Application Note

Moiré Superlattice in Twisted Bilayer Graphene

Nano-electrical characterization of tBLG at cryogenic temperatures

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Abstract

Two sheets of graphene stacked onto one another with a twist form a system which has garnered a lot of interest recently due to its fascinating electronic properties that often emerge at the scale of the resulting moiré superlattice which is typically 10-100 times larger than the graphene lattice constant. Particularly for small twist angles, the moiré superlattice constant falls within the range of 10-20 nm, making scanning probe microscopy (SPM) an ideal tool for investigation of twisted bilayer systems. With this application note we showcase how an attoAFM I cryogenic microscope, with its nanoscale lateral resolution, equipped with advanced AFM modes like conductive tip atomic force microscopy (ct-AFM) and piezo-response force microscopy (PFM) can be instrumental in exploring electrical and electromechanical properties of twisted bilayers.

Introduction

The exfoliation of graphene by Novoselov and Geim [1] proved to be a transformative event in the field of two-dimensional (2D) material research. The family of 2D materials has grown since to include a variety of new members like TMDs [2], monochalcogenides, mono-elemental 2D semiconductors, and van der Waals (vdW) heterostructures [3-4]. These materials possess unique optical and electronic properties and have shown a great deal of versatility in tunability of their properties [2,5]. These remarkable features have propelled them to the forefront of scientific research and technological innovation.

Stacking two 2D sheets on top of one another in a vdW heterostructure can generate long-range interference patterns called moiré patterns, which results in the creation of a new artificial 2D superlattice with larger periodicity in real space. These patterns arise either from a slight mismatch in the lattice constants of the stacking layers [6] like graphene on hexagonal boron nitride (hBN) or by changing the twist angle between the two stacking layers like, e.g., twisted bilayer graphene (tBLG) as illustrated in Fig. 1. The twist angle can serve as an extra parameter for tuning the electronic properties of a heterostructure. This was most notably demonstrated by the discovery of interaction-mediated insulating states and superconductivity in magic angle twisted bilayer graphene (MATBG) [7]. Hence, central to deciphering these moiré patterns and understanding their exciting properties, is the need for a platform offering robust and accurate electrical characterization techniques.

Due to their high lateral resolution, ct-AFM and PFM techniques have widely been used for the electrical and electromechanical characterization of materials [8]. attoAFM I makes these techniques readily available at variable temperatures and high magnetic fields which empowers researchers with additional control over experimental parameters to tune and explore exotic phenomena like quantum Hall effects and superconductivity in such heterostructures.

In this white paper, we have employed ct-AFM and PFM to image the moiré superlattice of tBLG twisted at an angle of 0.8° which results in a moiré period of about 18 nm.

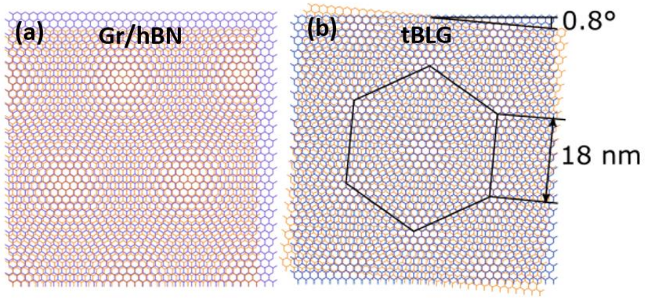

Fig. 1: Schematic of atomic scale moiré pattern created in a) graphene/hBN due to lattice mismatch, and b) by overlapping two twisted sheets of graphene.

Experiment and results

Sample preparation

The sample was prepared by transferring graphene layers onto a Si/SiO$_2$ substrate using a stamping method. The layer arrangement begins with a monolayer of graphene, followed by another layer of graphene with a twist angle of 0.8°, and finally, an hBN layer underneath.

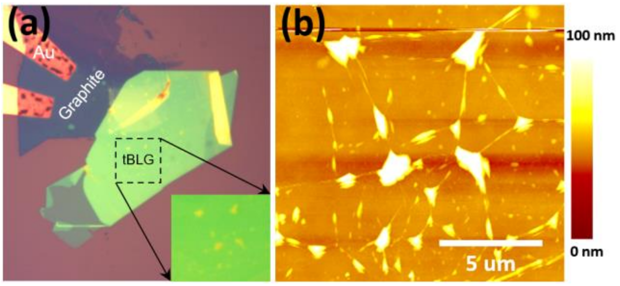

Fig. 2: a) Optical image of the device b) An AFM topography image of the region of interest.

