Epitaxial $(Bi,Sb)_2Te_3$/graphene/2D-Ga heterostructures towards topological superconductivity

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The interface of a conventional BCS superconductor (SC) and a topological insulator (TI) is predicted to host an exotic state of matter known as a topological superconductor, the elemental excitations of which could potentially enable quantum computing schemes that are robust against error produced by decoherence. The synthesis of a SC-TI heterostructure, however, is challenging due to structural dissimilarities and high interface reactivities between common superconductors and topological insulators. Here, we report on the synthesis and properties of wafer-scale $(Bi,Sb)_2Te_3$/graphene/2D-Ga heterostructures grown in-part by a new method, i.e. confinement heteroepitaxy (Chet), pioneered by the Robinson group [1]. Chet enables us to intercalate and form atomically thin gallium (Ga) layers at the interface of 2L epitaxial graphene (Gr) and its native SiC (0001) substrate. The graphene-encapsulated 2D-Ga films are predominantly 2-3L thick, crystalline, and epitaxially registered to the SiC, as confirmed by high-resolution scanning transmission electron microscopy (HR-STEM) (Fig. 1a). The Gr/Ga films exhibit a superconducting state with zero resistance at a transition temperature of $T_c \sim 4$ K (Fig. 1c), which is higher than that of bulk α-Ga ($T_c = 1.08$ K). In addition to serving as a capping layer for the 2D-Ga film, the Gr layer is both a reaction barrier and an ideal substrate for the subsequent molecular beam epitaxial (MBE) growth of a TI material $(Bi, Sb)_2Te_3$. HR-STEM and energy-dispersive X-ray spectroscopy (EDS) mapping shown in Fig. 1b demonstrates the atomically sharp interfaces and high-quality layer-by-layer growth of all constituent layers in a heterostructure of 6 quintuple-layer $(Bi,Sb)_2Te_3$ grown on Gr/Ga. Reflection high energy electron diffraction pattern and angle-resolved photoelectron spectroscopy measurements verify its crystalline integrity and the Dirac surface bands of the TI film. First-principles calculations reveal the electronic band structure of the heterostructure, which is conducive to proximity-induced superconductivity in the TI film. Our approach circumvents several key challenges in making high-quality SC-TI heterostructures to offer a new route towards the realization of topological superconductivity.

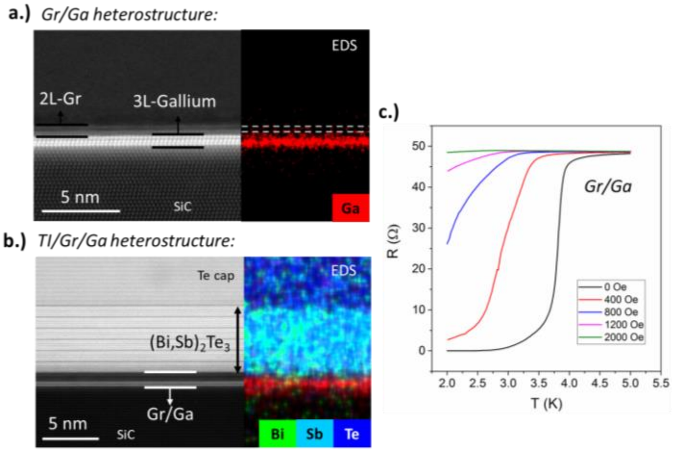

Figure 1: (a-b) HR-STEM and EDS mapping of Gr/Ga heterostructure before and after MBE-deposition of $(Bi,Sb)_2Te_3$ film, respectively. (c) RT curves for Gr/Ga film showing $T_c \sim 4$K.

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Supplementary Information

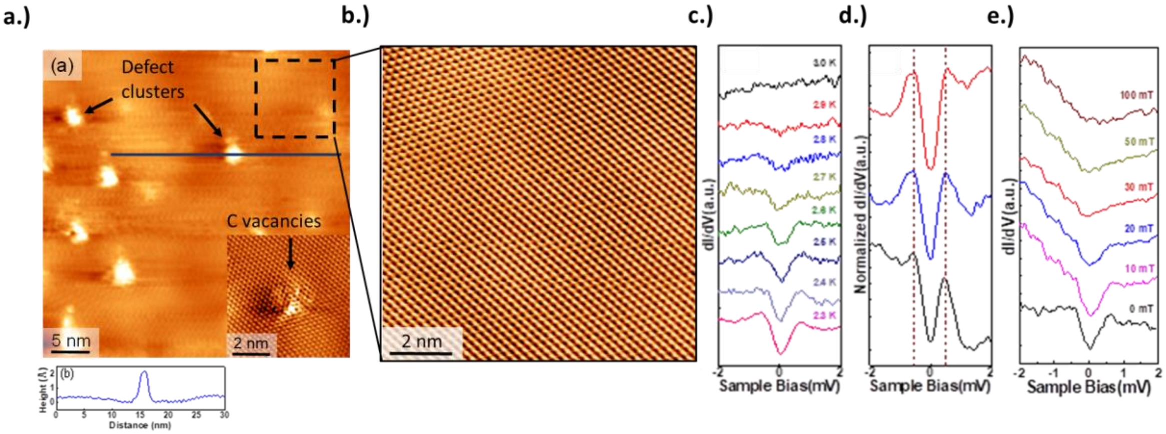

Figure S1: **Scanning tunneling microscopy and spectroscopy (STM/STS) of superconducting graphene/2D-Ga heterostructures.** (a) STM image of a typical Ga-intercalated terrace of epi-graphene/SiC (0001), highlighting the 2 types of defects we observe on the surface: large "defect clusters" and carbon vacancies which are a result of graphene processing and the intercalation synthesis. (b) A pristine region of bilayer graphene (on 2D-Ga). (c) Temperature-dependent differential conductance (dI/dV) measurements taken from STM imaged region in (a). At 2.3 K, there is a clear energy gap with decoherence peaks, further verifying the existence of a superconducting state. (d) Three different spatially averaged dI/dV differential conductance spectra (T = 2.2 K) normalized by spectra taken at 3 K. (e) Magnetic field-dependent dI/dV spectra taken at the same location as (c).

