Investigating SiC/Graphene/SiC(0001) Remote Epitaxy Using Hot-wall CVD

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Remote epitaxy (RE) is a promising new technique for epitaxial film removal and substrate reuse that utilizes monolayer graphene as a release layer [1]. Graphene grown directly on SiC(0001) substrates through Si sublimation or through propane chemical vapor deposition (CVD) is an ideal platform for remote epitaxy of wide bandgap (WBG) semiconductors as there is no need for a graphene transfer step, reducing the risk of introducing contamination or defects that can complicate the study of the remote epitaxy process. In addition, this materials system is compatible with commercially-viable WBG semiconductor growth and processing. However, SiC CVD growth is typically conducted using high-temperature hydrogen-based chemistries that could damage or remove graphene. This study investigates the effect of alternate CVD growth conditions on SiC/graphene/SiC(0001) remote epitaxy and optimizes CVD parameters to produce high-quality SiC epilayers while reducing damage to the graphene barrier. In addition, since the effect of epitaxial graphene features such as SiC macrostep morphology and associated layer inhomogeneity on the RE process is currently unknown, graphene preparation and associated morphology is varied to explore its effect on SiC epilayer formation.

Semi-insulating nominally on-axis 6H-SiC(0001) and n-type 4° off-axis 4H-SiC(0001) substrates were used to produce different SiC surface morphologies and graphene layer numbers. Ar:H_2 process gas flow ratio, growth precursor C/Si ratio, and growth temperature were optimized during hot-wall CVD RE to promote smooth film morphology. Nomarski optical microscopy, scanning electron microscopy, and atomic force microscopy found CVD deposition at 1620°C with Ar/H_2 ratios <20/5 slm, and C/Si ratios <1.55 to have the smoothest surface morphology and fewest polytype inclusions. Substrates with offcuts <0.1° from SiC(0001) exhibited lower epilayer macrostep density, but showed evidence of polytype impurities and 3D growth at C/Si ratios > 1.0. Point defect density in RE SiC epilayers using a graphene interface was shown to be lower than SiC homoepitaxy using similar conditions without graphene. Cross-sectional transmission electron microscopy was utilized to assess the growth interface and graphene layer integrity after CVD growth. Through this study, optimal RE growth processes are suggested for a balance of graphene survivability and SiC film morphology.

[1] Kim, Y., Cruz, S., Lee, K. et al. Nature 544, 340–343 (2017).

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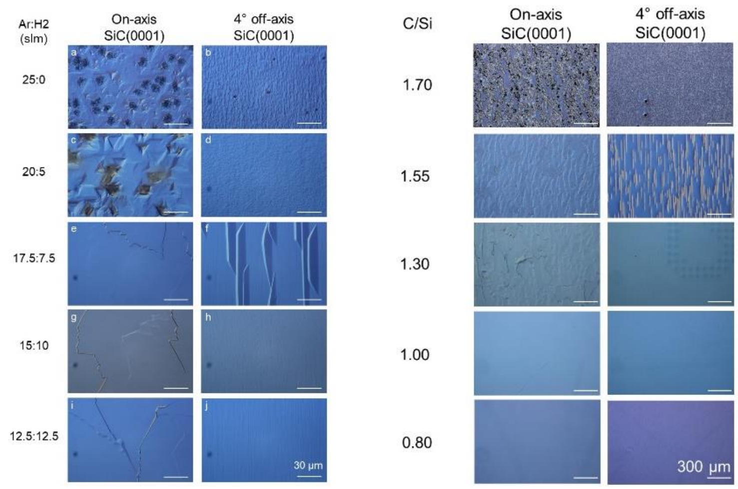
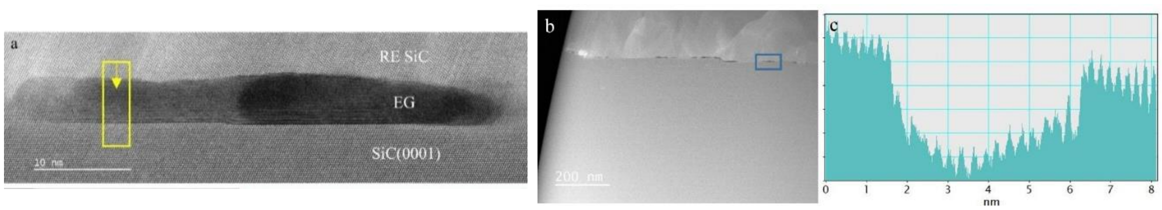
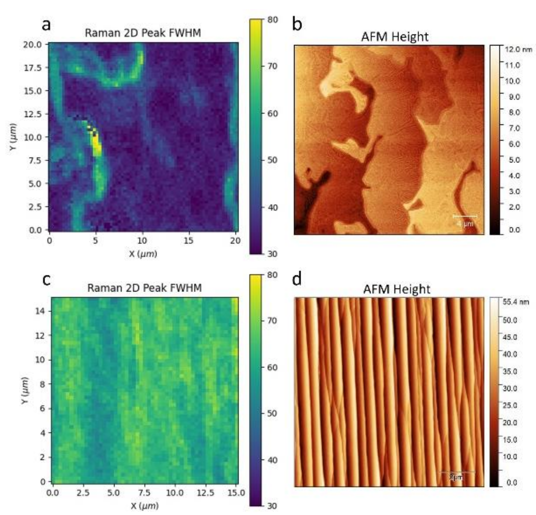
Supplemental Information



Nomarski micrographs for RE SiC films on epitaxial graphene (EG) substrates using various Ar:H_2 carrier gas ratios and a fixed C/Si ratio of 1.55 (left) and various C/Si ratios with a fixed Ar:H_2 ratio of 17.5:7.5 slm (right). Highest quality films are found with C/Si < 1.55 and Ar:H_2 < 20:5.



Cross-sectional HAADF-STEM of the SiC/EG/SiC(0001) interface. A close up of a multilayer segment of EG (a) shows multiple layers of graphene present in a small region. Lower magnification STEM (b) shows several multilayer EG regions at the interface. (c) An intensity profile taken from the yellow box in (a) shows the interplanar spacing of the multilayer graphene to be $d(0001)_G$ = 3.32 Å, close to graphite's interplanar spacing (3.35 Å)



Raman EG 2D peak FWHM maps (a, c) and AFM height images (b, d) of representative samples of ~1 ML EG grown on nominally on-axis, semi-insulating 6H-SiC(0001) (a, b) and ~4-5 ML EG on 4°-offcut, N-type 4H-SiC(0001) (c, d). FWHM color scale is measured in cm^{-1}.