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Suppression of intrinsic roughness in suspended van der Waals heterostructures

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1. Introduction

Intrinsic roughness/ripples

- Ripples in graphene are ubiquitous – seen for both for graphene on hBN, SiO_2 [1] and suspended graphene [2].
- Ripples may be a large factor limiting carrier mobility [3, 4].

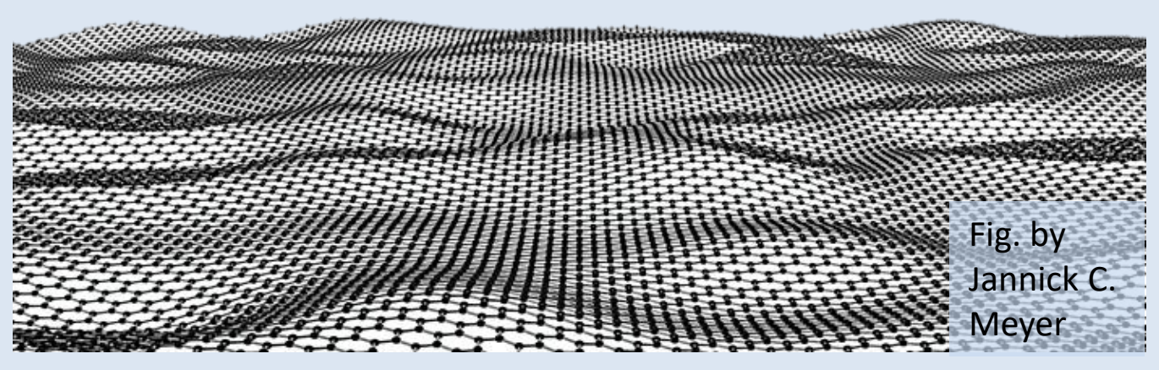


Fig. by Jannick C. Meyer

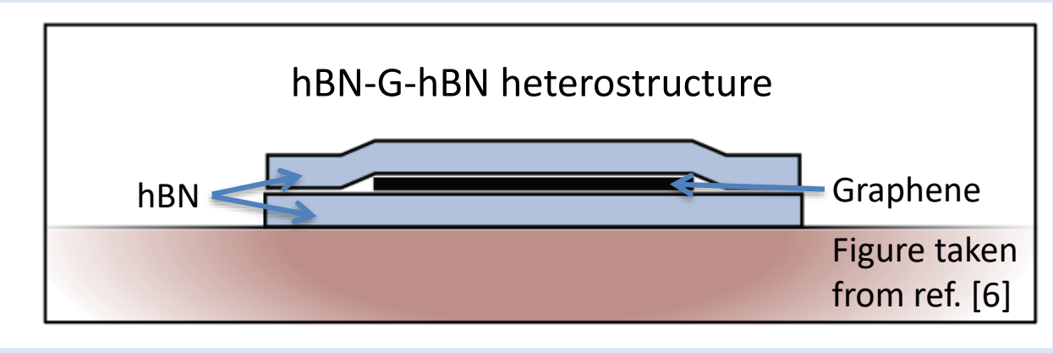


Figure taken from ref. [6]

Encapsulated graphene

- Making hBN-G-hBN heterostructures is a well known strategy for obtaining high quality grapene samples.
- Measurements of supported graphene on hBN have shown root mean square roughnesses (R_{RMS}) values of 100 pm even for thick hBN flakes [1].
- Here we show that the roughness can be reduced further by suspending the heterostructure.

2. Methods – In-situ TEM and Fabrication

Reciprocal space of rough graphene

The full 3D Fourier transform of rough graphene in reciprocal space (u,v,w) consists of a set of cones (Fig. 1).

If the graphene is rough the diffraction spots become diffuse when tilting the sample.

For G/hBN samples the spots remain the same.

