Application Note

Nano-electrical characterization of moiré systems

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Abstract

Understanding the microscopic properties of moiré systems at varying temperatures and magnetic fields is essential to comprehend the diverse electronic phenomena that it has showcased, such as correlated insulator states and unconventional superconductivity. In this Application Note we demonstrate on one of the classic bilayer moiré structures (graphene/hBN) how the correlative scanning probe microscope attoAFM I, operating at variable temperatures and high magnetic fields, can be utilized to perform the whole research process from identifying the region of interest with KPFM to revealing its local electrical and electromechanical properties using ct-AFM and PFM.

Introduction

Two-dimensional (2D) materials have been the subject of intense research and development for over two decades. This growing research interest was triggered by the discovery of graphene [1-3] and has since then expanded to encompass a wide variety of materials including hexagonal boron nitride (hBN) [4], transition metal dichalcogenides (TMDs) [5] and van der Waals heterostructures [6-7]. Their unique electronic, optical, and mechanical properties make them attractive for a variety of potential applications in diverse fields such as electronics [2, 8], energy storage [9], and biomedicine [10].

Recently, moiré systems have emerged as a new frontier in the field of 2D materials research. Moiré systems refer to materials exhibiting the superlattice-like patterns that arise from the slight misalignment between the honeycomb lattices of two or more layers in a 2D bilayer structure [11]. Apart from the formation of a moiré pattern due to a slight lattice constant mismatch between the individual layers (Fig 1a), another way to form these patterns is by twisting or rotating two individual layers relative to each other at a small angle (Fig 1b). This phenomenon is particularly evident in twisted bilayer graphene, where the resulting moiré pattern significantly alters the electronic properties of the material, leading ultimately to the emergence of unconventional superconductivity at the magic angle of 1.1° [12]

Moiré systems present a new unique opportunity for research. Next to the classic thermodynamical parameters (T, B, E), they provide a new, almost continuously tunable property, the twist angle. By adjusting these, one can find a broad range of electronic and optical behavior that can be manipulated and tuned continuously. In addition to superconductivity, they have been found to host intriguing strongly-correlated and topological phenomena, such as correlated insulator states [13] and orbital magnetism [14]. These discoveries have led to a surge of interest in twistronics with researchers exploring various other 2D materials with tailored electronic properties for use in next-generation electronic and optoelectronic devices.

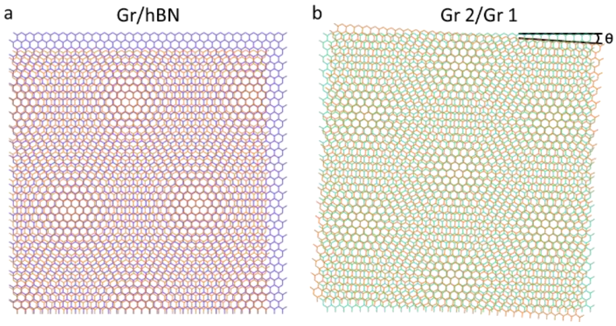

Fig. 1: Schematic diagram of moiré pattern in a) graphene/hBN heterostructure with 0° alignment, and b) twisted bilayer graphene where a second graphene layer (Gr2) is stacked at an angle θ relative to the first layer (Gr1).

Imaging, measuring and correlating the various properties of moiré systems at the nanoscale is a crucial aspect of their research. The most suitable technique is scanning probe microscopy (SPM) which includes conductive-tip atomic force microscopy (ct-AFM), Kelvin probe force microscopy (KPFM) and piezoresponse force microscopy (PFM).

Temperature-dependent studies are crucial for understanding the behavior of 2D materials. Properties such as electrical conductivity, electric polarization, piezoelectric properties, and magnetism can change dramatically, and many exciting phases, like 2D topological insulators and unconventional superconductivity only persist at low temperatures [11, 20]. Therefore, it is essential to understand their microscopic properties at low temperature and varying magnetic field. attocube variable temperature correlative SPM microscope, the attoAFM I, together with the low vibration attoDRY cryostats, are uniquely designed to be a precise and reliable tool for these investigations.

