Supplementary Information for

Extraordinary tunnel electroresistance in layer-by-layer engineered van der Waals ferroelectric tunnel junctions

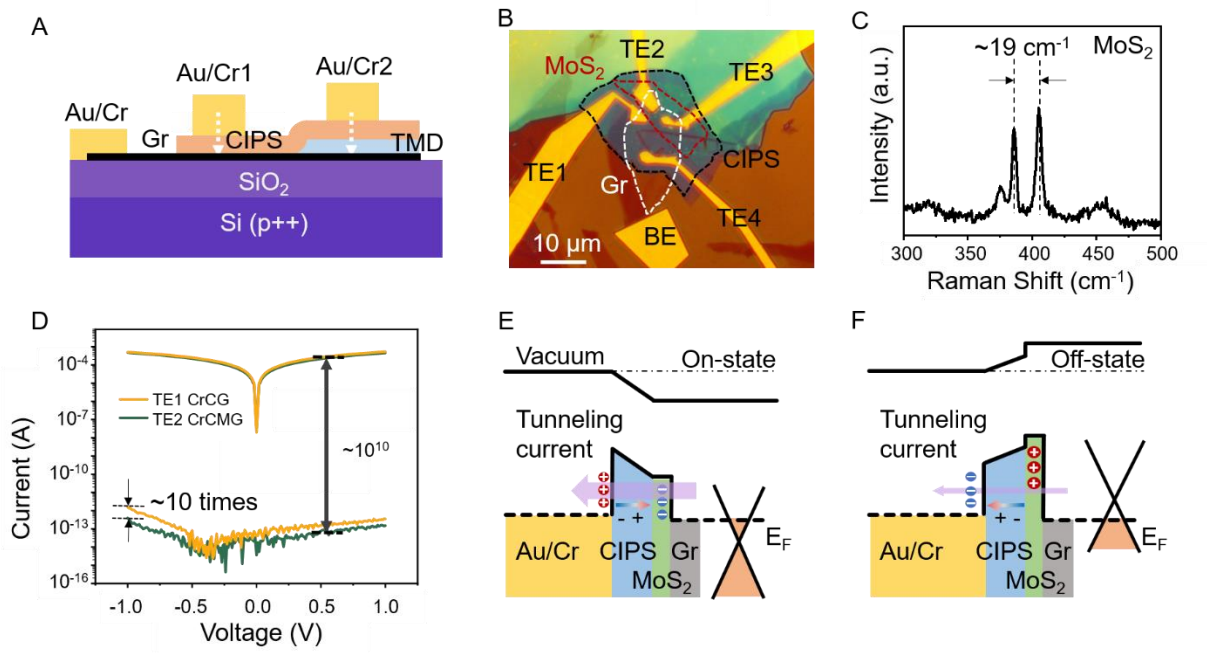

Figure 1. Layer engineering of the TER in Cr/CIPS/graphene (CrCG) tunneling device by inserting a monolayer MoS_2 in between CIPS/graphene

(A) Schematic of the device structure. Au/Cr1 and Au/Cr2 refer to TE1 (CrCG) and TE2 (Cr/CIPS/MoS_2/graphene (CrCMG)) in the optical image in (B). (B) Optical image of the CrCG and CrCMG tunnel junctions. There is a monolayer MoS_2 flake under the TEs labeled with TE2 and TE3, while the graphene directly contacts the top CIPS under the TEs labeled with TE1 and TE4. Graphene, MoS_2, and CIPS are circled by white, red, and black dashed lines, respectively. Scale bar, 10 μm. (C) Raman spectrum of the MoS_2 in the device shown in (B). The distance between two peaks is ~19 cm^{-1}, demonstrating that the MoS_2 flake is monolayer. (D) Current-voltage characteristics of the CrCG and CrCMG tunnel junctions. The largest difference between the two off-state currents is around 10 times. (E) Band diagram for the CrCMG tunnel junction in on state. (F) Band diagram of the CrCMG tunnel junction in off state. In (E) and (F), the built-in polarization fields in the CIPS are indicated by the blue-to-red arrows, and the tunneling currents are indicated by the purple arrows. The difference between the vacuum levels of MoS_2 and graphene is not the focus and thus is not specified in the illustrations in (E) and (F).

Reference

Wang Q, Xie T, Blumenschein N A, et al. Extraordinary tunnel electroresistance in layer-by-layer engineered van der Waals ferroelectric tunnel junctions. *Matter* **5**, 4425, (2022).