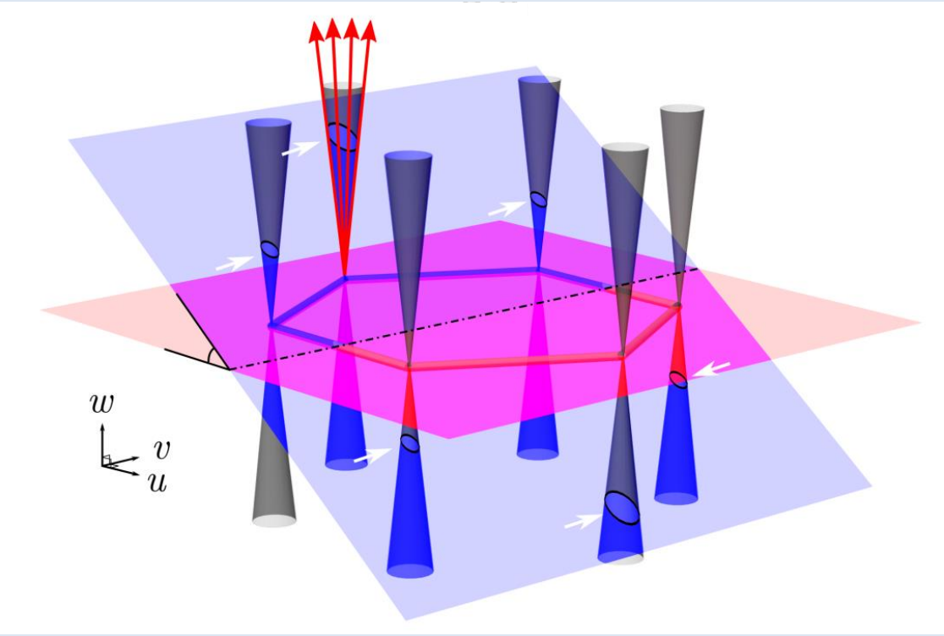


Fig. 1 – the 3D Fourier transform of rippled graphene consists of cones. The blue and red plane represent the Ewald sphere intersecting the density distribution to form diffraction patterns.

Roughness

The spot intensity of rough graphene varies as $I \propto \exp(-(2\pi G)^2\langle h^2\rangle)$, where $\langle h^2\rangle$ is the R_{RMS} [5]. Hence

$$\sqrt{\langle h^2\rangle} = \frac{1}{2\pi}\sqrt{\frac{d\ln(I)}{dG^2}}$$

Sample fabrication

The samples were made using the hot pick-up method (Fig. 3c) [6].

