suggest that future theoretical work could determine whether a one-dimensional topological phase might be realized in our system. Specifically, we might consider topological invariants on the one-dimensional band structure along at fixed . We also note that we may introduce a second nearest-neighbor hopping by using even thinner layers (see Fig. 5B). This may break the chiral symmetry, removing the one-dimensional topological invariant and shifting the surface state out of the superlattice bulk band gap. This would provide a direct experimental signature of the one-dimensional topological invariant. Another application of our topological insulator heterostructure is to break inversion symmetry using a unit cell consisting of four layers of different thicknesses (see Fig. 5C). This may lead to a superlattice band structure of spinful bands. More complicated topological insulator heterostructures may allow us to engineer time-reversal broken one-dimensional topological phases or large degeneracies implemented by fine-tuning hopping amplitudes. Our systematic ARPES measurements demonstrate the first atomic chain of topological insulator surface states. In this way, we provide not only an entirely novel type of condensed matter lattice but also a new platform to engineer band structures in true electron systems by directly controlling how electrons hop between lattice sites.

**MATERIALS AND METHODS**

**Heterostructure growth**

High-quality Bi$_2$Se$_3$/In$_x$Bi$_{2-x}$Se$_3$ heterostructures were grown on 10 mm × 10 mm × 0.5 mm Al$_2$O$_3$ (0001) substrates using a custom-built SVTA MOS-V-2 molecular beam epitaxy system with a base pressure of 2 × 10$^{-10}$ torr. Substrates were cleaned ex situ by 5-min exposure to ultraviolet-generated ozone and in situ by heating to 800°C at an oxygen pressure of 1 × 10$^{-6}$ torr for 10 min. Elemental Bi, In, and Se sources (99.999% pure) were thermally evaporated using effusion cells equipped with individual shutters. In all samples, the first layer of the superlattice was Bi$_2$Se$_3$, 3QL of which was grown at 135°C and annealed to 265°C for the rest of the film growth (23). For In$_x$Bi$_{2-x}$Se$_3$ growth, Bi and In were coevaporated by simultaneously opening both shutters, whereas the Se shutter was kept open at all times during the growth. To accurately determine the concentration of In in the In$_x$Bi$_{2-x}$Se$_3$ layers, source fluxes were calibrated in situ by a quartz crystal microbalance to one of the superlattice bulk band gap. This would provide a direct experimental signature of the one-dimensional topological invariant. Another application of our topological insulator heterostructure is to break inversion symmetry using a unit cell consisting of four layers of different thicknesses (see Fig. 5C). This may lead to a superlattice band structure of spinful bands. More complicated topological insulator heterostructures may allow us to engineer time-reversal broken one-dimensional topological phases or large degeneracies implemented by fine-tuning hopping amplitudes. Our systematic ARPES measurements demonstrate the first atomic chain of topological insulator surface states. In this way, we provide not only an entirely novel type of condensed matter lattice but also a new platform to engineer band structures in true electron systems by directly controlling how electrons hop between lattice sites.

**arpes measurements**

ARPES measurements were carried out using a number of synchrotron light sources, in particular at ANTARES, Synchrotron SOLEIL (Saint-Aubin, France); ADRESS, Swiss Light Source (Villigen, Switzerland); and the HRPES end station of Surface and Interface Spectroscopy, Swiss Light Source; I05, Diamond Light Source (Oxfordshire, U.K.); and CASSIOPEE, SOLEIL. Samples were clamped onto the sample holder using either an Mo or Ta clamp screwed onto the sample base or thin strips of Ta foil spot-welded onto the sample base. In addition to securely mounting the sample, the clamp also provided electrical grounding. The Se capping layer was removed by heating the sample in a vacuum preparation chamber at temperatures between 200°C and 300°C and at pressures lower than 10$^{-6}$ torr for ~1 hour. Following decapping, the quality of the exposed sample surface was checked by low-energy electron diffraction at a typical electron beam energy of 100 V. The presence of sharp Bragg peaks and their sixfold rotation symmetry (Fig. 1H) shows the high quality of the exposed sample surface. ARPES measurements were carried out at pressures lower than 10$^{-10}$ torr at incident photon energies between 15 and 320 eV. We take a short Fermi surface mapping near the center of the Brillouin zone for each sample, at relevant photon energies, to identify the rotation angles corresponding to . This allows us to take - cuts through the point to within a rotation angle of ±0.15°, minimizing the error in our measurement of the band gap due to sample misalignment.

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/3/3/e1501692/DC1

**Estimate of indium diffusion in the heterostructure**

Observation of a topological phase transition in numerics

Fine dependence on indium doping

Comparison with a single thin film of Bi$_2$Se$_3$ detailed analysis of bulk quantum well states

fig. S1. Characterization of In diffusion in the heterostructures.

fig. S2. Topological phase transition in numerics.

fig. S3. Change in Fermi level with In doping.

fig. S4. Dimerized-limit heterostructure and a single thin film of Bi$_2$Se$_3$.

fig. S5. Bulk quantum well states of 4QL/2QL 15% and 4QL/1QL 20%.

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