

# Unreconstructed moiré superlattice in marginally twisted graphene bilayers

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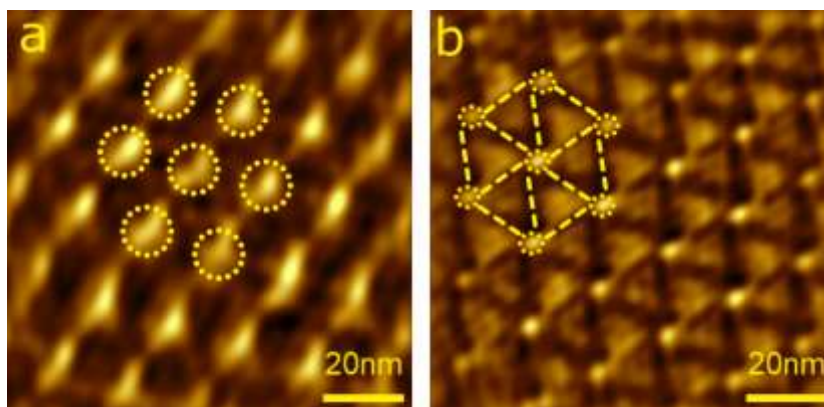
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In twisted bilayer graphene, at specific (magic) rotation angles ultra-flat electronic bands and enhanced density of states emerge in the vicinity of the Fermi energy [1]. Near the first magic angle of  $1.1^\circ$ , a variety of strongly correlated electronic phases, including unconventional superconductivity have been observed, establishing the field of graphene-based twistrionics [2]. Theoretical predictions indicate that at smaller twist angles interactions become even more dominant, potentially stabilizing novel correlated phases. However, the experimental access to such large-period moiré superlattices has been prohibited by atomic reconstructions emerging from the competition between interlayer stacking energy from van der Waals adhesion and intra-layer elastic energy from lattice distortions [3]. Here, we demonstrate that unreconstructed twisted bilayer graphene superlattices can be stabilized at twist angles below 1 degree, by weakening the effective interlayer mechanical coupling (the driving force for reconstructions) while preserving sufficient electronic coupling between the layers. This can be achieved by introducing a molecular monolayer between the twisted graphene sheets. Under these conditions unreconstructed moiré patterns with enhanced periodicity can persist even at sub-1-degree rotation and can be resolved by conductive atomic force microscopy (c-AFM) or scanning tunnelling microscopy (STM). Intercalated twisted bilayer graphene structures with reduced mechanical interlayer coupling can enable the exploration of the previously unreachable strongly correlated electronic phases at magic twist angles below one degree.

## References

- [1] Bistritzer, R. & MacDonald, A. H., *Proc. Natl Acad. Sci.* **108** (2011) 12233–12237.
- [2] Cao, Y. *et al.*, *Nature* **556** (2018) 43–50.
- [3] Yoo, H. *et al.*, *Nat. Mater.* **18** (2019) 448–453.

## Figures



**Figure 1:** Conductive AFM current images of bilayer graphene superlattices with a twist angle of  $\sim 0.7^\circ$  without (a) and with (b) full structural reconstructions, as indicated by the size of AA regions (dashed circles) and the absence/presence of the soliton domain walls (dashed lines).