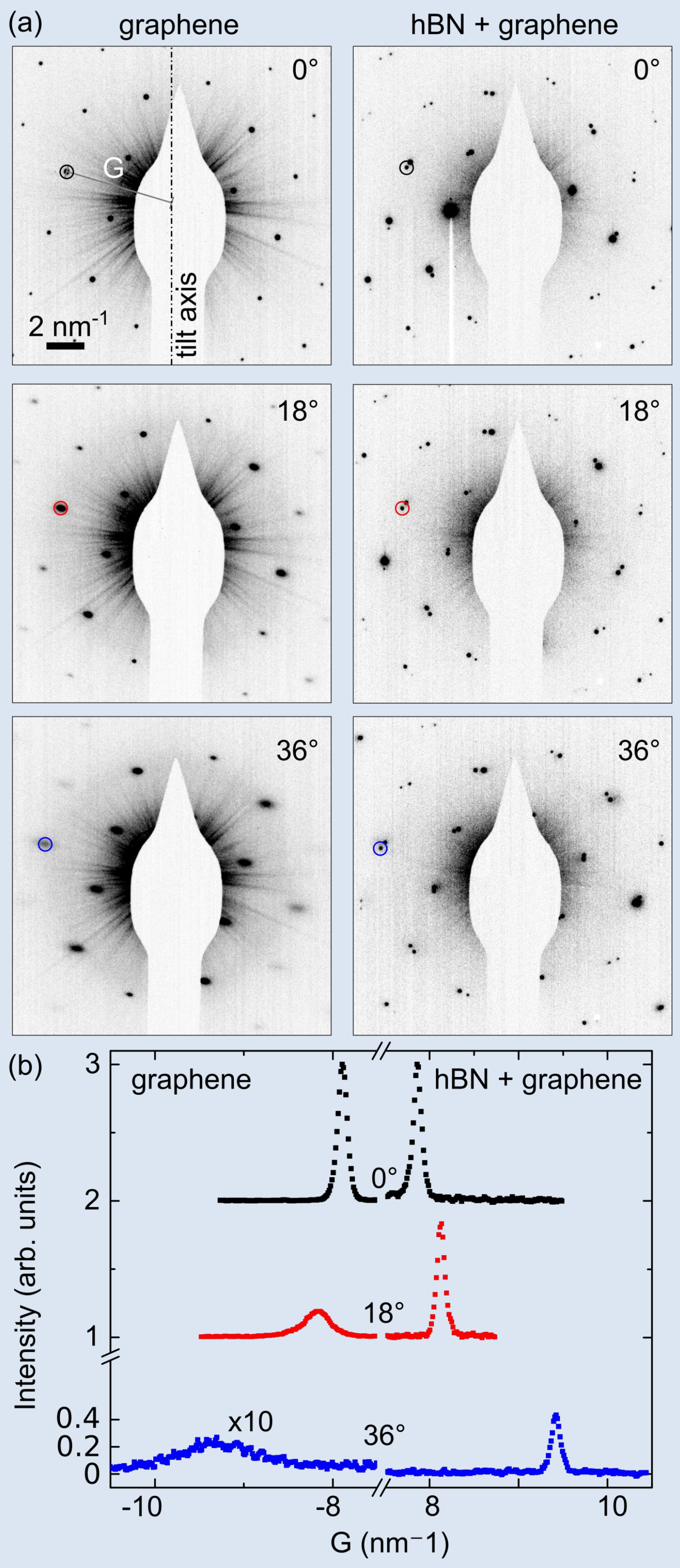


Fig. 2 – (a) diffration patterns of graphene and graphene/hBN at 0°, 18° and 36° tilt. (b) intensity of the circled second order graphene diffraction spots. Peaks are normalised to the 0° tilt intensity. Note that the intensity is 10x for suspended graphene at 36° tilt.

