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Electronic structure of graphene/hexagonal boron nitride heterostructure revealed by NanoARPES

Chaoyu Chen[1]**, Jos**é **Avila**[1]**, Shuopei Wang**[2]**, Rong Yang**[2]**, Guangyu Zhang**[2] **& Maria C. Asensio**[1*]

[1] ANTARES Beamline, Synchrotron SOLEIL & Université Paris-Saclay, L'Orme des Merisiers, Saint Aubin-BP 48, 91192 Gif sur Yvette Cedex, France.
[2] Beijing National Laboratory for Condensed Matter Physics and Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China

E-mail: `asensio@synchrotron-soleil.fr`

Abstract.   The ability to explore electronic structure and their role in determining material's macroscopic behaviour is essential to explain and engineer functions of material and device. Graphene/hexagonal boron nitride heterostructure (G/h-BN) has become a model system to study the emergent behaviour in 2D van der Waals heterostructure. Here by employing angle-resolved photoemission spectroscopy with sub-micrometric spatial resolution (NanoARPES), we give a full description on the electronic structure of G/h-BN, demonstrating the power of NanoARPES to detect the microscopic inhomogeneity of electronic structure for different materials.

1. Introduction
G/h-BN has become model system to study the emergent behaviour in 2D van der Waals heterostructure[1]. Previous ARPES measurements have shown its electronic band structure roughly[2, 3]. However, the lack of high-resolution (energy, momentum, space) is strangling the full understanding of G/h-BN' exotic behaviour. This is because that currently available high-quality G/h-BN on SiO_2/Si substrate[4, 5] are fabricated mainly by plasma-enhanced chemical vapor depositing (PECVD) or exfoliating graphene on mechanical cleaved h-BN, with typical domain size of several micrometer or sub-micrometer[6, 7, 8]. This scale is beyond the spatial resolution of conventional ARPES setups (typical $\sim 100\mu$m). In this work, by applying ARPES with sub-micrometer resolution (NanoARPES[9, 10]), we have been able to measure the comprehensive band structure and Fermi surface of high-quality PECVD-G/h-BN. We observe sharp features of valence bands of graphene Dirac cone and hBN, confirming that the G/h-BN system is atomically clean[11].

2. Experimental details
We first introduce the working principle of NanoARPES, as illustrated in Fig. 1a. Fresnel zone plate is used to focus the synchrotron radiation beam into nanometer size. Order sorting aperture can block the undesired orders of diffraction. The sample is mounted on a high-precision scanning stage with five degrees of freedom. Piezoscanners allow zero backlash and nanometric
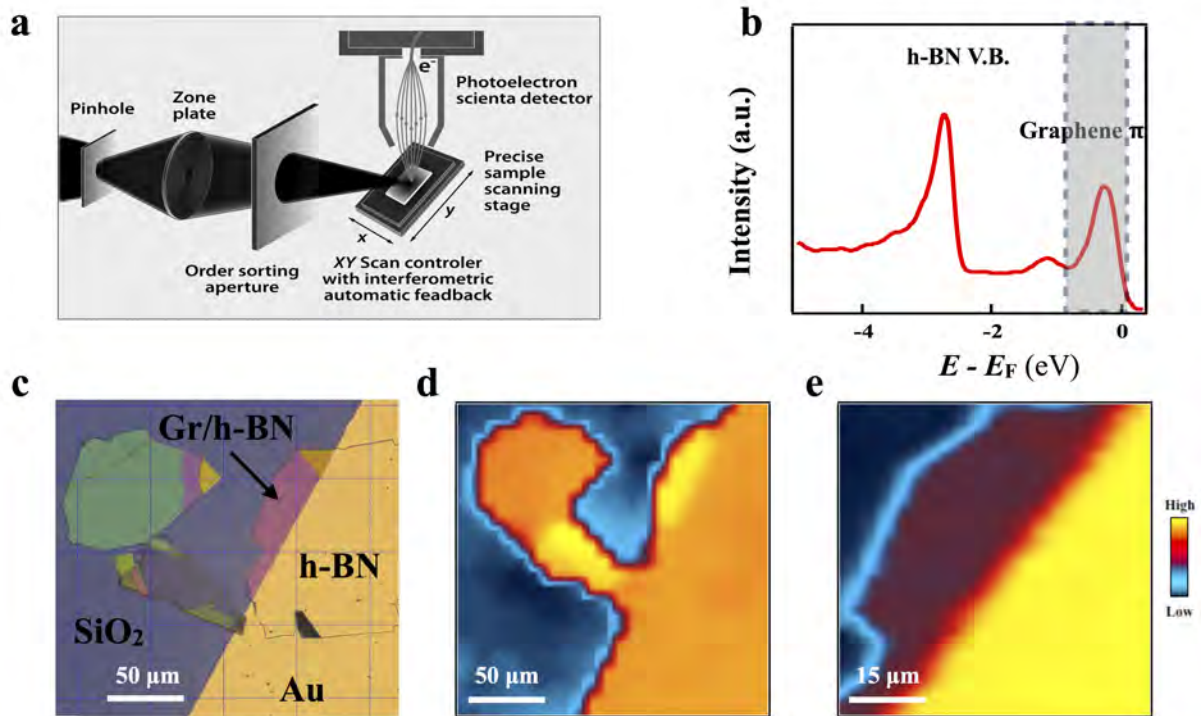
Figure 1. a, Working principle of NanoARPES. **b,** Valence band density of states of G/h-NB. The grey area indicates the energy window from which the valence band density of states images are integrated. **c**, Optical image of G/h-BN heterostructure. **d, e,** NanoARPES image of the valence band intensity.

