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, SiO₂ [1] and suspended graphene [2].
• Ripples may be a large factor limiting carrier mobility [3, 4].

Encapsulated graphene
• Making hBN-G-hBN heterostructures is a well known strategy for obtaining high quality graphene samples.
• Measurements of supported graphene on hBN have shown root mean square roughnesses (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.

Roughness
The spot intensity of rough graphene varies as I ∝ exp(−(2πG)²(β)²)), where (β) is the RMS [5]. Hence

\[ \beta = \sqrt{\frac{1}{2\pi^2} \frac{d\ln(I)}{dG^2}} \]

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

3. Roughness Measurements

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.

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):

RMS
Graphene on hBN
Graphene on bilayer hBN
Encapsulated graphene
35 pm
32 pm
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.

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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