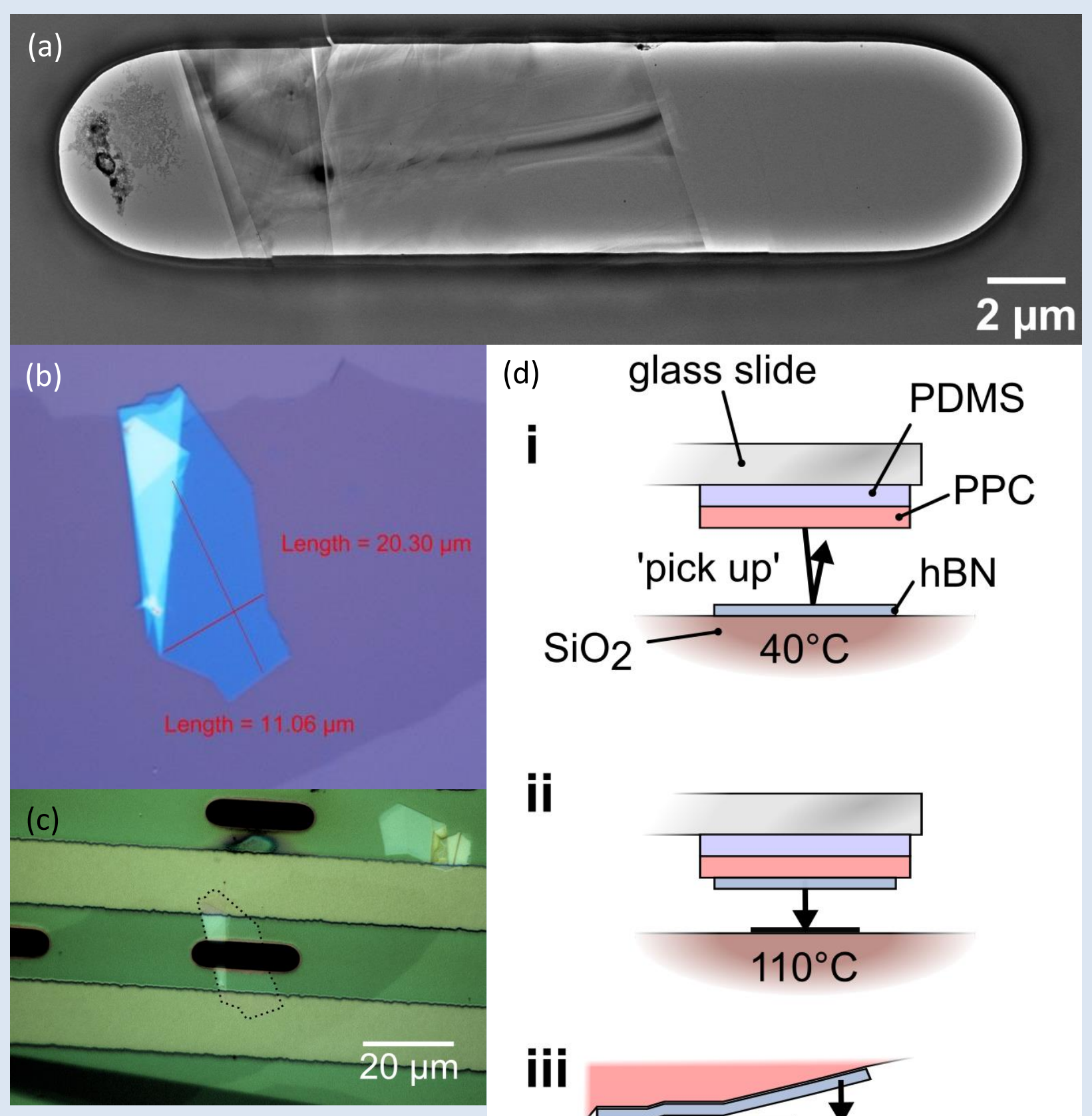
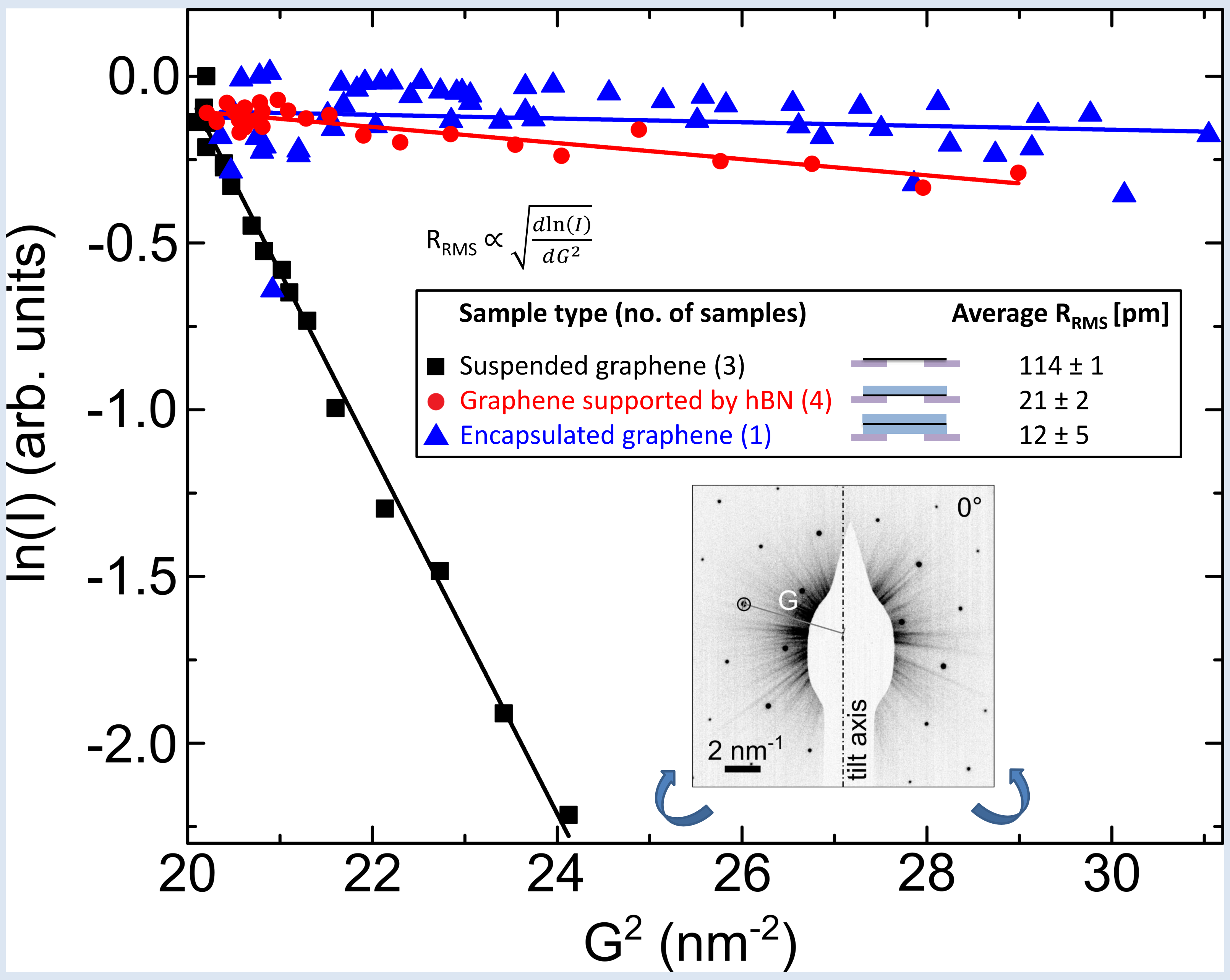


