

On the Way to Twisted Bilayer Graphene: Formation and Decoupling of 0°-Rotated Epitaxial Graphene

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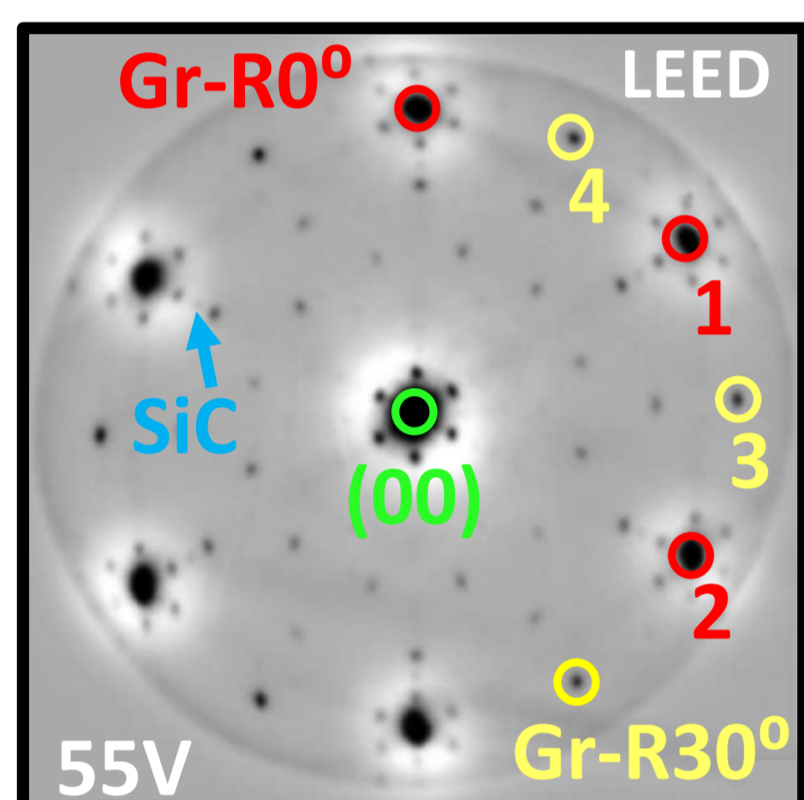
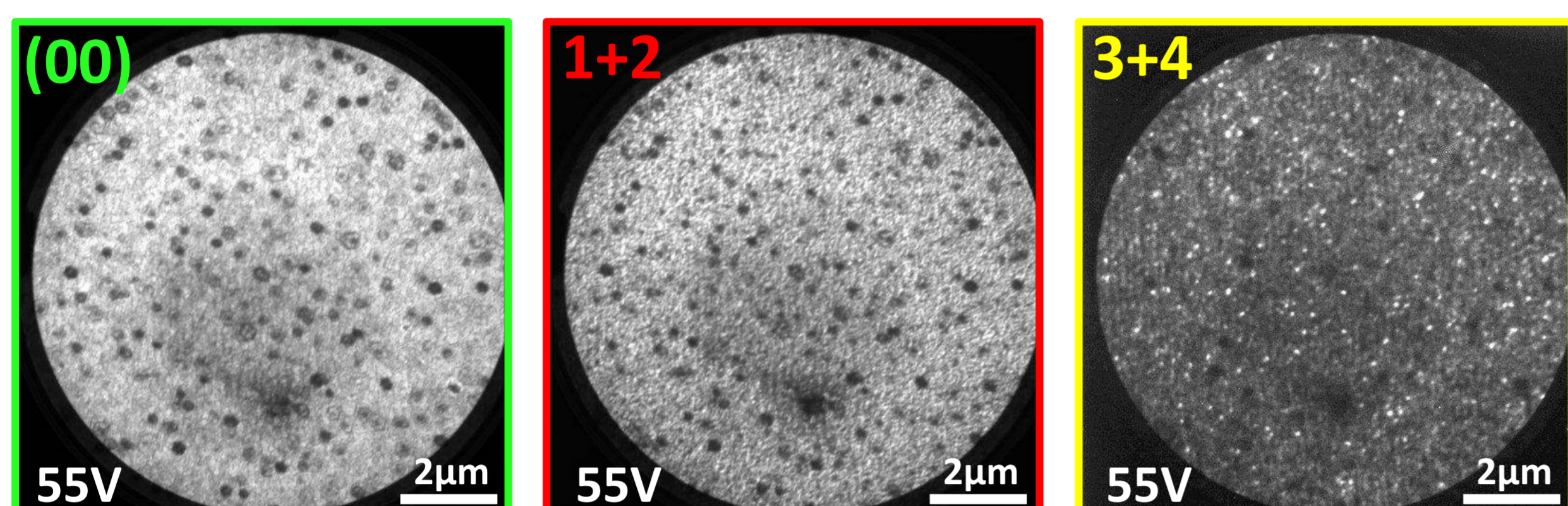
Introduction