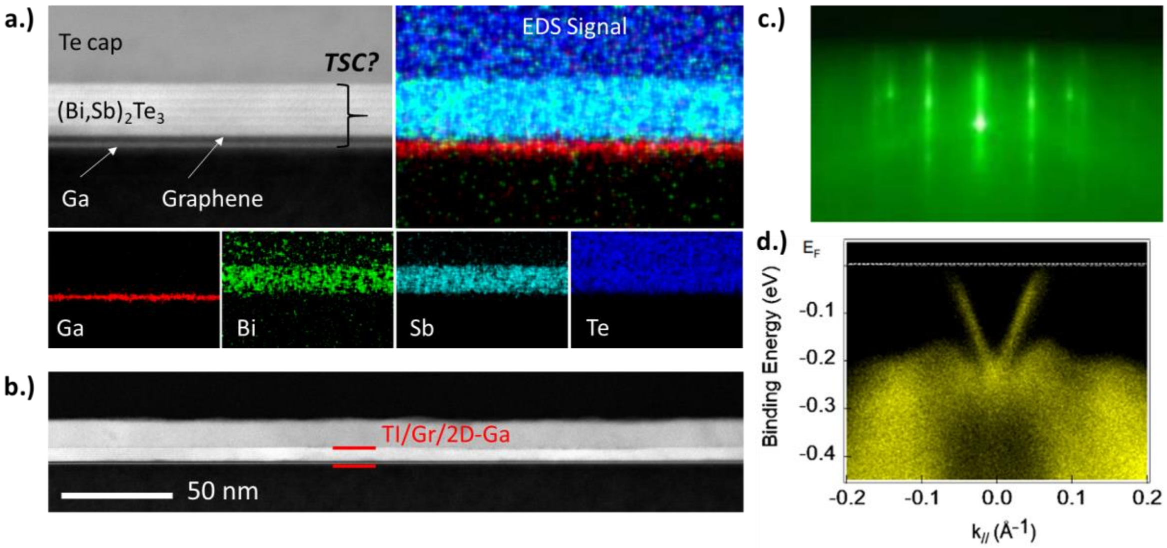

Figure S2: **Characterization of the $(Bi,Sb)_2Te_3$ (TI) growth on graphene/2D-Ga "substrate" for our proposed proximity-induced topological superconductivity platform**. (a) Cross-sectional STEM image of the heterostructure post-TI growth with energy dispersive spectroscopy (EDS) elemental mapping showing the clear co-location of Bi, Sb, and Te separated from the underlying Ga layer by a thin 2L-graphene sheet. (b) A low-mag STEM image of the same area showing uniformity and continuity over a 300 nm section. (c) Reflection high energy electron diffraction (RHEED) pattern of a nominally quintuple-layer (QL) thick $(Bi,Sb)_2Te_3$ film demonstrating high-quality, crystalline growth. Individual $(Bi,Sb)_2Te_3$ QLs with van der Waals spacing can be seen in the high-resolution STEM in Figure 1b and Figure S3 below. (d) Angle-resolved photoelectron spectroscopy (ARPES) measurement showing the Fermi level clearly within the conduction band of $(Bi,Sb)_2Te_3$ film. This material system can be tuned from n-type (Bi_2Te_3) to p-type (Sb_2Te_3) by varying the Sb composition.

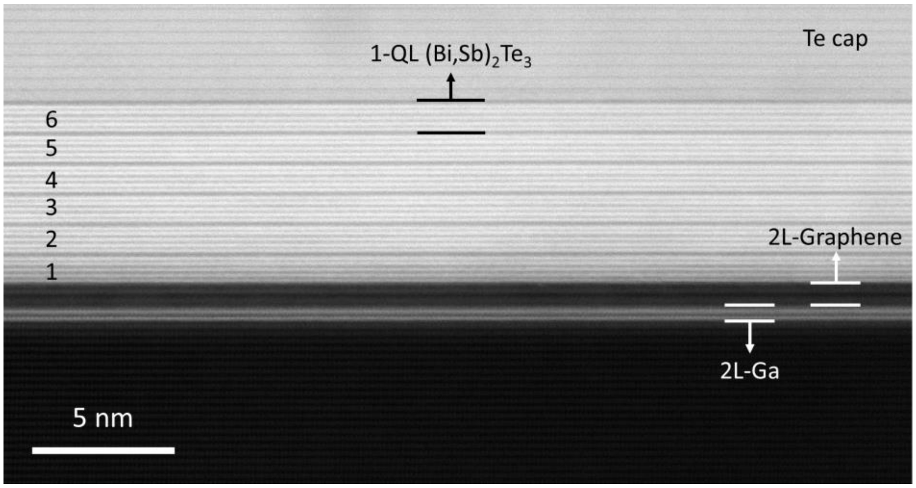

Figure S3: **High-resolution cross-sectional STEM image of epitaxial $(Bi,Sb)_2Te_3$/graphene/2D-Ga heterostructure**. Blown-up image from Fig. 1b for ease of viewing.