Fig. 3 – Sample fabrication. (a) hBN/graphene stack suspended over a hole in a TEM chip, (b) same hBN/graphene stack on SiO2, and (c) on TEM chip. (d) Hot pick-up procedure, (i) pick-up of hBN, (ii) drop down on graphene, (iii) release of polymer stack. Fig. taken from ref. [6]

3. Roughness Measurements

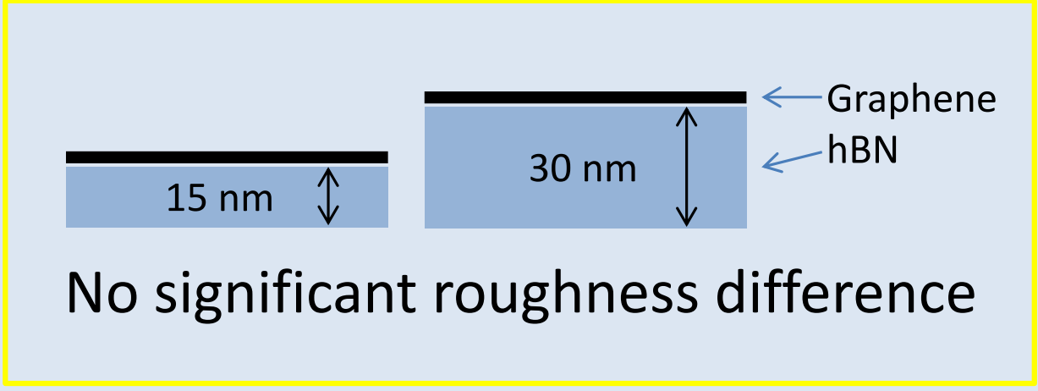


Sample type (no. of samples)	Average R_{RMS} [pm]
■ Suspended graphene (3)	114 ± 1
● Graphene supported by hBN (4)	21 ± 2
▲ Encapsulated graphene (1)	12 ± 5

