Electronic supplementary Information

MOCVD of AlN on epitaxial graphene at extreme temperatures

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1) As-grown epitaxial graphene

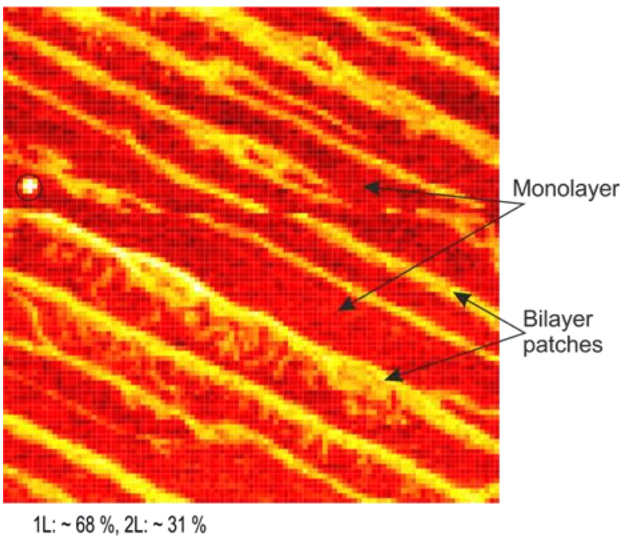

Fig. S1 Reflectance map of as-grown epitaxial graphene collected on a 30 µm × 30 µm sample area obtained using the method described in [1]. The map displays the number of layers, monolayer (red, 68 % of the total area) and bilayer (yellow) covering mainly step edges. The white spot surrounded with black circle is most likely due to a micropipe. The steps are related to the unintentional misorientation of the substrate which occurs during substrate production. The consequence is that formation of bilayer graphene is promoted on the steps resulting in bilayer patches (31% of the total area). Note that the graphene samples grown on the Si-face of SiC substrates always contain a buffer layer, which transforms into an extra graphene layer upon hydrogen intercalation.

2) TEM characterization

Transparent TEM specimens were thinned by conventional Ar ion milling using Technoorg Linda millers and further studied by using a JEOL3010 (LaB6, 300 keV) and a JEOL ARM 200F (200 keV probe corrected) microscopes.

