Supporting Information: Low Temperature Growth of Graphene on a Semiconductor

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Photoelectron Spectroscopy Measurements

High-energy-resolution X-ray Photoelectron Spectroscopy (XPS) measurements of the gra/Fe/SiC and gra/Ru/SiC were performed at the MatLine and “Soft X-ray” (SXR) endstations of the synchrotron radiation sources ASTRID (Aarhus) and the Australian Synchrotron (Melbourne, Victoria), respectively. At MatLine, core levels Fe 3p, Si 2p, C 1s, Ru 3d and O 1s were measured with photon energies ranging from 120-600 eV using a SCIENTA SES-200 analyzer, with energy resolutions 100-850 meV. The exact photon energy used, and therefore the binding energy of each core level spectrum, was calibrated using the core level signal generated by second order harmonic light from the same excitation energy as the principal peak. At SXR, the same core levels were recorded using excitation energies ranging from 100-1200 eV using a Specs Phoibos 150 analyzer, with an overall energy resolution in this range of 150-175 meV. The binding energy of each peak was calibrated to the Fermi level position of the metalized SiC substrate recorded over the corresponding photon energy range. All core level spectra were fitted using the Levenberg-Marquardt nonlinear least square method, using asymmetric pseudo-Voigt approximations to the Mahan functions of each chemical component as described by Schmid et al.¹ For the core level backgrounds, an “active” approximation to the Shirley-Vegh-Salvi-Castle background² was used: individual contributions to the background from each chemical component peak were modeled by error functions and fitted dynamically together with all peaks and background components to simulate the full photoemission signal of the region.

NEXAFS Measurements

Near-Edge X-Ray Absorption Fine Structure (NEXAFS) measurements were performed at the SXR endstation, in partial electron yield (PEY) mode and using linearly polarized light at incidence angles roughly 20-100° relative to the sample plane. For each measurement of the C 1s K-edge, a reference sample of highly oriented pyrolytic graphite (HOPG), as well
as a grounded gold mesh were placed upstream from the analysis chamber. During each scan, NEXAFS spectra for the reference sample and the drain current on the Au mesh were recorded simultaneously using a small portion of the beam. Additionally, the absolute photon intensities coming into the analysis chamber were subsequently measured over the same scan regions using a calibrated photodiode. The recorded NEXAFS signal was normalized to the drain current on the Au mesh (for details, see Ref. 3), and the spectral response of the photodiode was used as a secondary normalization to correct for any carbon contamination on the Au mesh. Using the well-known exciton resonance in HOPG occurring at 291.65 eV, the spectra of the HOPG reference that was measured in parallel with each scan was used to absolutely calibrate the energy scale of the data via a rigid shift. All energy and intensity flux corrections were performed using the Quick As NEXAFS Tool (QANT).

Small Area LEEM, XPEEM, LEED and ARPES Measurements

Spatially resolved characterization of the surface graphene growth was performed using low-energy electron microscopy (LEEM), X-ray photoemission electron microscopy (XPEEM) and surface diffraction (µ-LEED). Measurements were carried out at the BL3.2Ub (aka “PEEM”) endstation at the Synchrotron Light Research Institute (SLRI), Muang Distrect, Thailand, and at the UE49-PGM-SMART endstation at BESSYII, Helmholtz-Zentrum Berlin, Germany. Additionally, bandstructure measurements were performed at UE49-PGM-SMART using small area angle-resolved photoemission spectroscopy (µ-ARPES).

Samples of gra/Fe/SiC and gra/Ru/SiC were prepared in a similar manner at the two facilities, using similar settings for the LEEM/PEEM instruments wherever possible. Clean 6H-SiC(0001) was confirmed by the lack of O 1s signal seen from plane XPS measurements, as well as a (1 × 1) or a ($\sqrt{3} \times \sqrt{3}$)R30° reconstruction, indicating an atomically clean surface. Small differences in C-C signal from the C 1s were also observed. Neither the difference in
initial surface reconstruction nor the mentioned differences in XPS signal had any significant impact on the subsequent growth steps.

Metals were deposited using calibrated Fe and Ru sources, and the thickness of each film was confirmed from XPEEM and LEEM measurements. All \(\mu\)-LEED measurements were recorded from selective areas of 1.5 \(\mu\)m diameter. Furthermore, similar measurements were performed in several different regions within a roughly 500 \(\mu\)m range of each sample to verify the homogeneity of the surface layers. A 40 eV excitation energy was used for all \(\mu\)-LEED measurements, except the diffraction patterns after graphene formation at 620 °C (gra/Fe/SiC) and 800 °C (gra/Ru/SiC). Here, 140 eV and 50 eV were used, respectively, to reveal both the features native to graphene and the underlying SiC substrate. The \(\mu\)-ARPES measurements of the graphene formed was recorded from 1.5 \(\mu\)m areas at room temperature. The measurements were performed with excitation energy \(h\nu = 115\) eV, in energy steps of 100 meV and with an overall energy resolution of 500 meV.

References

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