STEM and EELS study of the Graphene/Bi₂Se₃ Interface

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Bi₂Se₃ is a 3D topological insulator (TI) that has attracted a lot of research interest due to its exotic properties [1], associated with topologically-protected helical two-dimensional surface states and one-dimensional bulk states associated with line defects such as dislocations. Recent theoretical studies [2] have shown that when graphene is placed near Bi₂Se₃ the strong spin orbit interaction due to proximity effects will open the band gap in graphene for 0.2 eV. Therefore TI/graphene heterostructures are promising platform for developing electronic and spintronic graphene based devices.

It has been recently shown that when Bi₂Se₃ is grown either on epitaxial graphene or free-standing graphene flakes [1,2], a rich grain structure develops due to the spiral nature of the film growth. The grain boundaries and growth-induced screw dislocations in this system provide grounds for interesting new physics that can be accessed by a combination of scanning tunneling microscopy and transmission electron microscopy techniques such as STEM-HAADF and EELS. In this work we investigate the nature of the graphene/Bi₂Se₃ interface in order to understand the complex epitaxy between the film and substrate as this ultimately determines the structure and functional properties of the graphene/Bi₂Se₃ interface topological states. The Bi₂Se₃ films were grown on epitaxial graphene/SiC(0001) and free-standing graphene (Figure 1a) by chemical vapor deposition.

The structure and electronic structure of the Bi₂Se₃/epitaxial graphene/SiC(0001) interface were studied in cross-section by state-of-the-art aberration-corrected STEM and atomically-resolved EELS. HAADF imaging (Figure 1) confirmed the presence of an epitaxial carbon layer at the Bi₂Se₃/epitaxial graphene/SiC(0001) interface. Combined imaging and atomically-resolved EELS maps confirm that both the Bi₂Se₃/carbon and carbon/SiC(0001) interfaces are atomically sharp with a Se termination of the Bi₂Se₃ layer. Analysis of the C K fine structure across the interface stack confirms that the observed interface carbon layer is in fact epitaxial graphene, indicating that the bonding between the Se atomic plane and the epitaxial graphene has a Van der Waals nature. This weak bonding is further corroborated by strain analysis of the HAADF images, showing the absence of strain at the Bi₂Se₃/epitaxial graphene/SiC(0001) interface. Such weak bonding would be the key factor for the multiple epitaxial relations which leads to both low- and high-angle boundaries observed in Bi₂Se₃ thin films when grown on graphene substrate [3,4].

References:

[1] Y Ando, J. Phys. Soc. Jpn **82** (2003), p.102001

[2] L. Kou et al., Nano Lett. 13 (2012), p. 6251

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- [3] Y Liu et al., Phys. Rev. Lett. 110 (2013), p.186804
- [4] Y Liu et al., Nat Phys, **10** (2014), p. 294
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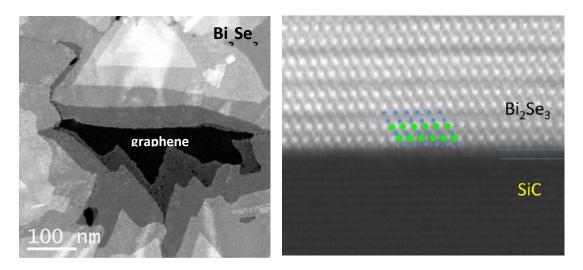


Figure 1. a) planar view (HAADF STEM) image of a Bi₂Se₃ film grown on epitaxial graphene showing the spiral growth of the Bi₂Se₃ film and b) cross-sectional view (HAADF STEM image) of the Bi₂Se₃/epitaxial graphene/SiC(0001) interface showing an epitaxial graphene layer.

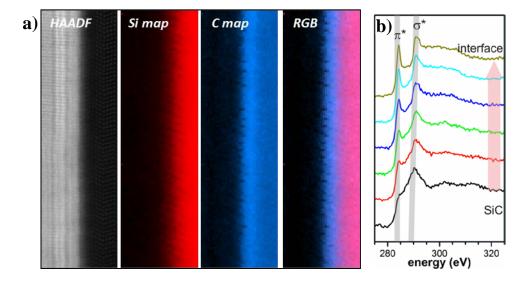


Figure 2. a) cross-sectional view (HAADF STEM image) and EELS maps confirming the presence of an epitaxial graphene layer at the Bi_2Se_3 /epitaxial graphene/SiC(0001) interface and C K spectra across the interface showing the sp^2 bonding character of the epitaxial graphene layer.