In the exploration of obtaining twisted bilayer graphene (TBG) with a twisting angle of 30°, we investigated the graphene growth on 6H-SiC(0001) using an unconventional epitaxial method named “surfactant-mediated growth”, which is based on annealing the SiC surface in borazine atmosphere. Here, we report a LEEM-based study on two different samples, on which we observed different surface morphologies with varying number of stacked graphene layers.

Multilayer Graphene R0° and R30°

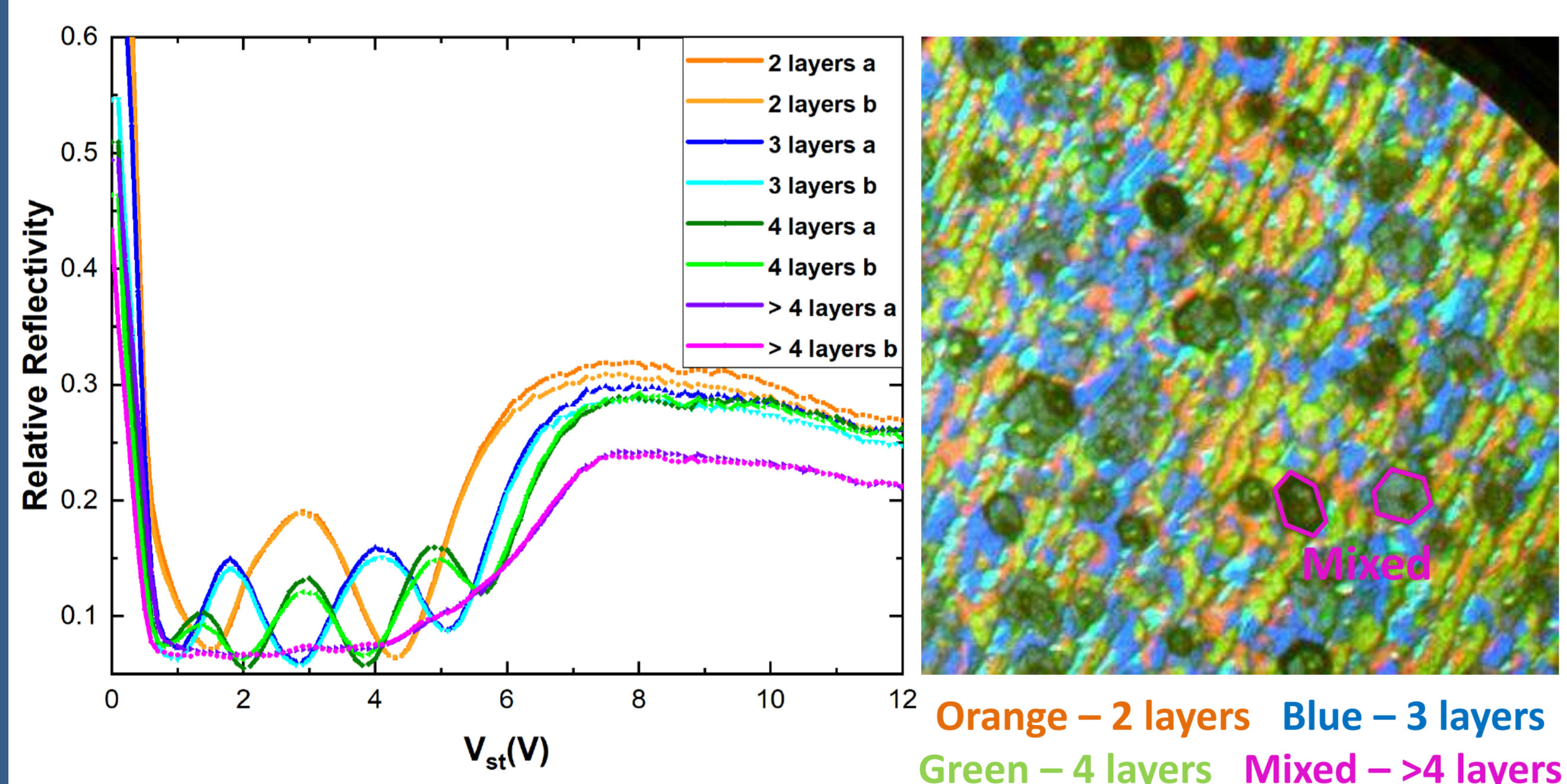
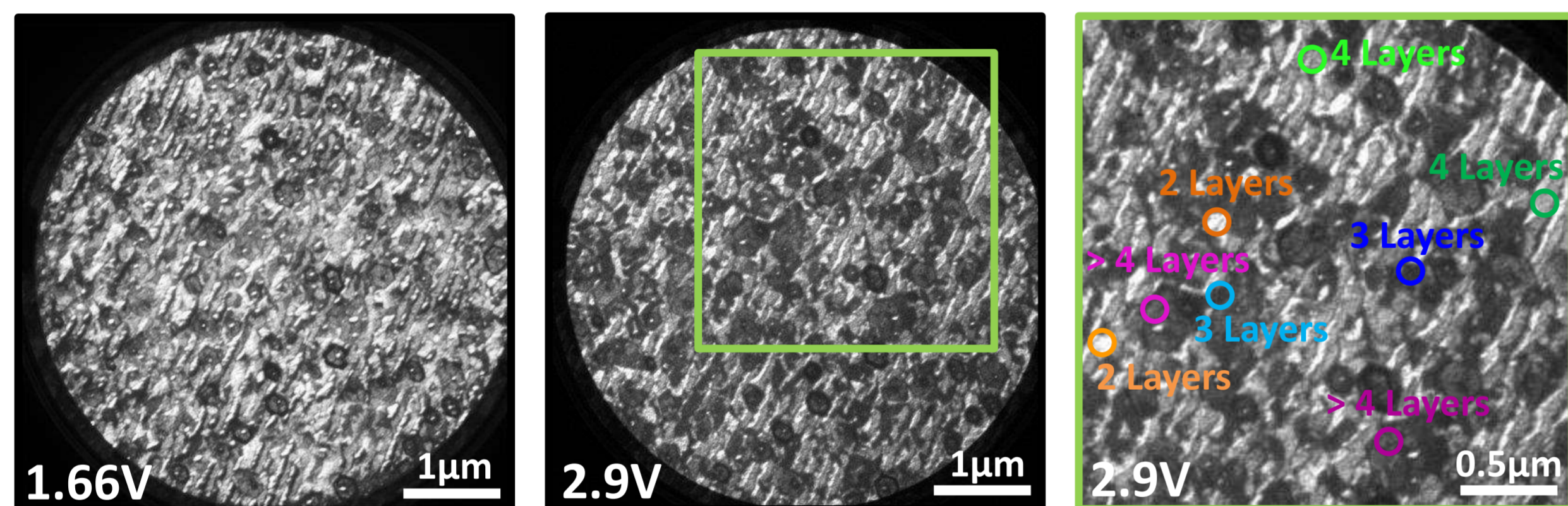
DF-LEEM

