Atomic composition of the SiC/graphene structure obtained by high-temperature sublimation process

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We report some new insights into elemental composition of the SiC/buffer layer (BL)/graphene layers using transmission electron microscopy (TEM) characterization techniques and density functional theory (DFT) calculations. Previous studies have shown the atomic rearrangement of the BL and the further growth of the graphene layers by assuming the stoichiometry of the SiC substrate to be maintained despite the high-temperature sublimation process used for the growth of graphene[1,2]. We record scanning (S)TEM images at the SiC/BL/graphene interfaces in the cross-section geometry (Fig. 1a). Their direct comparison with STEM simulation[3] of perfect structural atomic model shows a discrepancy at the BL contrast (Fig. 1b). The perfect structural model is relaxed using DFT calculations. The difference between the STEM simulations of the relaxed and none-relaxed structures is shown Fig. 1d. The experimental intensity of the buffer layer is better reproduced for the STEM simulations based on the DFT relaxed structure. However, the amplitude of the experimental STEM profile in the SiC substrate is found to decrease toward the interface, it could be due either to the substitution of Si by C atoms and/or by the presence of vacancies (V). Atomic resolution electron energy-loss spectroscopy (EELS) in STEM mode configuration is performed to determine the absolute concentration of Si, C, V_{si}, and V_C (Fig. 2b). The vertically averaged EELS profile shows the decrease of the Si and C concentrations toward the interface as well as the presence of Si in the BL (Fig. 2c). The gradual modification of the C fine structure is also observed. Based on the concentrations and fine structure information obtained from EELS measurements, a new atomic model has been created and examined by DFT calculation. Then, we used new relaxed structure as input for the STEM simulations and test the results against the experimental STEM image. The obtained results will enable us to gain deep insights into true stoichiometry of the SiC and BL layers, which is essential for future commercial applications of epitaxial graphene grown by high-temperature sublimation process.

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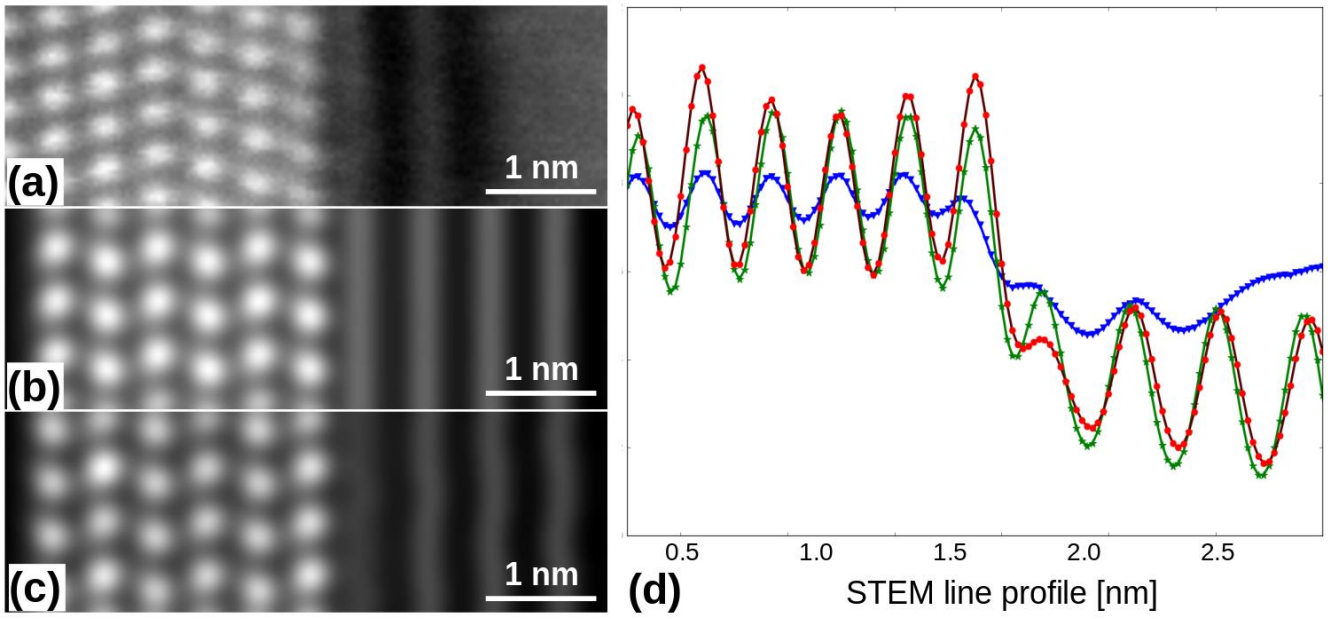

Figure 1: SiC/BL/graphene cross-section data. (a) High-angle annular dark-field (HAADF) STEM experimental image, (b, c) STEM simulation of the perfect (b) and DFT relaxed (c) structures. (d) Vertically averaged intensity profiles of the experimental data (blue), perfect (orange) and DFT-relaxed (black) structures. The experimental data has been scaled to fractional intensities matching the mean value of the simulation.

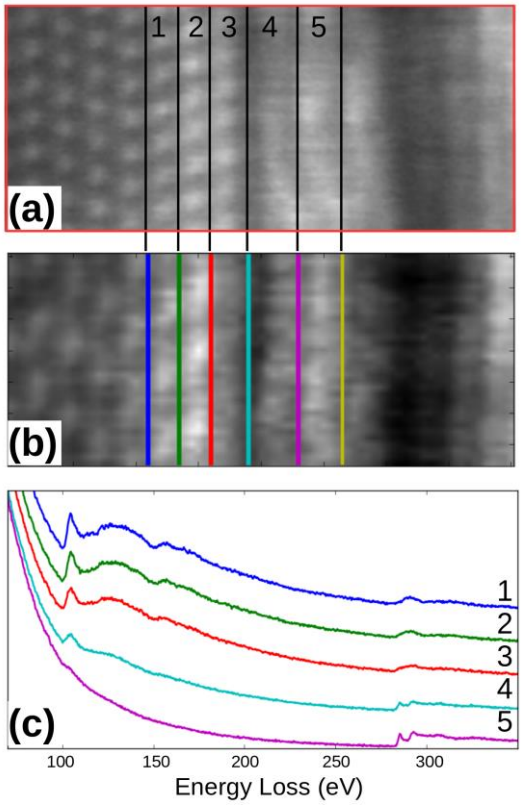

Figure 2: SiC/BL/graphene cross-section data. (a) Experimental HAADF-STEM image recorded at full resolution prior the recording of the EELS data. (b) Live HAADF-STEM image recorded simultaneously with the EELS dataset. (c) vertically averaged EELS profiles at the positions shown in (a).