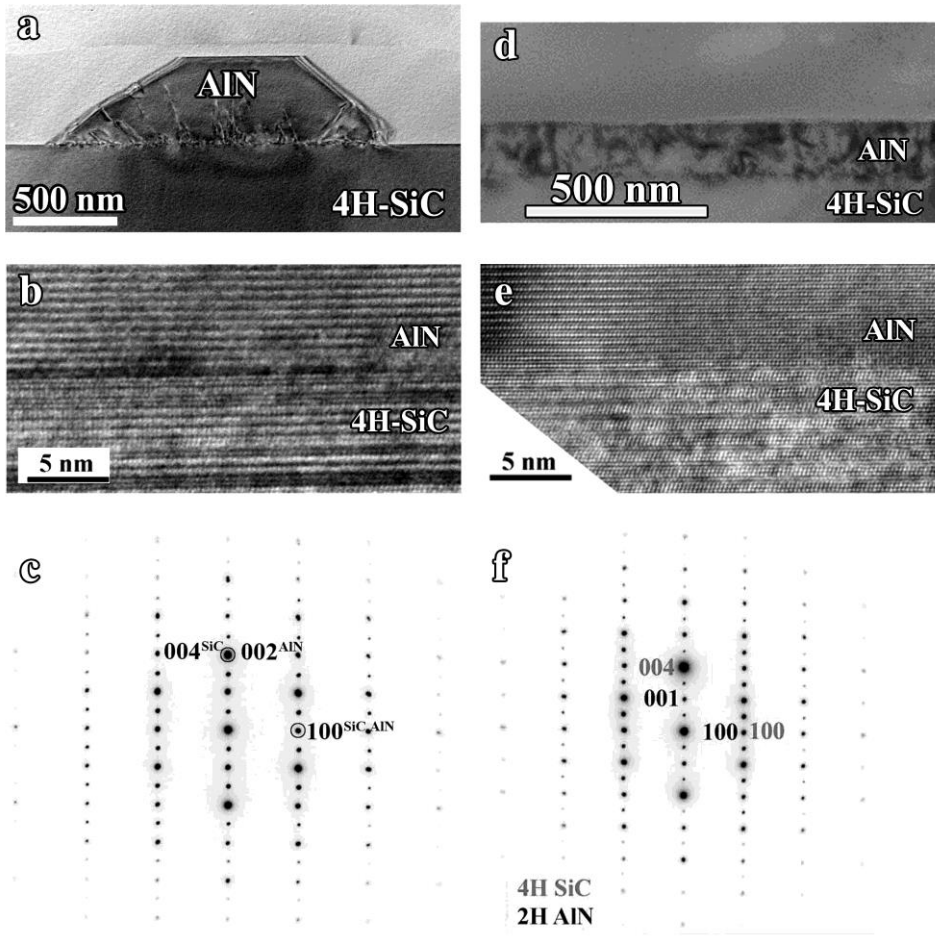

Figure S2. Cross-sectional TEM images of thin AlN crystallites and coalesced areas associated with the MOCVD of AlN on epitaxial graphene at the temperature of 1410°C (a) and 1240°C (d). Graphene undergoes breakage and decomposition at these extreme temperatures (see main text); therefore, it cannot be located at the AlN/SiC interface. Due to the faceted structure of the AlN crystallites, most of the dislocations generated at the interface bend and do not extend to the surface of the AlN. High-resolution TEM images of the AlN/SiC interface at 1410°C (b) and 1240°C (e). The associated selected area electron diffraction patterns are presented in (c) and (f), respectively. Despite of the planar defects occasionally detected in the SiC substrate and shown in (b), there are no such defects in the AlN above the SiC. It is rare that the grown layer quality is better than the quality of the substrate. AlN was grown onto SiC with epitaxy and well-aligned along the indicated crystallographic orientations.

3) Notes on Raman spectroscopy

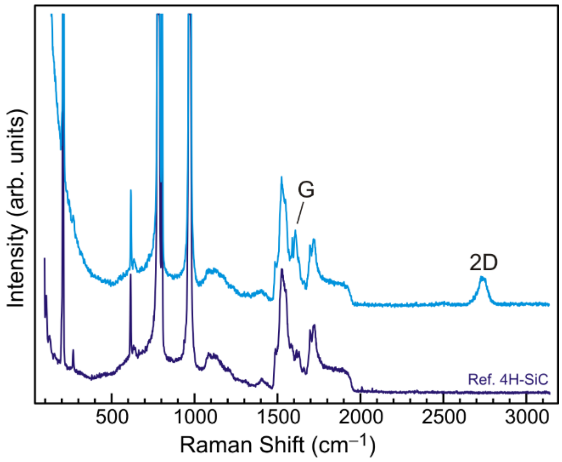

Fig. S3 Raman spectrum of the sample associated with the MOCVD of ultrathin GaN on epitaxial graphene using 532 nm laser excitation. The G and 2D mode of graphene can readily be observed. Reference Raman spectrum of 4H-SiC showing the first and second order Raman contributions of the substrate in the spectrum. By using UV excitation which will be absorbed in the top ultrathin GaN, thus bringing up its contribution and reducing the SiC substrate contribution in the Raman spectrum, the registration of the phonon mode A1(LO) in the ultrathin GaN may be possible [2].

4) Notes on SEM characteriazation

Considering the ultra-small thickness of the GaN, and to achieve a relevant signal contrast, the surface of the sample was imaged using In-lens detector for collection of secondary electrons. The In-lens detector, besides morphology and surface topography with high spatial resolution, can image differences in the work function on the sample with high lateral resolution. Thus, the ultrathin GaN with high work function appears with dark contrast on the SEM image in **Figure S4**.

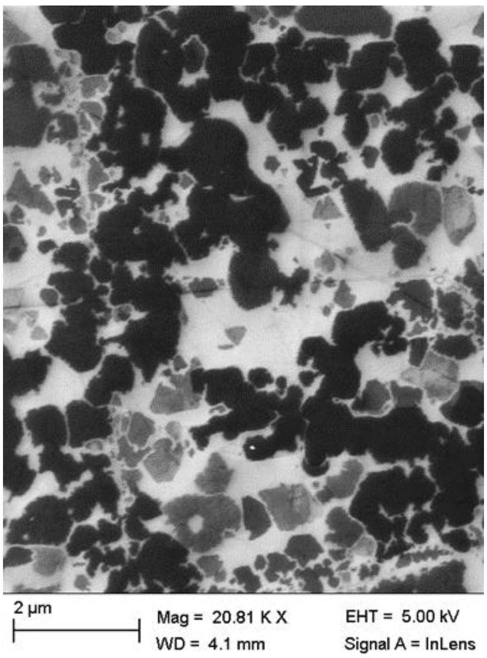

Figure S4. SEM image of ultrathin GaN domains grown on epitaxial graphene on C-face 4H-SiC. The collection of type SE1 electrons was done using In-lens detector. The ultrathin GaN with high work function appears with dark contrast on the image. Similar images, and in correlation with the CAFM images in the main text, have also been observed for ultrathin GaN grown on epitaxial graphene on Si-face 4H-SiC.

References

[1] I. G. Ivanov, et al., *Carbon* 2014, **77**, 492.

[2] Z. Y. Al Balushi, et al., Nature Materials, 2016, 15, 1166; Supplementary information; https://doi.org/10.1038/nmat4742.