Annealed to 1380 °C



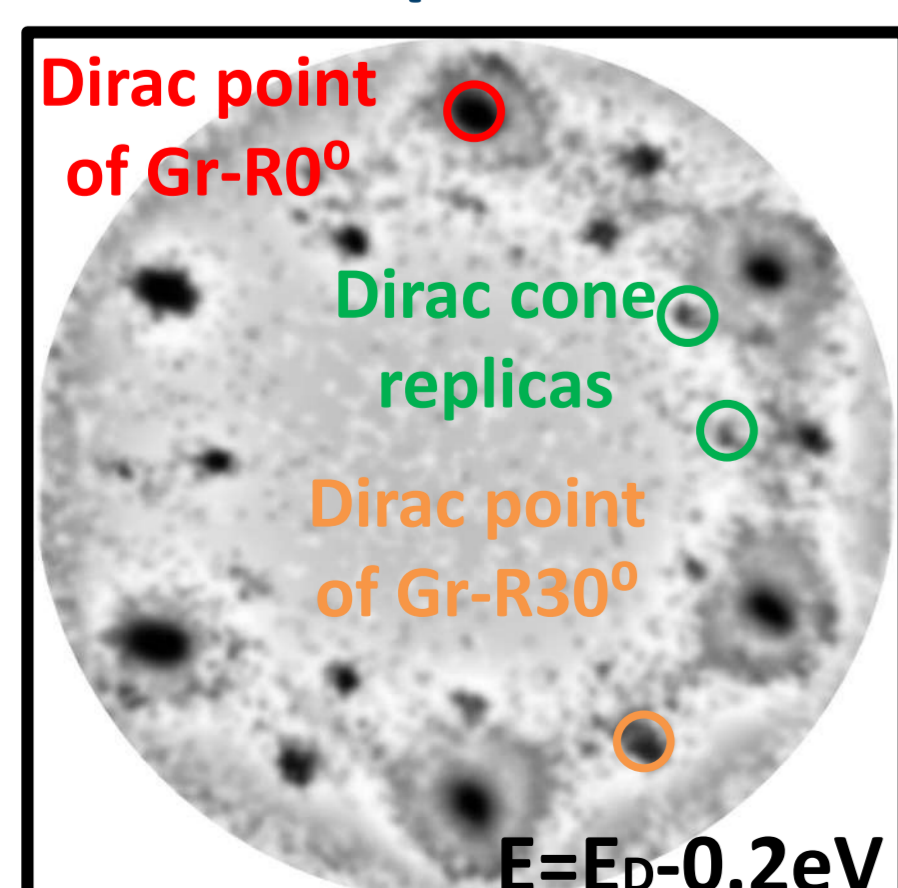
- The surface is much more inhomogeneous than for the 1330°C sample ((00) and '1+2');
- Image '(00)' and '1+2' are very similar → Gr-R0° is present (almost) everywhere on the surface;
- Gr-R30° appears randomly as small bright spots (image '3+4');
- Substrate LEED spots are weaker than for the 1330°C sample → Multilayer graphene.