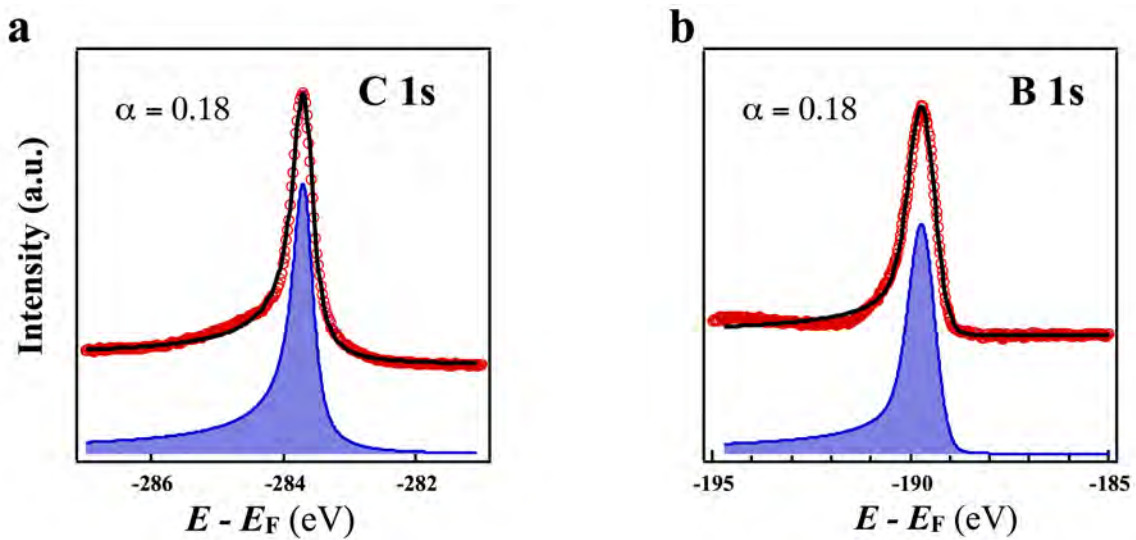**Figure 2. a,** C 1s core level. **b,** B 1s core level. The core level peaks are fitting by asymmetric Lorentzian line shape (blue). Red circles are the original data. Black solid lines are fitted spectra.
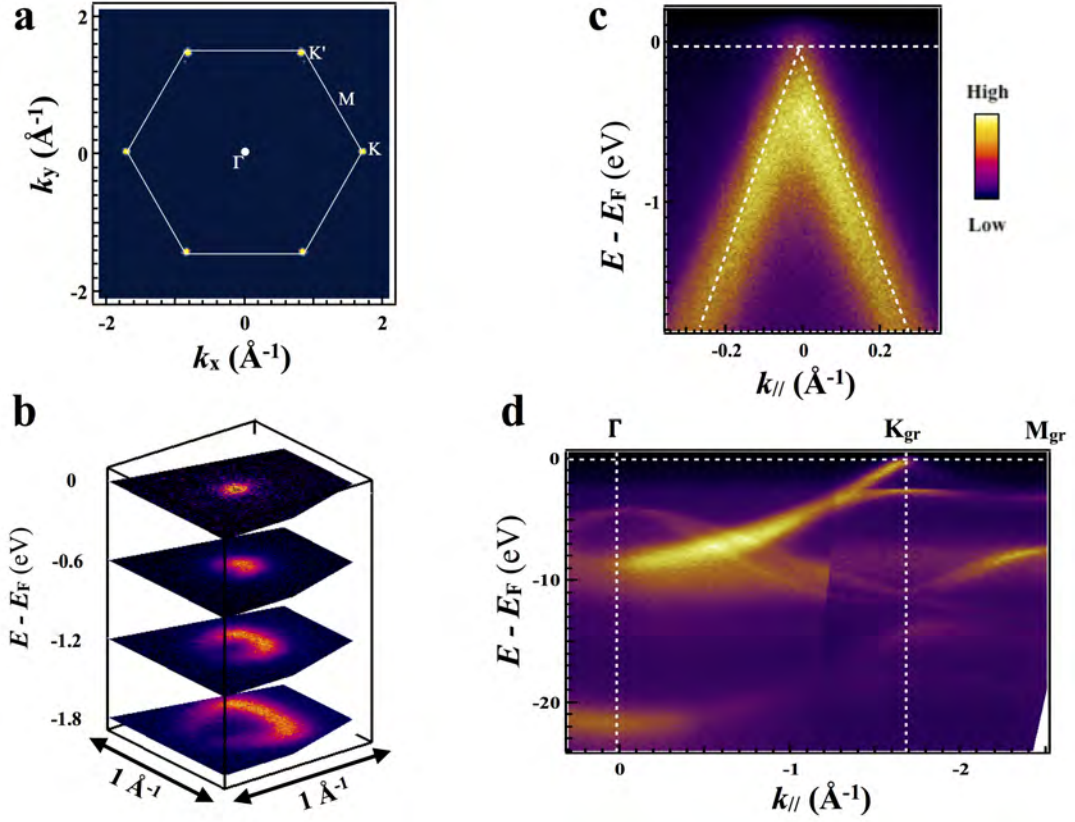
Figure 3. a, Symmetrised Fermi surface of G/h-BN . **b,** Spectral intensity distribution in E-k_x-k_y space. **c**, Detailed electronic structure of Dirac cone. **d**, Complete valence band structure of G/h-BN.