As seen in the optical image in Fig. 2a, the monolayer graphene is contacted with multilayer graphite, which is contacted to the

gold electrode for wire bonding. The sample fabrication resulted in the formation of the bubbles which are evident in both optical and AFM images. These bubbles serve as markers to effortlessly locate the region of interest (ROI). ROI was identified by corelating the sample's optical image with the topography images obtained.

The measurements were carried out with an attoAFM I cryogenic scanning probe microscope based on an interferometric displacement sensor in an attoDRY2200 cryostat at the temperature of 1.8 K. The microscope was equipped with closed loop scanning feature which made navigation around the sample easy by employing an optical readout of the X and Y positions. This feature also aided in mitigating nonlinearities intrinsic to piezo scanners, thereby minimizing potential imaging artifacts. For electrical measurements, ct-AFM was employed in contact mode with a Nanosensors PtSi-FM probe and for PFM measurements same probe was used with an AC voltage applied at the contact resonance frequency of 377 kHz.

ct-AFM measurements

ct-AFM technique has been extensively utilized in previous studies [9,10] to image the local conductivity of such twisted bilayer systems. This technique measures the current flowing through the ohmic contact between the probe and the sample surface.

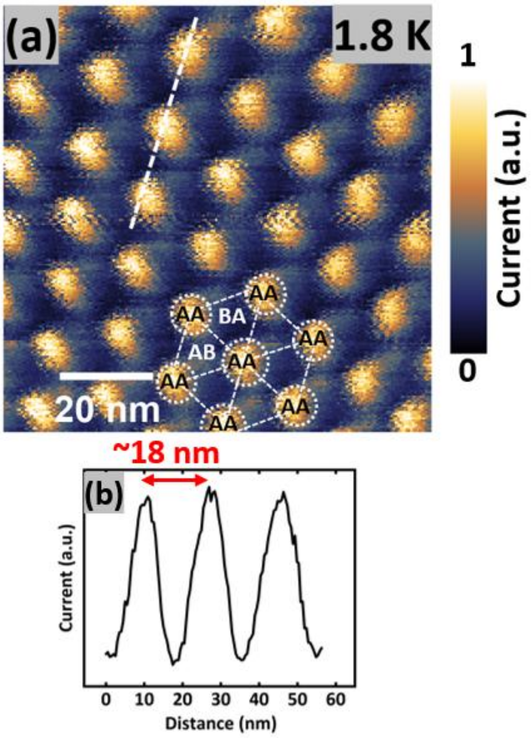

Fig. 3: a) Current map of tBLG revealing the moiré pattern generated because of the twist between the stacked layers. b) Current profile taken along the indicated dashed line showing a period of 18 nm.

Figure 3a shows a current map obtained while scanning the tip across a small sample area with a 68 mV bias applied to the sample. The image reveals the newly reconstructed moiré superlattice resulting from the twist between the two layers as variations in the local conductivity.

By correlating the twist angle with the moiré periodicity, a twist angle of 0.8° can be inferred from the recorded moiré pattern, aligning closely with the expectation. The relation between the twist angle and the moiré period is given by the following relation [15]:

$$d = \frac{\alpha}{a\sin\left({}^{\theta}/_{2}\right)}$$

where $\alpha = 0.246$ nm is the lattice constant of graphene and θ is the twist angle between the layers. It is evident that the conductivity in the AB-/BA-stacked regions is lower than in the AA-stacked regions, which is consistent with findings from prior studies [10,11]. This heightened vertical conductivity in the system is attributed to increased interlayer conductivity and a higher interfacial carrier density [10,12].

We also observed a variation in the moiré period when recording ct-AFM images at various locations on the sample, which suggests a twist angle inhomogeneity. These twist angle inhomogeneities are commonly observed and have been reported before [13].

