A versatile tool

Investigating the properties of 2D layered materials via Atomic Force Microscopy

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After the first experimental characterization of graphene by Geim and Novoselov in 2004 [1], two-dimensional (2D) layered materials became more and more sought-after due to their exotic behaviour and possible nanotechnological applications [2 – 5]. In this article, we describe systems formed by a single atomic layer including graphite, boron nitride, transition metal dichalcogenides and others or by stacking such layers. The interplay of chemical and geometrical factors, such as the types of the stacked layers, the out-of-plane relative alignment of their unit cells, the twisting angle between consecutive layers, and the presence of defects or strain result in novel material properties. As the properties of such systems are modulated at the nanoscale, the experimental characterization of 2D layered materials requires probing techniques with high resolution.

The Atomic Force Microscopy (AFM) is a versatile tool to assess complementary properties of these materials in detail. In the most simple implementation of AFM, a sharp tip mounted on a microlever is in contact with the surface of the sample. Strong, short-range repulsive forces bend the lever once the tip reaches the surface. Any vertical or torsional bending of the lever is detected via reflecting a laser or SLD beam from the back of the lever onto a position sensitive photodetector (PSPD). A feedback loop during the scanning of the surface allows to control the bending and the force applied by the probe to the sample using piezo actuators with sub nanometer positioning precision.

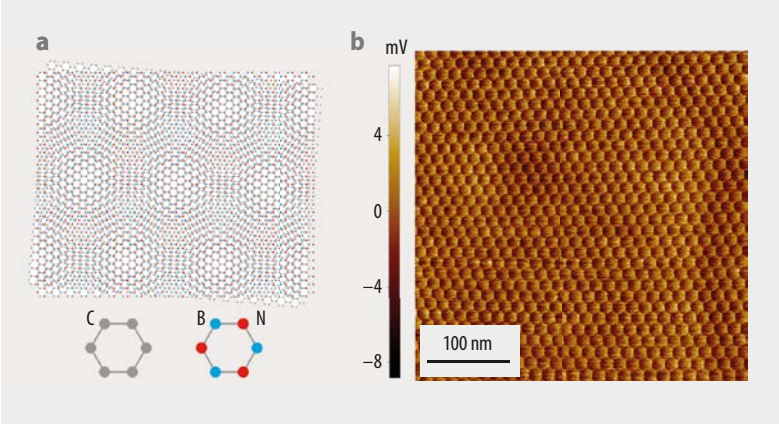

Fig. 1 The relative twist between superpositioned graphene (grey) and hBN (blue/red) layers results in a honeycomb moiré pattern (a). The measurement of the LFM map of graphene/hBN with a Park Systems FX40 AFM shows a moiré pattern in the lateral deflection signal (b).

During the scanning of the surface with a constant force being applied, the probe experiences variations of the tip-sample friction determined by mechanical or material differences between neighbouring areas. This translates into a torsion of the cantilever and a horizontal shift of the reflected beam on the PSPD. Thus, it is possible to measure a map of the lateral force known as Lateral Force Microscopy (LFM). LFM yields the twisting angle of 2D bilayers and the presence of domain boundaries or strain, as the fine corrugation caused by the lattice mismatch generates a moiré-like pattern which is easily detected. A graphene layer on hexagonal boron nitride (hBN) results in a honeycomb pattern (**Fig. 1**) due to the misalignment of the top hBN and graphene layers;

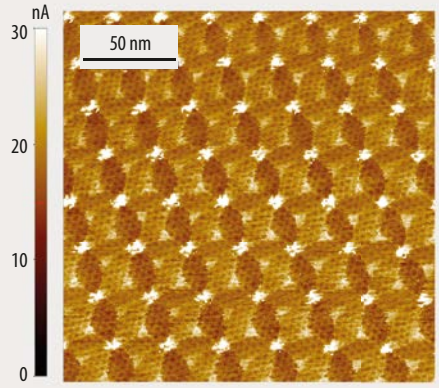

Fig. 2 This C-AFM map of a twisted graphene bilayer over hBN was measured with a Park Systems FX200 AFM and an applied bias voltage of 50 mV.

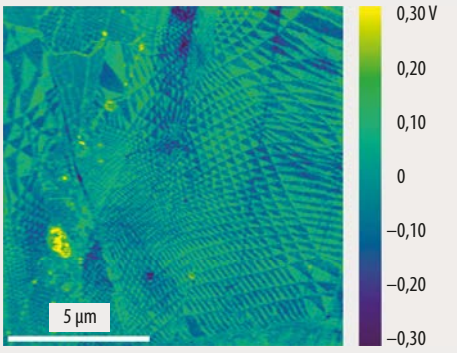

Fig. 3 This KPFM image of the surface potential of a hBN bilayer over graphene was measured with a Park Systems NX20 AFM.

the periodicity is a function of the relative twisting angle.

Conductive AFM (C-AFM) accounts for more direct evidence how superposed monolayers influence the electronic properties of layered structures. This technique uses a contact mode with an additional bias voltage applied to the sample. Thus, a current of typically femto- to micro-Amperes flows through a conductive tip during contact with the sample surface and is collected with a current amplifier. The profile of the current allows to produce conductivity maps associated to the sample topography.