Fig. 4 – plots of the logarithm of the diffraction spot amplitude as a function of the square of the distance from the zero order diffraction spot to the spot center. The slope of the linear fits is proportional to the RMS roughness of the graphene.

hBN thickness dependence

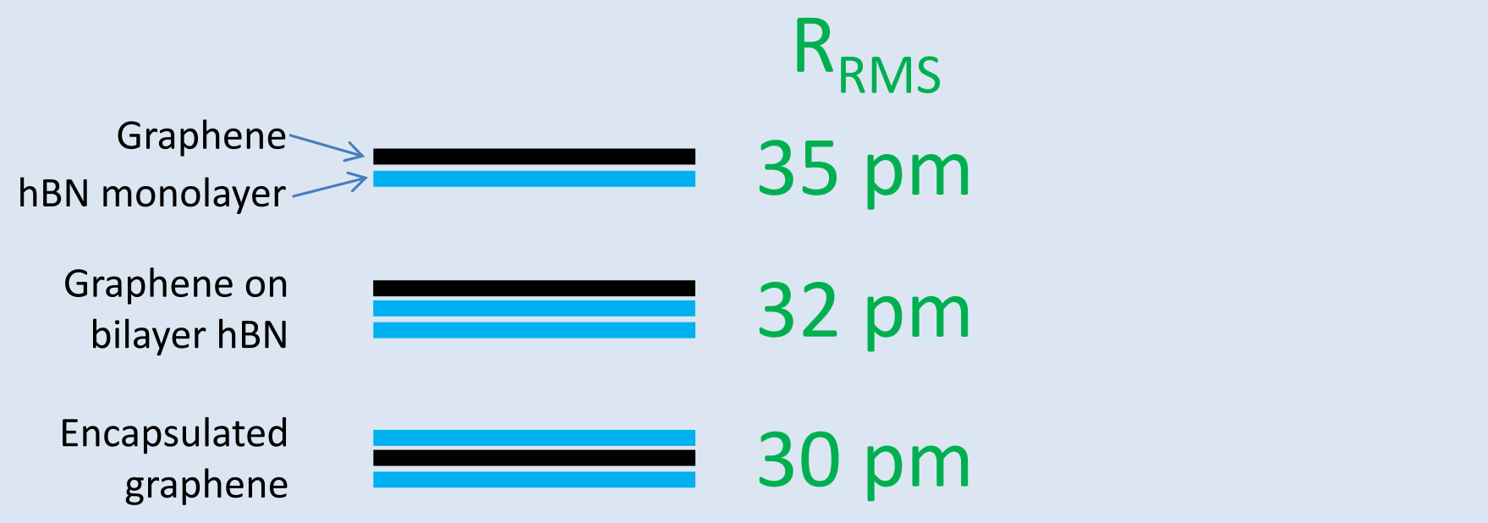
For graphene on hBN we measured two hBN flakes that were 15 nm and 30 nm thick, respectively, and found no dependence on roughness.



No significant roughness difference

4. DFT Simulations

From first principles calculations of hybridized phonon bands we calculate RMS out-of-plane atomic vibrations for carbon atoms in the following systems (all monolayers, 300 K):



	R_{RMS}
Graphene hBN monolayer	35 pm
Graphene on bilayer hBN	32 pm
Encapsulated graphene	30 pm

We find a stronger impact on flexural displacements by encapsulation compared to the effect on an additional layer. The low RMS carbon vibrations are due to localisation of the flexural acoustic phonon mode in the hBN.