LEEM-IV



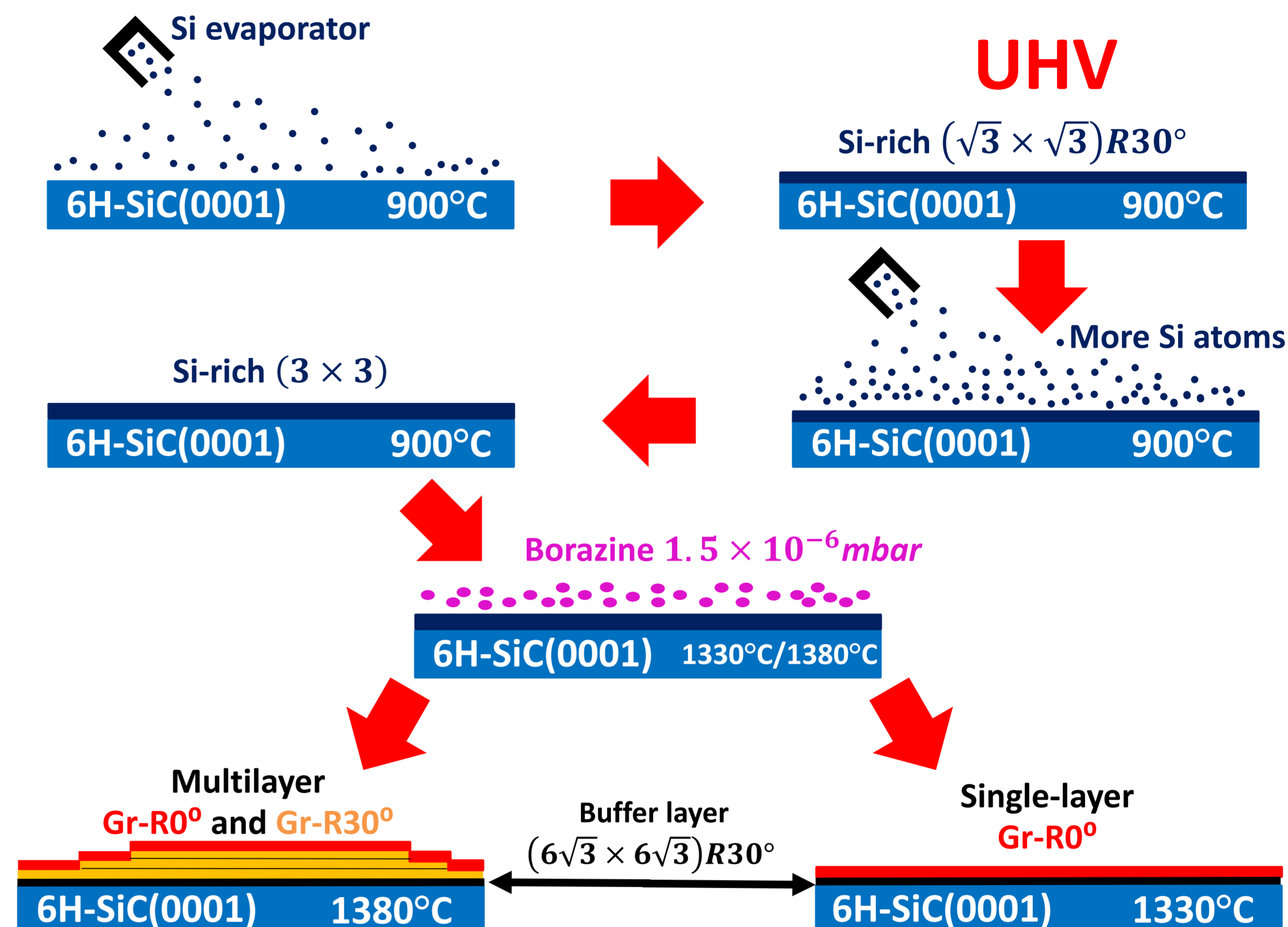
- Small domains with various number of graphene layers, corresponding to the number of minima in the IV curves;
- Domains with 1 to 4 layers clearly identified, and areas with more than 4 layers;
- The false color composite image presents the distribution of the different multilayer graphene domains.

ARPES (Constant Energy Map)



- Dirac points of Gr-R0° and Gr-R30° can be distinguished, with a much higher intensity for Gr-R0°. Twelve Dirac cone replicas (in green circles) can be explained by Umklapp-scattering in 30°-rotated TBG layers.