The following example shows the effect of the interplay in the charge density among twisted bilayer graphene (TBG) deposited on a hBN crystal (courtesy: Prof. Machida, Institute of Industrial Science, University of Tokyo, Japan). The bias applied to the bottom graphene layer generates a current flowing to the contact point of the AFM tip through the top graphene layer (**Fig. 2**). Any moiré patterns should depend on the relative angle between the two graphene layers as observed in LFM. However, an additional moiré contrast arises depending on the presence of the underlying hBN and its charge density which modulates the overall transport of current on a very fine scale as described in [6].

More complex AFM implementations often rely on additional hardware but provide information on electrical properties of the samples like the work function or the ferroelectric polarizability. Among others, Kelvin Probe Force Microscopy (KPFM) allows to measure the contact potential difference between the conductive AFM probe and the sample. Applied to 2D structures like the hBN bilayers over graphene (**Fig. 3**), the local atomic reconstruction and layer strain result in distinct domains due to the out-of-plane polarization stemming from the different stacking [4].

Complementary results

AFM modes provide complementary information for a deeper insight into the behaviour and characteristics of 2D materials. Users want commercial AFMs which easily switch between different modes observing the same area of the sample. The Park Systems FX AFMs enable swapping from standard to advanced modes without any hardware modifications. Moreover, advanced implementations such as Sideband™ and heterodyne KPFM are available as default FX features.

As an example (**Fig. 4**), different techniques were used on a Park

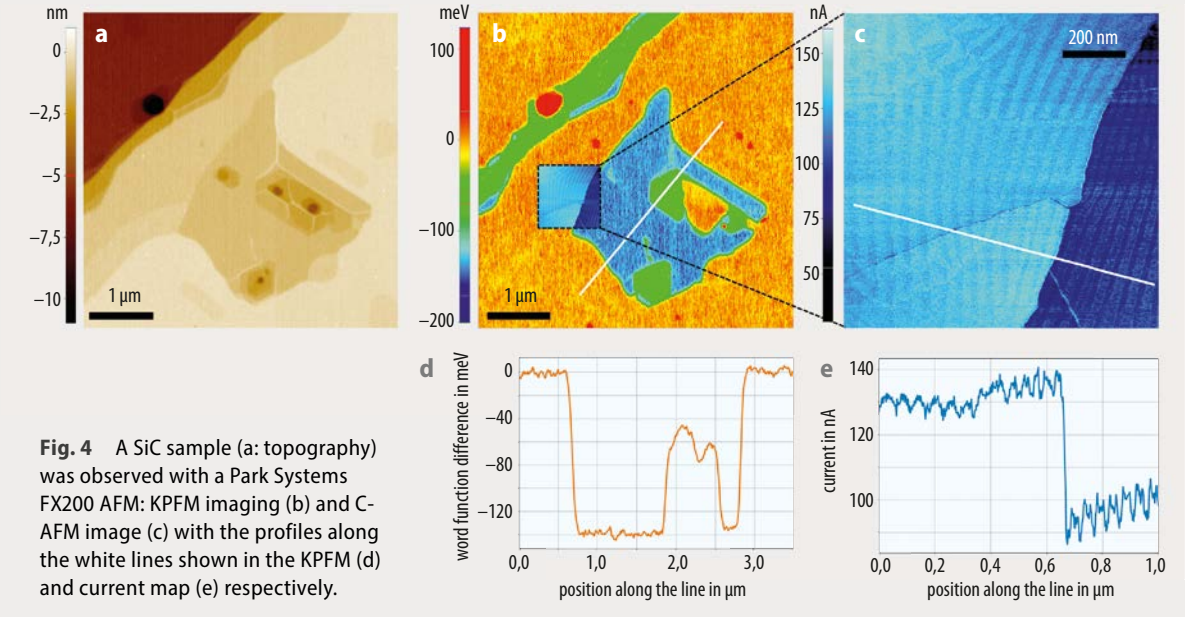

Fig. 4 A SiC sample (a: topography) was observed with a Park Systems FX200 AFM: KPFM imaging (b) and C-AFM image (c) with the profiles along the white lines shown in the KPFM (d) and current map (e) respectively.

Systems FX200 to observe the same area of a conductive Silicon Carbide (SiC) sample (courtesy: Dr. Rejhon from Institute of Physics, Charles University, Prague, Czech Republic). SiC is a precursor to form large flakes of graphene. Thermal treatment at high temperature causes Si atoms close to the surface to progressively leave the crystal resulting in mono- or multilayer graphene stacked on an intermediate graphene-like buffer layer on the SiC crystal. C-AFM reveals the formation of stripe-like domains and the evidence of a surface potential mismatch between areas with different conductivity and domain topology [7].

The work function variation correlates with the presence of incomplete terraces on the main steps of SiC (**Fig. 4b**). In addition, the KPFM contrast changes for areas close to crystalline defects. Subsequent switching to C-AFM mode enables the mapping of conductivity in the same region. Data acquired at higher resolution in an area at the border between the regions with different potential reveal a shift in the average current flow at a constantly applied sample bias of 5 mV. Fine shapes of stripes and quasi-triangular domains occur in both regions. They might stem from a structural reorganisation of multiple graphene layers.

In conclusion, AFM provides a platform to characterize 2D samples using all the information offered by different operational modes. This versatility allows to study the fundamental properties and applications of these structures, making AFM an essential technique to further explore the complex and fascinating world of 2D materials.

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