Supplementary Information for

Layer-Dependent Topological Phase in a Two-Dimensional Quasicrystal and Approximant

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Materials and Methods:

$Ta_{1.6}Te$ Synthesis:

Samples of the $Ta_{1.6}Te$ were prepared *via* reduction of $TaTe_2$ with elemental tantalum at high temperatures. $TaTe_2$ was produced by combining elemental materials (Ta: H.C. Starck, 99.9%, Te: Honeywell Fluka, 99.999%) in the appropriate ratios in an evacuated silica ampoule and heating at 950° C.

For conversion to $Ta_{1.6}Te$, a 1:3 compressed mixture of $TaTe_2$ and Ta was placed in a molybdenum crucible and sealed under argon. Trace iodine was added to promote crystal growth. The mixtures were heated at 1700 ° C for 3-4 hours and cooled to room temperature within an hour. The resulting samples were 100s of μm in lateral dimensions with a gold luster, with excess tantalum. The excess tantalum does not exfoliate and is separated out during preparation of 2D materials. Bulk materials were handled in ambient for only short (t < 1 h) periods of time.

2D Material Preparation:

Two-dimensional (2D) samples were fabricated by mechanical exfoliation of the synthesized bulk materials using polydimethylsiloxane (PDMS) stamps. Ultrathin flakes were identified by optical contrast. The exfoliated materials were then directly transferred onto arbitrary substrates including SiO_2 and SiN TEM grids with a micro-manipulator transfer stage. All fabrication, handing, and transfer of bulk crystals and two-dimensional samples were done in a nitrogen filled glove-box with O_2 and H_2O levels kept below 0.1 ppm. For imaging, samples were transported in vacuum and exposed to air for <<1 min.

52 **(S)TEM Characterization**

53 Transmission electron microscope imaging and electron diffraction were conducted using
54 a JEOL 2010 microscope at 80 kV. Aberration-corrected scanning transmission electron
55 microscope imaging and energy dispersive x-ray spectroscopy were conducted with and FEI
56 Titan3 (60−300) equipped with a SuperX energy dispersive X-ray spectrometry (EDS) system at
57 80 kV. A camera length of 115 mm and a beam current of 50 pA were used for image acquisition.
58 We performed the EDS elemental mapping in the STEM mode at 80 kV with a 5min acquisition
59 time.

60

61 **Theoretical Methods:**

62 Density functional theory (DFT) calculations were performed using the projector
63 augmented wave (PAW) method in the Vienna ab initio Simulation Package (VASP)[1,2] code with
64 the Perdew-Burke-Ernzerhof functionals (PBE).[3] Ta (6s, 5d) and Te (5s, 5p) electrons were treated
65 as valence, and their wavefunctions expanded in plane waves to an energy cutoff of 600 eV.
66 Gamma-centered k-point grids of 2x2x4 (bulk), 2x2x2 (monolayer with vacuum) and 2x2x1
67 (bilayer with vacuum) were used for Brillouin Zone sampling. Spin-orbit coupling (SOC) was
68 included self-consistently. We fixed the lattice parameters and internal coordinates to the
69 experimental values of the *P6mm* structure (a = 19.5, c = 10.3 Å). Structural parameters for
70 $Ta_{21}Te_{13}$ are extracted from the approximant $Ta_{181}Te_{112}$, and manually fitted to the unit cell of
71 $Ta_{21}Te_{13}$. For the monolayer and bilayer calculation, a vacuum of 10 Å was included in the out-of-
72 plane direction. Topological classification was carried out using DFT calculations and the
73 SymTopo package.[4]