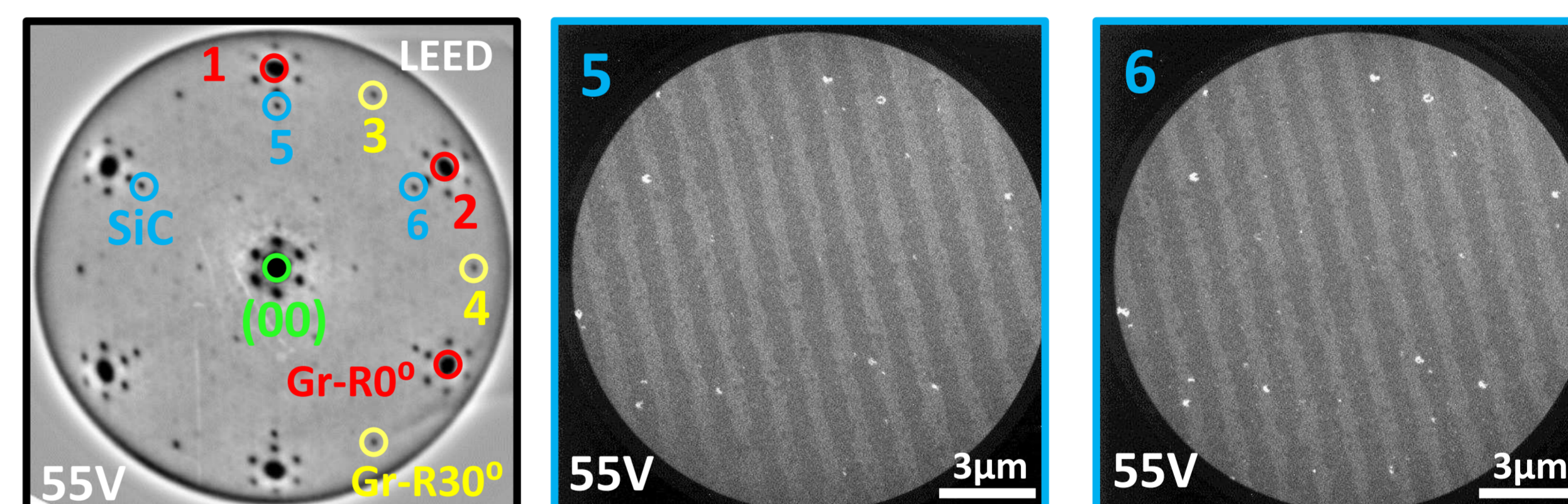
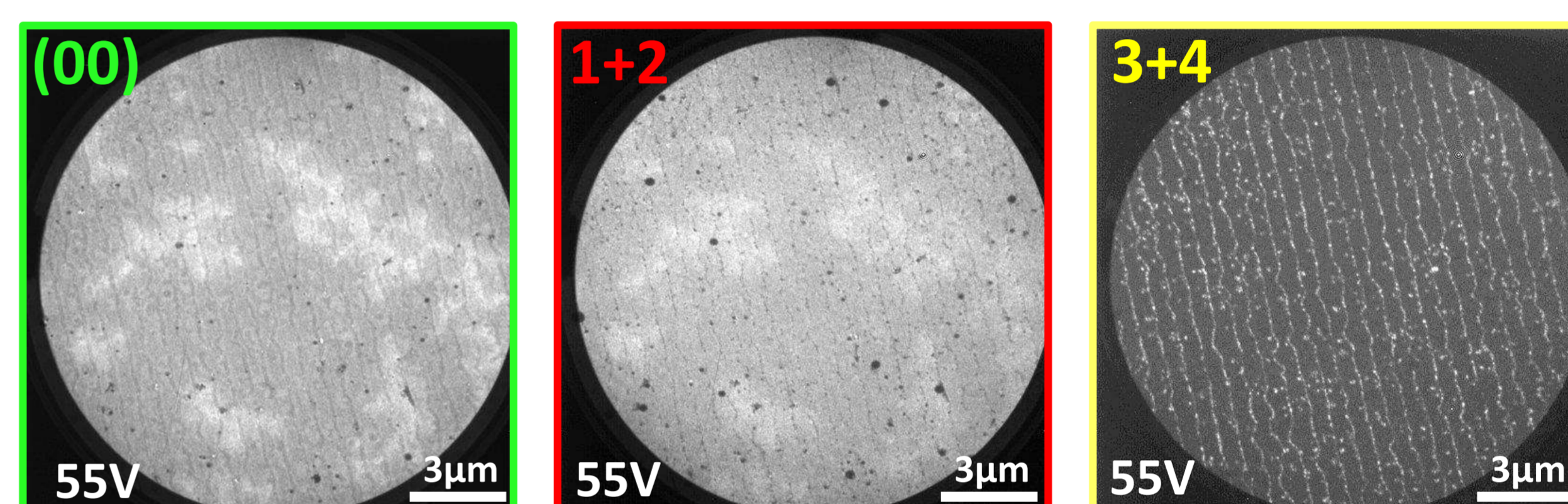
Surfactant-Mediated Epitaxial Growth