In this Note, we demonstrate how attocube systems attoAFM I can be utilized for measuring various local electrical properties of graphene/hBN sample using ct-AFM, KPFM and PFM at low temperature.

Measurement Results

The identification of different 2D layers can be achieved through quantitative AFM analysis of the step heights between them. However, this approach requires careful analysis and a correction for any absorbed surface layer or contamination. Alternatively, electrical modes of SPM provide a simpler tool for differentiating between 2D grains based on imaging alone. By mapping out the surface potential of the sample using KPFM, it

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is possible to differentiate between layers with varying layer thicknesses based on variations in their electrical properties. KPFM utilizes a conductive AFM tip, which is scanned over the sample surface while applying an AC voltage between the sample and the probe. The resulting electrostatic force is detected and used to measure the surface potential. Different modes of KPFM can be used to detect the electrostatic interaction. The two main detection modes of KPFM are amplitude modulated KPFM (AM-KPFM), where the changes in the oscillation amplitude allows for detecting the electrostatic force, and frequency modulated KPFM (FM-KPFM), where the frequency variation is recorded, giving access to the derivative of the electrostatic force.

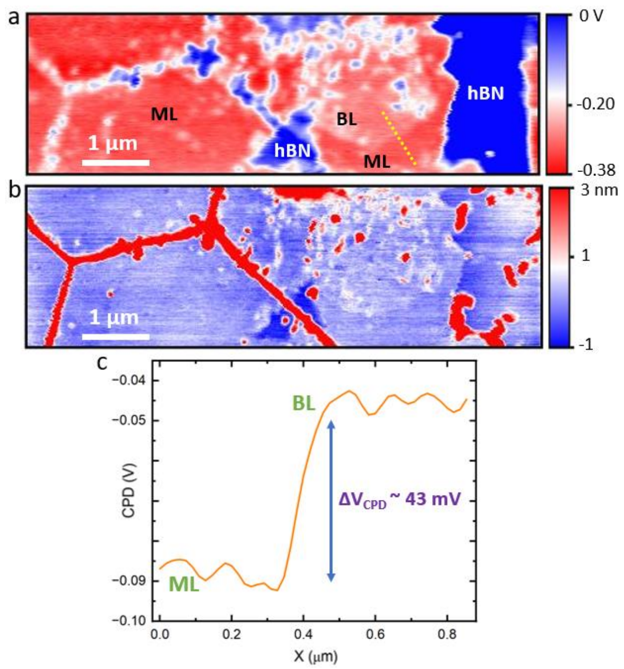

Fig. 2: KPFM image of graphene/hBN heterostructure. a) FM-KPFM image. ML, BL and hBN correspond to monolayer graphene, bilayer graphene and hexagonal boron nitride, respectively. b) Topographic AFM image corresponding to (a). c) The line profile of KPFM signal along the yellow dashed line shown in (a).

The FM-KPFM signal obtained from the graphene/hBN heterostructure is shown in Fig. 2a, while the corresponding topography image is shown in Fig. 2b. The topography image alone does not allow us to distinguish between monolayer and bilayer graphene or hBN exposed between the graphene flakes. However, the KPFM image in Fig. 2a clearly reveals variation in the surface potential of monolayer, bilayer graphene and hBN, making it possible to differentiate between them. The variation in CPD can be attributed to the distinct electrical properties of each layer stack, which influences their respective responses to the applied electric field. A line profile of KPFM signal, Fig. 2c, across the monolayer (ML) and bilayer (BL) graphene reveals the contact potential difference (CPD) of ΔV_{CPD} ~ 43mV, which is consistent with previous reports [17, 18]. The enhanced KPFM signal of BL graphene comes from the presence of interlayer potential difference between the two graphene layers. The CPD between the ML graphene and twisted BL graphene can be different and may depend on twist angle between the layers [19]. The large bandgap of hBN results in a much lower conductivity and higher surface potential than graphene. As a result, the KPFM signal is higher than that from graphene, as shown in Fig. 2a.

