

Study of thickness uniformity and wrinkling in epitaxial graphene grown on SiC polytypes

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Abstract

Growth of epitaxial graphene was performed on the Si face of SiC substrates in an inductively heated furnace at a temperature of 2000°C and at an ambient argon pressure of 1 atm. We have studied thickness uniformity and wrinkling on graphene grown on 4H-SiC(0001), 6H-SiC(0001), and 3C-SiC(111) substrates. Graphene surface morphology, thickness, band structure, structure and electronic properties have been assessed by using AFM, LEEM, ARPS, STM, and STS respectively.

Graphene formation has been analyzed in respect to step bunching and surface decomposition energy differences created by the SiC basal plane stacking sequence on different SiC polytypes. Differences in the thickness uniformity of the graphene layers on different SiC polytypes is related mainly to the minimization of the terrace surface energy during the step bunching process. The uniformity of silicon sublimation is an important factor for obtaining large area homogenous graphene. It is also shown that a lower substrate surface roughness results in more uniform step bunching and consequently better quality of the grown graphene. We have demonstrated a monolayer (ML) graphene growth on all SiC polytypes, but larger area, over $50 \times 50 \mu\text{m}^2$, on cubic SiC (Fig. 1a). To study the surface restructuring during SiC sublimation we examined around 300 steps for each sample using AFM. The corresponding histogram of the step height for 4H-SiC (Fig. 1b) indicates that two bilayer-height steps are the most probable and four bilayer-height steps show a significant probability. For the 6H-SiC sample (Fig. 1c) two and three bilayer-height steps dominate. On the 3C-SiC graphene sample one Si-C bilayer height has the highest percentage (48%) of appearance although some larger steps are present (Fig. 1d). The sublimation rate of 3C-SiC is the same over the whole defect-free substrate surface due to the similar decomposition energy on all step terraces, this providing a uniform source of C on the surface which results in a superior uniformity of the grown graphene layer. It is worth noting that C contained in one unit cell (three Si-C bilayers) of 3C-SiC is sufficient to feed the formation of 1ML graphene. The 6H-SiC polytype shows close quality of graphene to that on the 3C-SiC polytype, because half of the unit cell contains three Si-C bilayers. The results for the 4H-SiC substrate coverage by graphene show that graphene formation process has narrower window of growth parameters. We have found that single Si-C bilayer steps with the same decomposition energy in the beginning of the graphene formation are the controlling factors for the uniformity of Si subtraction. Having a rather low step height distribution is one advantage of our results, since it has been reported that the resistance of epitaxial graphene on SiC increases linearly with step height on the substrate [1].

Wrinkling is a very general phenomenon in nature with dimensions spanning across length scales from meters down to nanometers. Graphene wrinkles (Fig. 2a) easily and often. They are larger in dimension and form by compressive strain induced during cooling from the growth temperature due to the

difference in thermal expansion coefficients of graphene and SiC. Wrinkles are linear defects which can cause carrier scattering and decrease mobility. [2] Deep understanding and sufficient control of the wrinkle appearance are central to our current research interest. By modifying substrate conditions we have been able to change the wrinkle orientation from a random network to a full alignment (Fig. 2b) in a particular direction or radial, by partially reducing strain. By these results we have found out how the step size and point defect can rule wrinkle morphology. The behaviour of wrinkles during thermal cycling at the same temperature and different temperatures and also cooling down to 4 K has been studied. We also examine to what extent the electronic and structural integrity of graphene is preserved upon wrinkle formation by STM and STS. We will also present, from typical AFM images, an approximate evaluation of strain for the top graphene layer forming wrinkle network on different SiC samples with diverse wrinkle morphology and size. We observed that the wrinkles appear very rare in SiC wafers in comparison with small size of SiC substrates. It seems that the larger size of graphene sheets can sustain more compressive strain and avoid wrinkling. Wrinkling was studied in a series of computer simulation. The simulations were performed with the molecular dynamic method using AIREBO forcefield which is quite suitable for simulation of hydrocarbons and the Tersoff potential for Si-C simulation. The simulated samples have about 150,000 atoms including both graphene and substrates. The results confirmed the experimental findings of wrinkle formation.

References

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 [2] S.V. Morozov, K.S. Novoselov, M.I. Katsnelson, F. Schedin, L.A. Ponomarenko, D. Jiang, and A.K. Geim, Phys.Rev. Lett., **97** (2006) 016801.

Figures

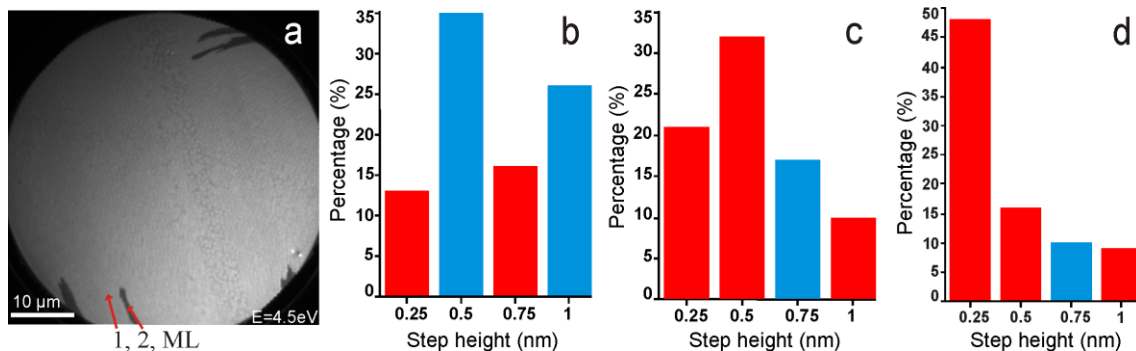


Fig. 1- a) LEEM image of graphene on 3C-SiC, Histograms of step heights for a) 4H, b) 6H, and c) 3C-SiC

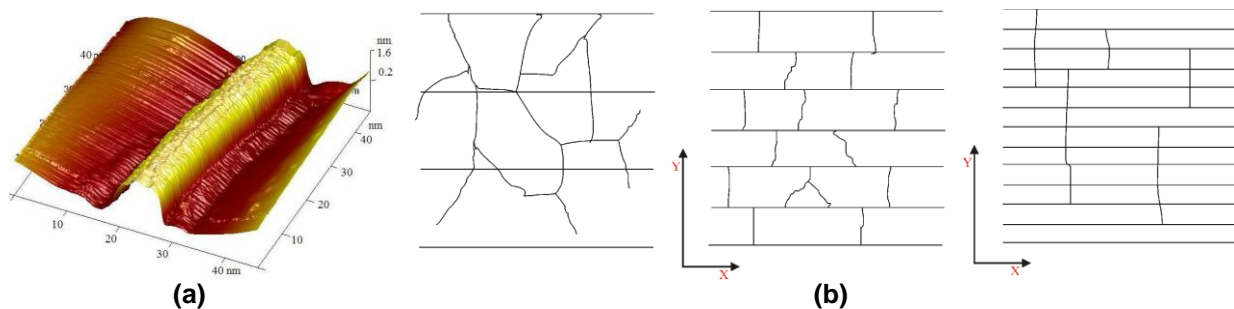


Fig. 2- a) STM image of part of a wrinkle b) Sketch of step size effect on wrinkles orientation