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Supplemental Figures

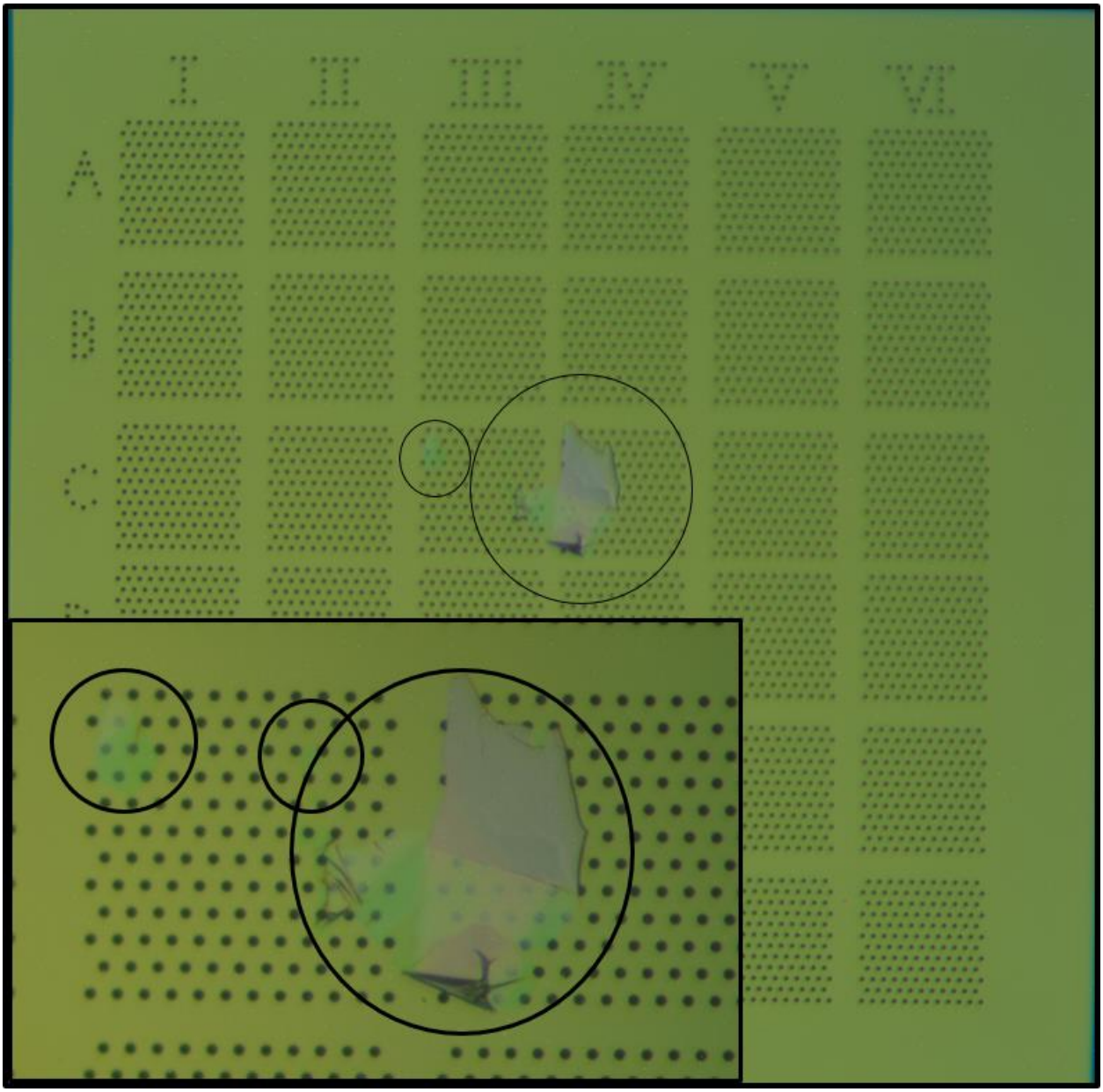

Figure S1. Optical image of exfoliated $Ta_{1.6}Te$ transferred onto SiN TEM grid. Inset: Enlarged section showing exfoliated/ultrathin materials. Each hole 2 μm in diameter.