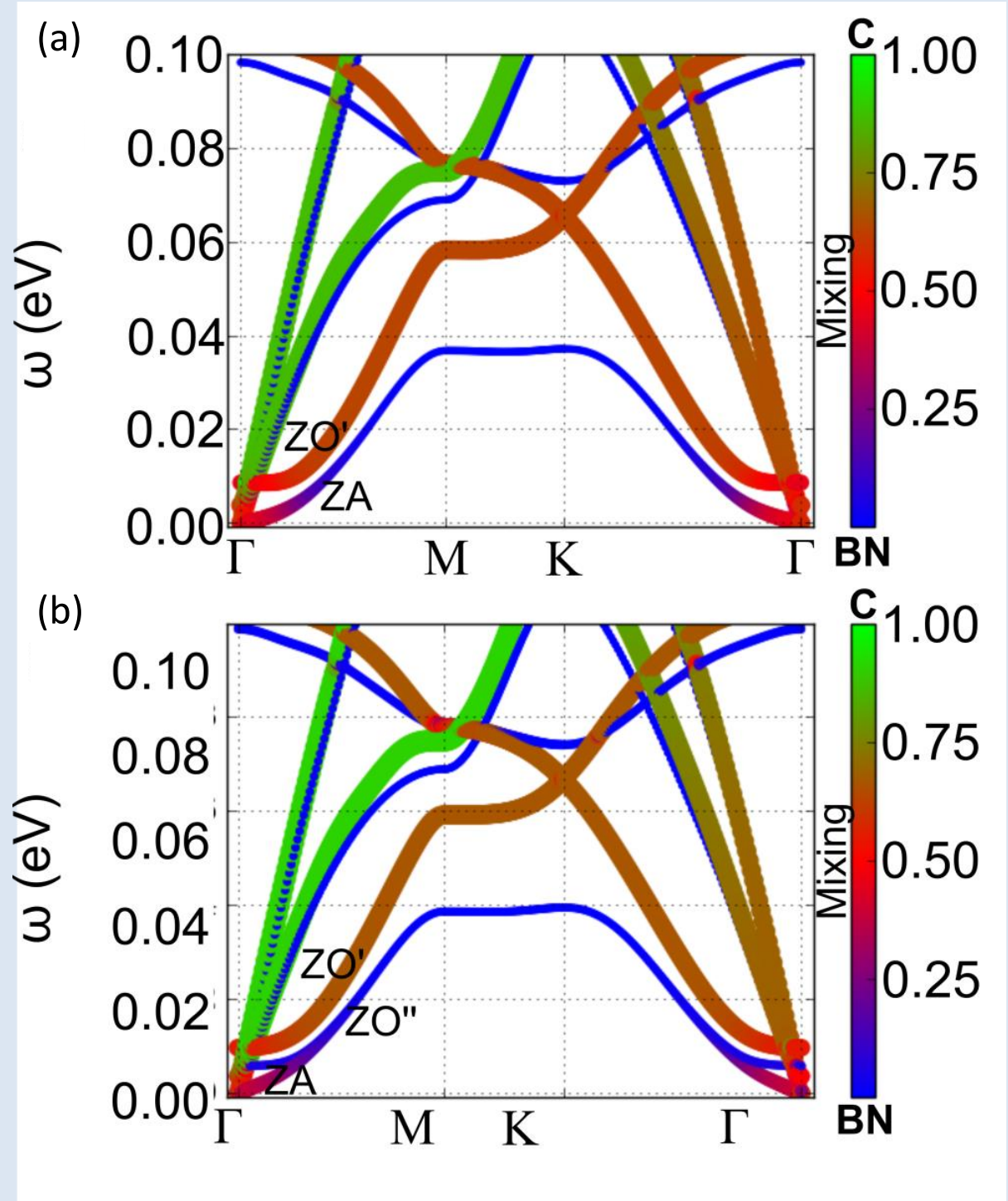


Fig. 5 – Phonon dispersions of (a) AB stacked graphene on monolayer hBN and (b) ABA' stacked graphene encapsulated in monolayer hBN with a C atom in between a B and N atom.

5. Conclusion

- By suspending the heterostructures we have measured a significant decrease in RMS roughness – 21 pm for hBN/graphene and 12 pm for hBN/graphene/hBN
- DFT calculations showed that carbon vibrations in graphene/hBN systems are suppressed due to localisation of flexural phonon modes in the hBN layers.

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