Single-layer Graphene R0°

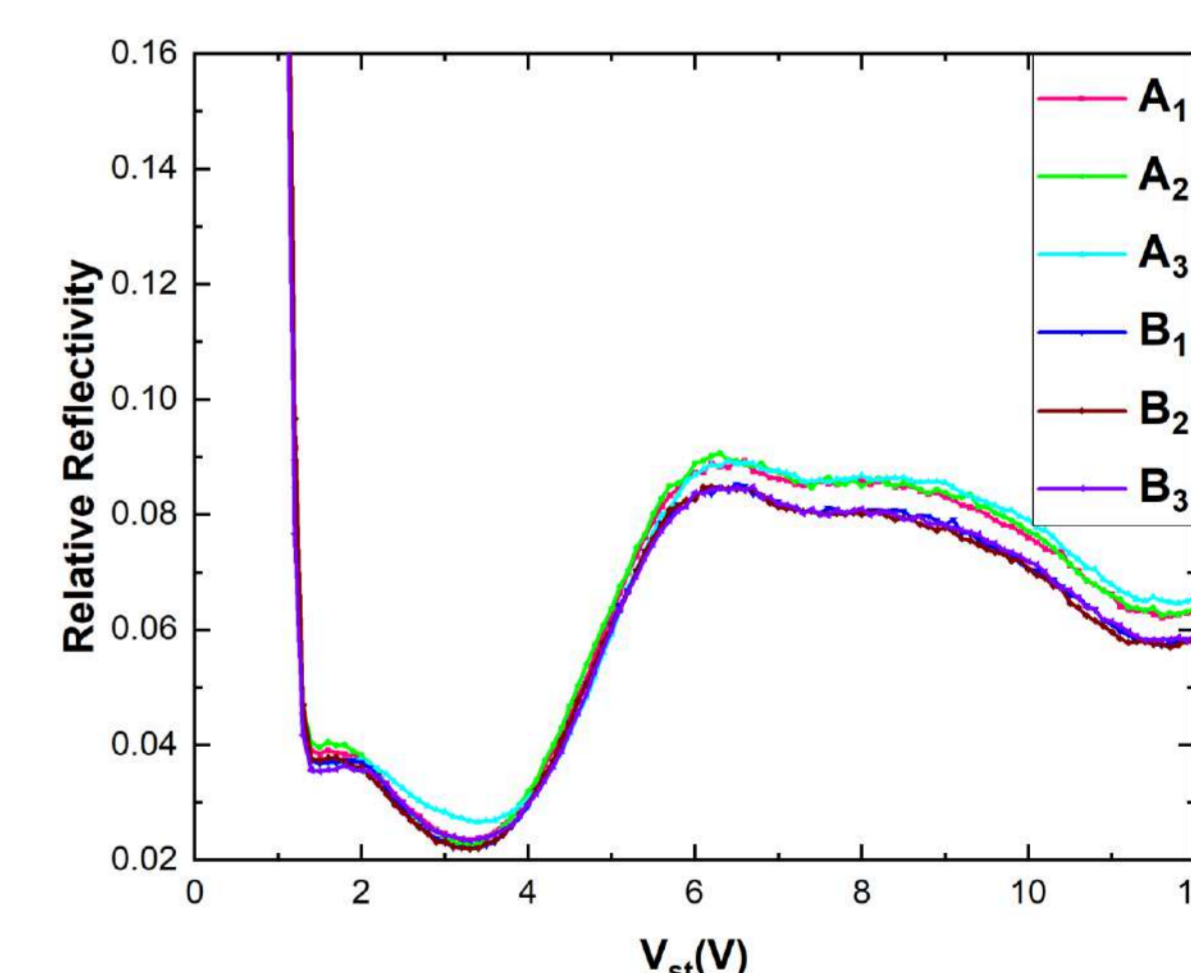
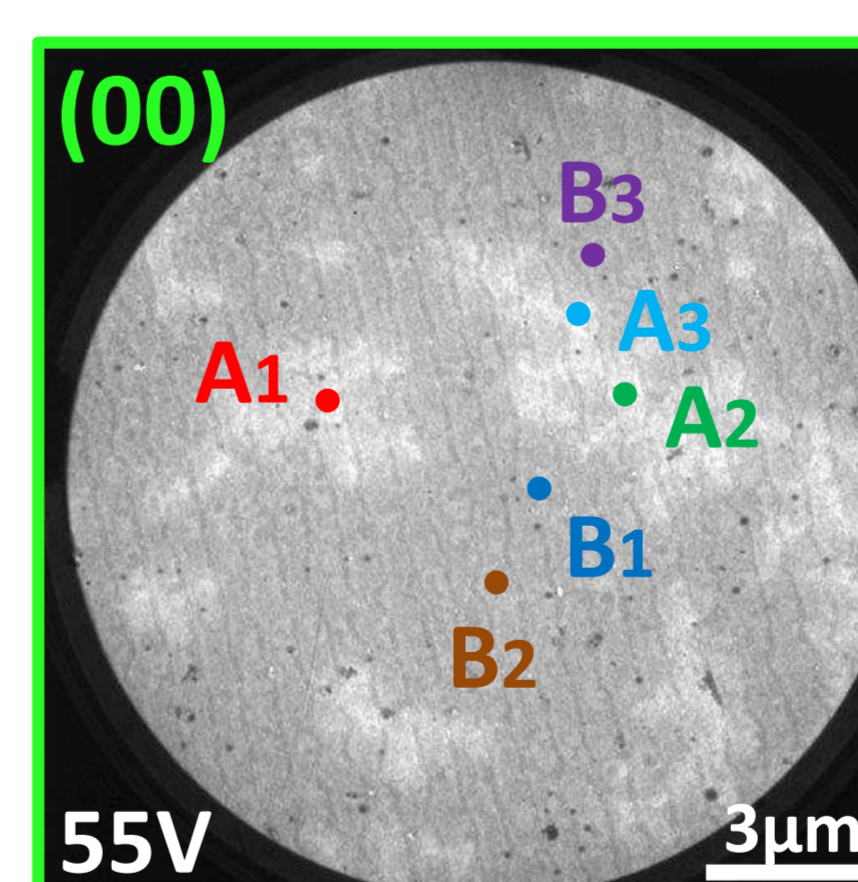
DF-LEEM