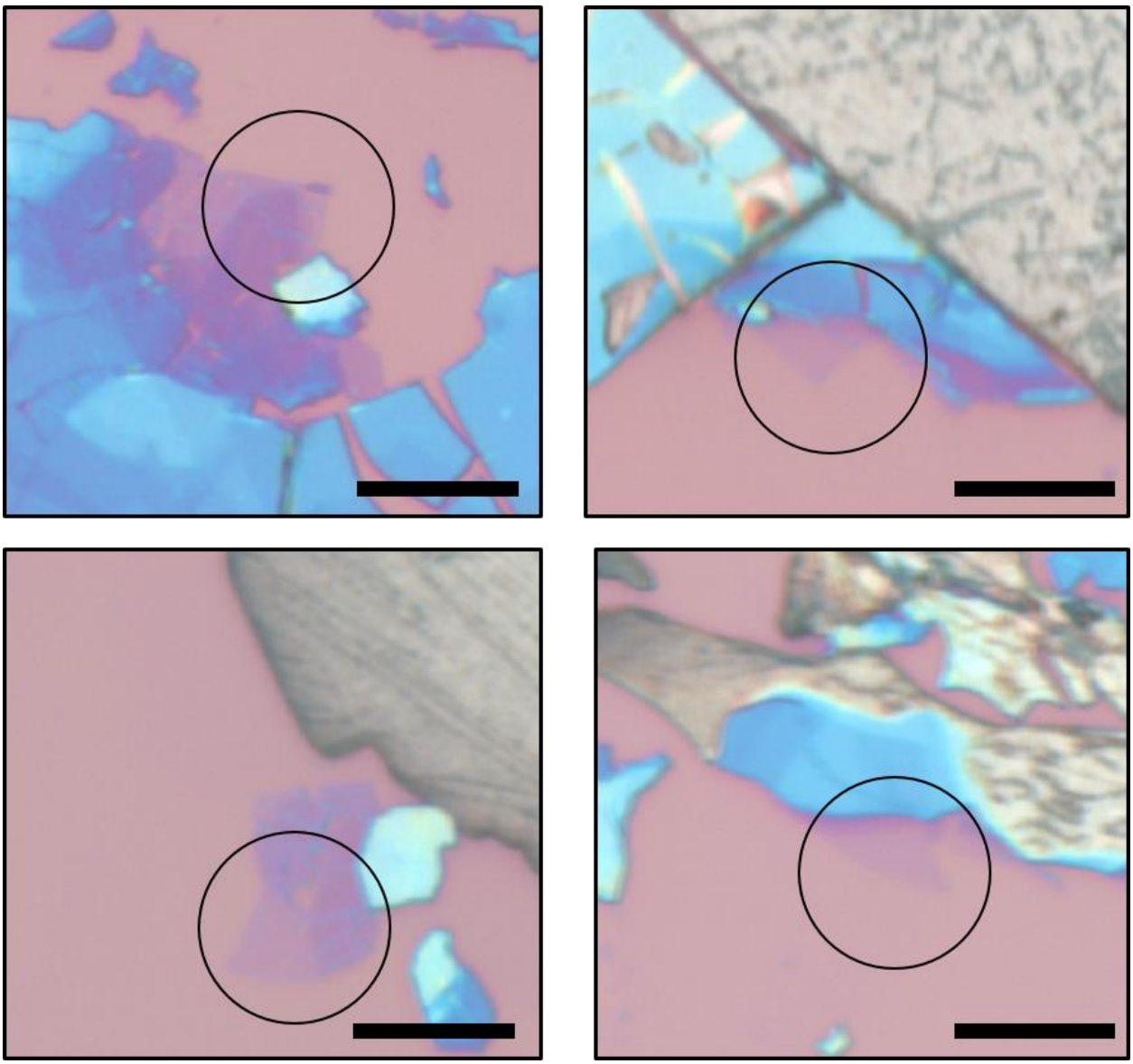

Figure S2. Optical images of exfoliated $Ta_{1.6}Te$. Circles highlight the two-dimensional sections. Scale bars: 10 μm