We also employed ct-AFM to probe the local electric response of the graphene/hBN heterostructure. ct-AFM enables the mapping of a sample's electrical behavior in response to an applied bias voltage. This technique involves scanning a conductive tip over the sample surface in contact mode while a bias voltage is applied. The electrical current between the tip and the sample is then detected, which is directly proportional to the local conductivity of the material.

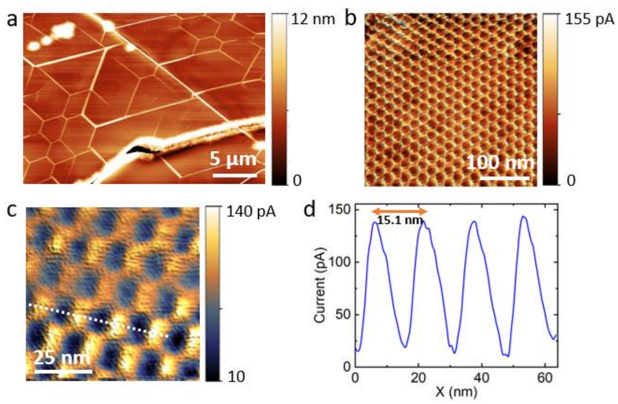

Fig. 3: Conductive AFM image of moiré superlattice. a) Topography image of graphene/hBN heterostructure. b-c) ct-AFM image of moiré pattern at 70 K. d) The line profile of the current signal along the dashed line shown in (d).

Fig. 3a shows an AFM image of the graphene/hBN heterostructure where monolayer graphene was deposited on hBN using chemical vapour deposition (CVD) method. The monolayer graphene grains are separated by boundaries arising from the deposition process. hBN has a 1.8% higher lattice constant than graphene. [12]. This leads to the formation of a moiré pattern in graphene/hBN heterostructure. Fig. 3b-c show ct-AFM images of the moiré pattern on the monolayer graphene taken at 70 K. As shown in Fig. 3d, a line profile of the image in Fig. 3c reveals that the moiré pattern has a periodicity of 15 ± 1 nm. Since this sample was grown by the CVD method, wherein monolayer graphene is epitaxially grown on hBN, the alignment angle between the two layers is precisely 0°. This results in a moiré lattice with a periodicity of 14 nm [15], which is in consistent with our findings.

Furthermore, we employed PFM technique to probe the local electromechanical response of the graphene/hBN heterostructure. PFM measures the piezoelectric response of a material based on the inverse piezoelectric effect, where applying an AC electric field to the material leads to its local

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periodic deformation. This deformation can be detected by the AFM tip, which is in contact with the sample surface. It has been shown that all moiré superlattices, irrespective of the presence of inversion symmetry in the constituent layers, exhibit a mechanical response when subjected to out-of-plane electric fields [16].

In our case of graphene/hBN heterostructure, the PFM image in Fig. 4 shows that the moiré pattern is not only present in the conductive response of the sample, but also in a piezoelectric response. This demonstrates the ability of PFM to investigate the electrical properties of 2D materials, particularly in cases where ct-AFM may not be suitable, such as when the material is insulating.

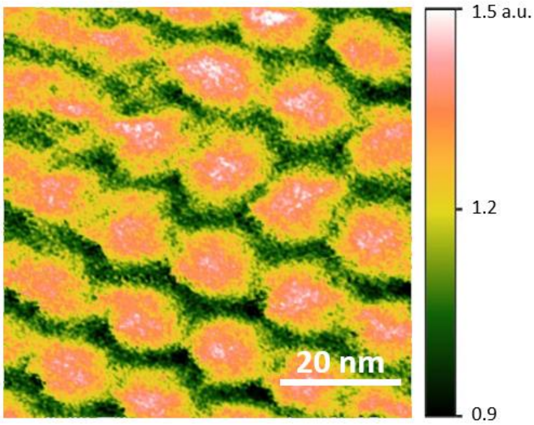

Fig. 4: PFM amplitude image of moiré superlattice in graphene/hBN heterostructure.