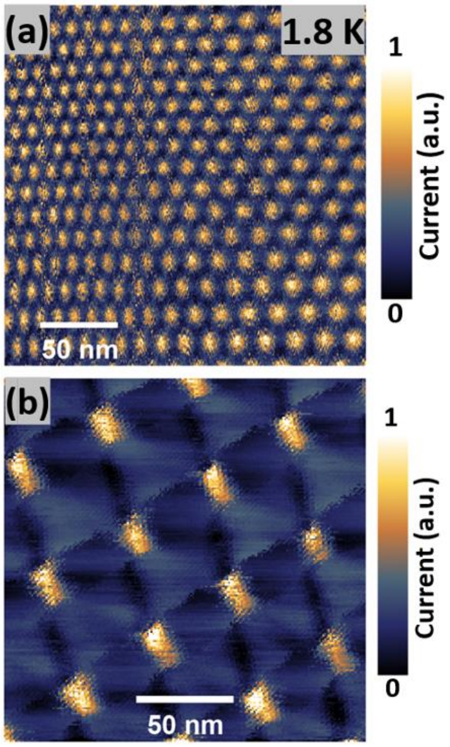

Fig. 4: a) ct-AFM image of tBLG showing variations in moiré periodicity over a big scan area b) moiré superlattice of tBLG imaged at a different sample area showing a period of about 50 nm.

Such inhomogeneities are believed to be introduced during sample fabrication by small variations in local strain, blisters, or

substrate imperfections [13]. The variation in period is noticeable in the current map in Fig. 4a as one moves from left to right. Fig 4b shows area of the sample where moiré period is constant (~50 nm), but substantially deviating from the expected nominal value (18 nm).

PFM measurements

We also performed PFM measurements on the tBLG to image the moiré pattern. The technique is based on the inverse piezoelectric effect and utilizes a sharp conductive probe which is brought into contact with the sample surface. An AC voltage is applied to the conductive probe at its contact resonance frequency to create an electric field between the probe and the back electrode of the sample. This electric field interacts with the sample and induces periodic mechanical deformation. By detecting the amplitude and phase of these deformations one can get information about the local electromechanical response of the sample.

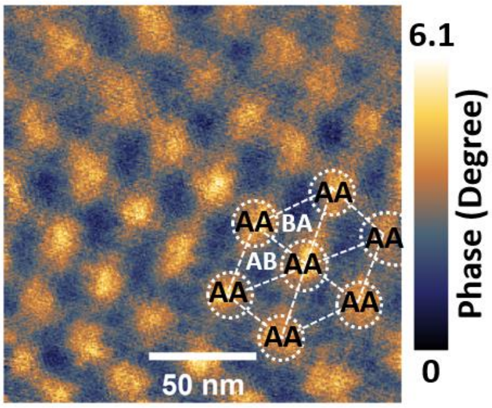

Fig. 5: PFM phase image response of tBLG

Fig. 5 shows the PFM phase response image recorded while scanning on the tBLG system at 3 K. The phase contrast displayed in the image is different from what one would expect in a typical PFM phase image contrast (sharply 180° of difference). This suggests that the origin of the PFM response of the tBLG cannot be simply attributed to piezoelectric or ferroelectric behavior and hence more detailed study needs to be undertaken to rule out other contributions (electrostatic, electrochemical, in-plane electric dipoles and cantilever dynamics), as also reported by McGilly et al. [14]. From Fig. 5 one can, however, clearly differentiate the AB-/BA-stacked regions (dark contrast) from the AA- stacked regions (bright contrast) which proves that PFM can be advantageous for quick and effortless characterization of the moiré superlattice in twisted bilayer systems.

Summary

We demonstrate how the combination of nano-electrical scanning-probe-microscopy modes (ct-AFM and PFM) can serve as a powerful approach for analyzing moiré patterns in twisted van der Waals bilayers and for exploring their electronic properties. attoAFM I, equipped with ct-AFM and PFM upgrades, operating at variable temperature and variable magnetic field, offers indeed unprecedented capabilities for studying such twisted bilayer systems. By combining precise control over experimental parameters with high-resolution imaging and electrical measurements, attoAFM I can help researchers unravel the intricacies of these fascinating materials.

References

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