accuracy in the linear motion. Interferometers are used to control the simultaneous movement of sample and zone plate. By synchronised scanning of the sample along in-plane two directions and collecting the photoelectron intensity by energy analyser, images of the sample chemical composition (by element-specified core levels) and angle-resolved valence-band states can be obtained.

G/h-BN was prepared by PECVD growing graphene on exfoliated h-BN micro crystals. As described in the previous work[8], the growth of single crystalline monolayer graphene follows the van der Waals epitaxy mode, with zero crystallographic alignment angle respect to h-BN (Fig. 1c). SiO_2/Si was chosen as substrate for the convenience of transport measurement and also to avoid the possible interference in the valence band of G/h-BN from the d-bands of metals or transition metals. G/h-BN was grounded by Au contact for ARPES measurement (optical image in Fig. 1c).

As shown in Fig .1b, the spectra density of states of G/h-BN shows the peaks from h-BN valence band and graphene Dirac cone. By integrating the intensity within the selected energy window while scanning the sample along two in-plane direction, the spectral intensity distribution image can be generated, as shown in Fig. 1d and e. Compared with the optical image in Fig. 1c, it is clear that we can locate the G/h-BN sample (grounded) precisely with NanoARPES. From any position within the scanning range we can extract or remeasure the detailed electronic structure, including the core level spectra and valence band dispersion, as well as the Fermi surface, which will all be discussed in detail in the following.

3. Core level spectra of G/h-BN

Fig. 2 shows the core level spectra of C $1s$ and B $1s$, measured by NanoARPES with phonon energy of 350 eV. Both panels present sharp core level peaks. For a more quantitative analysis, we have fitted the peaks by a procedure involving the deconvolution of the peak with a Doniach and Sunjic analysis[12], using a Lorentzian line shape. Both C $1s$ and B $1s$ show an asymmetry parameter of 0.18 and the fitting results are satisfactory with the usage of a linear background.

4. Fermi surface, Dirac cone and valence band structure

We show here, for the first time, the complete Fermi surface of G/h-BN heterostructure. As proved in Fig. 3a, the Fermi surface of G/h-BN originates from the Dirac cone of graphene, composing a standard hexagon, with the K-K' distance $\sim$1.7 $\mathring{A}^{-1}$. This clearly indicates that there is no structural change for graphene from the weak van der Waals interaction with h-BN. The detailed Dirac cone structure plotted in Fig. 3b and c clearly proved that there is no perturbation for the standard Dirac cone. The graphene is non-doped. The linear fitting for the Dirac cone shown in Fig. 3c gives a Fermi velocity $\sim$ 7.5 eV $\mathring{A}$ ($\sim$1.1 $\times$ 10^6 m s^{-1}), in agreement with reported value[13].

The complete band structure plotting in Fig. 3d clearly show the sharp features of well-defined dispersions for valence band of G/h-BN. Generally speaking, the G/h-BN valence band can be treated as the superposition of free graphene and h-BN valence bands, however with detailed but significant modification to be discussed elsewhere[14].

Up to now we have shown in detail that, by employing NanoARPES, we are able to measure the complete electronic structure, including the core levels and valence bands, for G/h-BN heterostructure. This paves the way for future exploration of microscopic novel heterostructures' electronic structure by NanoARPES.

5. Acknowledgments

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