To summarize, attoAFM I in the low vibration attoDRY cryostat incorporates all the necessary SPM techniques to facilitate a thorough exploration of interlayer twisting, as well as electrical, mechanical and magnetic properties of 2D layered materials under varying temperatures and magnetic fields. The integration of these techniques offers a powerful approach to gaining a more comprehensive understanding of the behavior and potential applications of these materials.

Outlook

attoAFM I system also allows for the combination of bulk transport measurements with SPM imaging of 2D materials. While electrical transport measurements are valuable in providing insights into the electrical properties of the materials, SPM imaging offers a local nanoscopic understanding of the materials. By combining both techniques, researchers can gain a more profound understanding of the material's behavior at both the macroscopic and nanoscopic levels. This correlative approach can be advantageous in guiding the development of new materials for various applications. Thus, the attoAFM I system, with its unique features, is a valuable tool for researchers in the field of 2D materials and twistronics.

References

[1] K. S. Novoselov et al., *Electric field effect in atomically thin carbon films*. Science **306**, 666 (2004).
[2] A. K. Geim et al., *The rise of graphene*. Nature Mater **6**, 183 (2007).
[3] A. K. Geim, *Graphene: status and prospects.* Science **324**, 1530 (2009).
[4] C. R. Dean et al., *Boron Nitride Substrates for High-Quality Graphene Electronics*. Nature Nanotech **5**, 722 (2010).
[5] K. F. Mak et. al., *Atomically Thin MoS2: A New Direct-Gap Semiconductor*. Phys. Rev. Lett. **105**, 136805 (2010**).**
[6] K. S. Novoselov et al., *2D materials and van der Waals heterostructures*. Science **353**, aac9439 (2016).
[7] A. K. Geim et al., *Van der Waals heterostructures*. Nature **499**, 419 (2013).
[8] Q. H. Wang et al., *Electronics and optoelectronics of two-dimensional transition metal dichalcogenides*. Nature Nanotech **7**, 699 (2012).
[9] Y. Xu et al., *Graphene-based electrodes for electrochemical energy storage*. Energy Environ. Sci. **6**, 1388 (2013).
[10] K. Kalantar-zadeh et al., *Biosensors Based on Two-Dimensional MoS2*. ACS Sens. **1**, 5 (2016).
[11] J. Xue et al., *Scanning tunnelling microscopy and spectroscopy of ultra-flat graphene on hexagonal boron nitride*. Nature Mater **10**, 282 (2011).
[12] Y. Cao et al., *Unconventional superconductivity in magic-angle graphene superlattices*. Nature **556**, 43 (2018).
[13] Y. Cao et al., *Correlated insulator behaviour at half-filling in magic-angle graphene superlattices*. Nature **556**, 80 (2018).
[14] X. Lu et al., *Superconductors, orbital magnets and correlated states in magic angle bilayer graphene*. Nature **574**, 653 (2019).
[15] M. Yankowitz et al., *Emergence of superlattice Dirac points in graphene on hexagonal boron nitride*. Nature Phys **8**, 382 (2012).
[16] L. J. McGilly et al., *Visualization of moiré superlattices*. Nat. Nanotechnol. **15**, 580 (2020).
[17] O. Kazakova et al., *Epitaxial Graphene and Graphene–Based Devices Studied by Electrical Scanning Probe Microscopy*. Crystals 3, 191 (2013).
[18] D. Ziegler et al., *Variations in the work function of doped single- and few-layer graphene assessed by Kelvin probe force microscopy and density functional theory*. Phys. Rev. B **83**, 235434 (2011).
[19] J. T. Robinson et al., *Work Function Variations in Twisted Graphene Layers*. Sci Rep **8**, 2006 (2018).
[20] Z. Y. Fei et al. *Edge conduction in monolayer WTe2*. Nature Phys **13**, 677 (2017).

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