Annealed to 1330 °C



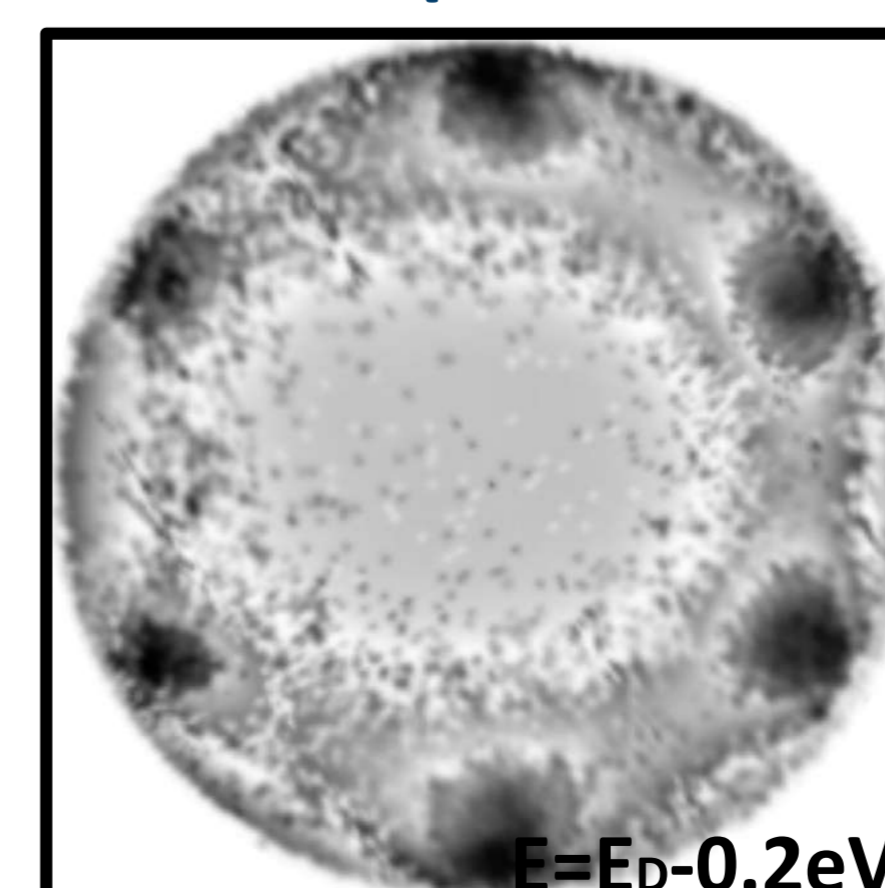
- Uniform Gr-R0° layer on large scale (> 200μm) (image '1+2');
- Gr-R30° appears at step edges and a few spots on terraces ('3+4');
- Stripes with alternating brightness in images '5' and '6' reveal the substrate symmetry of 6H-SiC regarding the 60° orientation difference between neighboring terraces.

LEEM-IV



- Only one minimum in the energy range of 0-7V indicates a single graphene layer on the sample surface;
- Brightness contrast exists at specific start voltages, which could be related to a different degree of decoupling from the substrate.

ARPES (Constant Energy Map)



- The existence of only six Dirac points illustrates the dominance of the Gr-R0° layer.