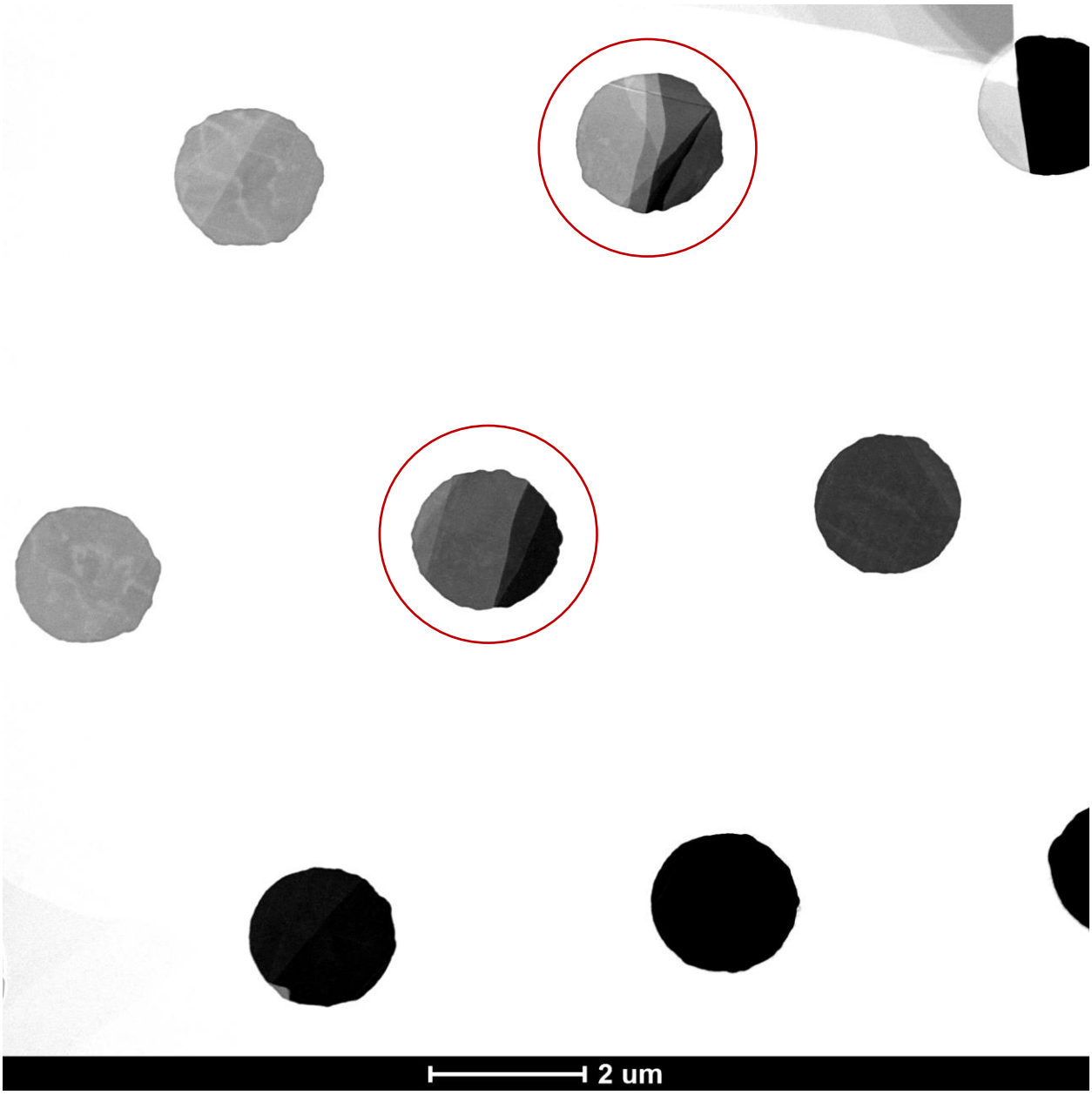

Figure S3. Low-magnification ADF-STEM image of $Ta_{1.6}Te$ showing discrete layers of exfoliated material.

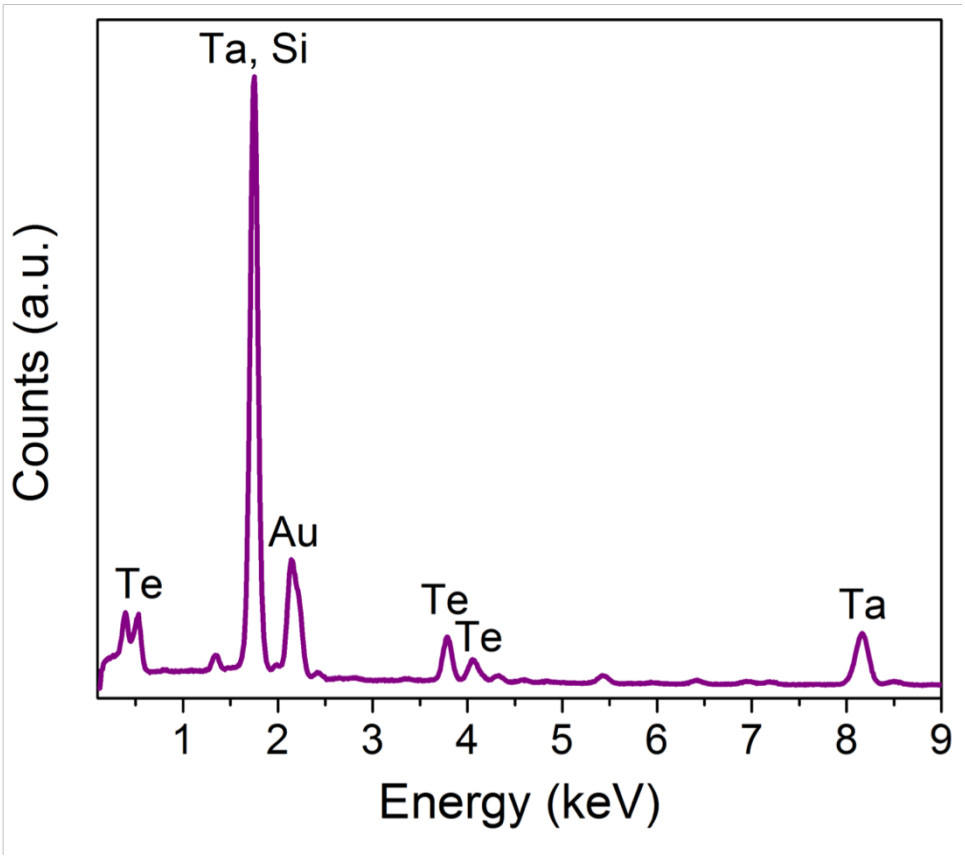

Figure S4. EDS spectrum of $Ta_{1.6}Te$ showing presence of tantalum and tellurium

