Twisted graphene membranes: Layer-selective hydrogenation and proton transport

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Abstract
Recent advances in electrochemical hydrogenation have revealed that graphene can undergo reversible electronic transitions under controlled proton environments, providing a versatile platform to modulate its electronic and structural properties. From a theoretical perspective, understanding how such transitions emerge from the interplay between electronic decoupling, electrostatics, and surface chemistry remains an open challenge.
In this work, we demonstrate layer-selective conductor–insulator transitions in twisted bilayer graphene driven by hydrogenation under strong transverse electric fields. Our results show that large twist angles suppress interlayer electronic coupling, enabling independent control of the carrier density in each layer. Moreover, we identify proton transport pathways through high-symmetry AA-stacked regions of the lattice, highlighting the role of local stacking geometry in governing permeation across atomically thin membranes.
By combining atomistic insight with device-level implications, this work positions twisted bilayer graphene as a model system for coupling charge transport, chemical functionalization, and proton conduction, with potential applications in hydrogen technologies, energy storage, and nanoelectronic devices.
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References

[1] J. Tong, *et al.* Control of proton transport and hydrogenation in double-gated graphene. Nature **630** (2024) 619–624. https://doi.org/10.1038/s41586-024-07435-8

Figures

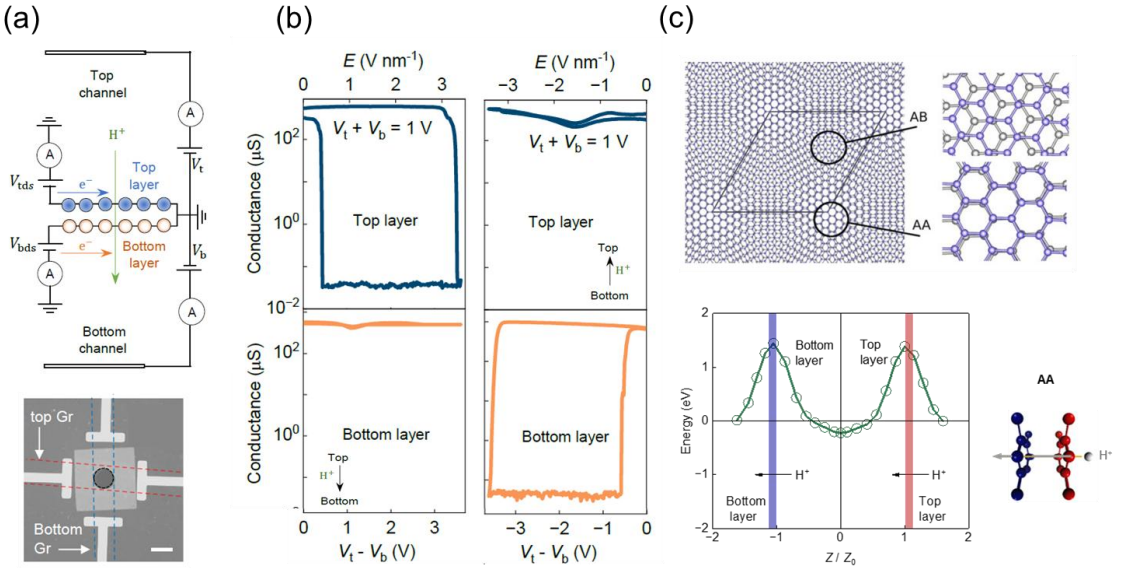

Figure 1: (a) Schematic and optical image of bilayer graphene device with twist angle. **(b)** In-plane electronic conductance of top (blue) and bottom (orange) graphene layers under electric field. **(c)** Calculated proton permeation through the AA region of twisted bilayer graphene.