Moiré Buckling Transition and Bending Stiffness Collapse of Twisted Bilayer Graphene

Jin Wang[1,*], Ali Khosravi[1,2,3], Andrea Silva[1,3], Michele Fabrizio[1], Andrea Vanossi[1,3], Erio Tosatti[1,2,3]

1. *International School for Advanced Studies (SISSA), I-34136 Trieste, Italy*
2. *International Centre for Theoretical Physics, I-34151 Trieste, Italy*
3. *CNR-IOM, Consiglio Nazionale delle Ricerche - Istituto Officina dei Materiali, c/o SISSA, 34136, Trieste, Italy*

[*] Corresponding author: jinwang@sissa.it

Twisted bilayer graphene (TBG) is widely investigated when deposited onto, or encapsulated into, solid frames, that keep them flat. Their properties are much less known and potentially exciting once freely suspended TBG states could be experimentally realized. Realistic simulations show that for decreasing twist angle θ the TBG structure should spontaneously buckle with twice the moiré periodicity [1]. The buckling transition is predicted to be continuous with a displacive order parameter $Q \sim (\theta_c - \theta)^\beta$ with $\theta_c \approx 3.7°$ and an unusual exponent $\beta = 0.7$ at T=0. Unexpectedly, at the same critical point, the TBG bending rigidity also undergoes a critical collapse with its own novel exponents, a behavior which implies critical flexural fluctuations, which we verify in simulations and explain by a mean-field analytical model. Choosing the well know magic twist angle $\theta \approx 1.08°$ as an example, the freestanding TBG's buckled state is akin to a zigzag washboard with alternating off-horizontal angles around $\pm 5°$ and a corrugation larger than 1 nm. Moreover, the buckled state is robust against tensile stress up to ≈ 1 GPa, and survives high temperatures well above 300 K. Electronically, the buckled TBG is still endowed with narrow bands but with interestingly modified properties compared to those of the standard flat TBG. We believe in summary that these and other remarkable structural, critical, flexural, frictional, and electronic predicted properties make an experimental study of low twist angle freestanding 2D materials highly desirable. Works on the spontaneous buckling of other important bilayers, including transition-metal dichalcogenide (TMDC) and heterostructures is currently going on.

[1] J. Wang, A. Khosravi, A. Silva, M. Fabrizio, A. Vanossi, and E. Tosatti, Phys. Rev. B **108**, L081407 (2023).