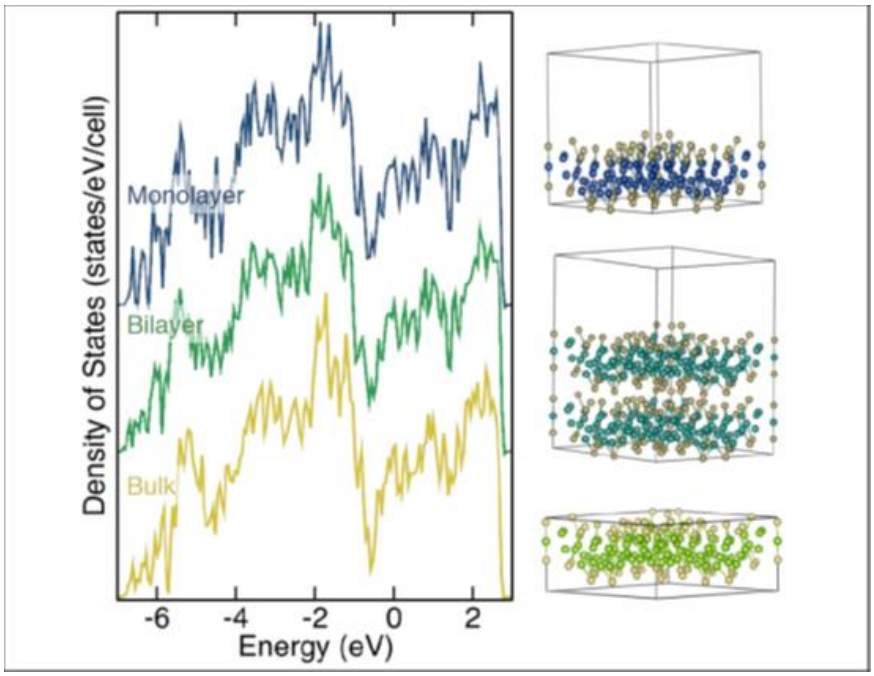

Figure S5. Calculated density of state of monolayer, bilayer, and bulk $Ta_{21}Te_{13}$.

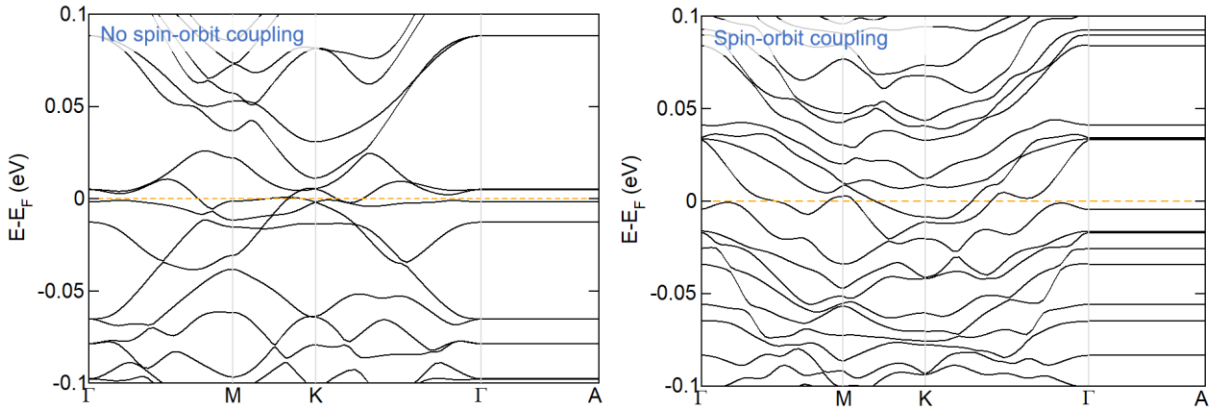

Figure S6. Calculated band structure of monolayer $Ta_{21}Te_{13}$ with and without spin-orbit coupling.

References

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