Quantitative characterization of epitaxial graphene for the application of quantum Hall resistance standard

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Abstract — **Monolayer graphene grown epitaxially (epitaxial graphene) on silicon carbide (SiC) is the best graphene material for the application of quantum Hall resistance standards (QHRS). The quality of epitaxial graphene is critical for the performance of QHRS with accuracy of ≈ $1{\times}10^{-9}$.**

In this paper, we will characterize the morphology-related defects (multilayer graphene domains and substrate terraces) in epitaxial graphene quantitatively by confocal laser scanning microscopy (CLSM) and Raman spectroscopy. We will correlate the characteristics parameters (size and density of multilayer graphene domain, layer number of graphene, size of terraces) to the performance of graphene QHRS. Understanding the influence of scattering at the morphology-related defects in epitaxial graphene will help further improve the morphology of epitaxial graphene to enhance the performance of graphene QHRS.

Index Terms —**anisotropic scattering, epitaxial graphene, morphology, defects, multilayer graphene domain, quantum Hall effect, silicon carbide, steps, terrace.**

I. INTRODUCTION

Recently, epitaxial graphene has been intensively studied by national metrology institutes (NMIs) for the application of quantum Hall resistance standard (QHRS). With the improvement of the quality of epitaxial graphene, quantum Hall effect (QHE) devices made of epitaxial graphene have been demonstrated to outperform GaAs-based devices, operating with accuracy of ≈ $1{\times}10^{-9}$ at lower magnetic field (5 T or below), higher temperature (up to 5 K) and larger working current (up to hundreds of µA). As monolayer epitaxial graphene can be grown with minimum lattice defects at high temperature on SiC substrate, it contains defects such as the multilayer graphene domains and terraces on the SiC surface. Further enhancement of the graphene-based QHRS performance requires understanding of the scattering at these defects.

Epitaxial graphene used for QHRS is grown on the si-terminated surface of SiC substrate by a high temperature annealing process. At high temperature, initial SiC sublimation leads to small steps on the SiC surface, which transform into terraces through step bunching as temperature increases. Carbon atoms diffuse and nucleate on the SiC surface to form graphene. Thicker graphitization usually occurs along the edge of the terraces due to anisotropic diffusion of carbon atoms. Breakdown of quantum Hall effect in epitaxial graphene can be caused by anisotropic scattering at the terrace edges and the multilayer graphene domains. [1, 2].

Between the terrace morphology and the multilayer graphene domain, it is not clear which is the dominant defects to limit the performance of the QHE devices. In this paper, we will investigate the correlation between the performance of graphene QHE devices and the characteristics of these two common defects in epitaxial graphene.

II. SAMPLE FABRICATION AND CHARACTERIZATION

We grow epitaxial graphene on SiC substrate (7.6 mm to 23 mm size) at high temperatures up to 1950 °C in an Ar background gas. The sublimation rate of SiC is suppressed by placing the Si-side surface of SiC substrate toward a graphite plate [3,4]. Terraces with width of a couple of µm to above 10µm will form on the Si-side surface (Fig. 1a), Multilayer graphene stripes (the stripes with bright contrast in Fig. 1a) extend along the edge of the terraces to above 20 µm, while the monolayer graphene on the terraces is almost bilayer-free.

Combined with a polymer-assisted sublimation method developed at Physikalisch-Technische Bundesanstalt (PTB) [5,6], we can grow nearly bilayer-free epitaxial graphene of about 100 µm size (Figure 1b), because the PTB method can effectively suppress the step bunching and therefore reduce the multilayer graphene domain size. The width of the terraces in Fig. 1b is below 1 micrometer, which can not be detected by confocal laser scanning microscope (CLSM). The yield of bilayer-free samples is very low due to the difficulties to control the local sublimation rate. Usually, multilayer graphene domains of various sizes are distributed randomly across the SiC substrate (Figs. 1c and 1d).

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We fabricate Hall bars with width of 100 µm to 400 µm, where the graphene in the Hall bar region is either free of bilayer graphene or has evenly distributed multilayer graphene domains. Our clean fabrication process [7] prevents contamination from photoresist used for lithography steps. We characterize the defects of graphene in the Hall bar region quantitatively by atomic force microscope (AFM), CLSM [8] and Raman spectroscopy and correlate the characteristic parameters (graphene layer number, multilayer graphene domain size and density etc.) to the performance of the QHE devices. The graphene in the Hall bar region is open to ambient environment so that its carrier density can be tuned by exposing the sample to p-doping gas molecules (such as air or nitric acid vapor) and subsequent vacuum annealing.

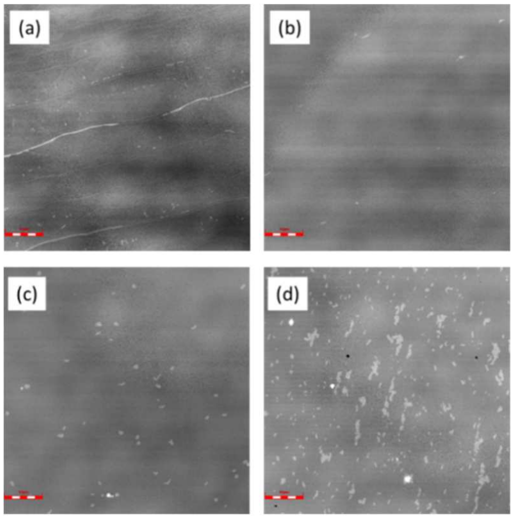

Figure 1. Reflective intensity images of epitaxial graphene on SiC substrate obtained by CLSM. Features (dots, patches, stripes) with bright contrast are multilayer graphene domains. The scale bar is 10 µm. (a) Epitaxial graphene with wide (2 µm to 10 µm) terraces. (b) Nearly bilayer-free monolayer epitaxial graphene. (c) Monolayer graphene region with multilayer domain coverage rate of 0.63 %. (d) Monolayer graphene region with multilayer domain coverage rate of 5.63 %.

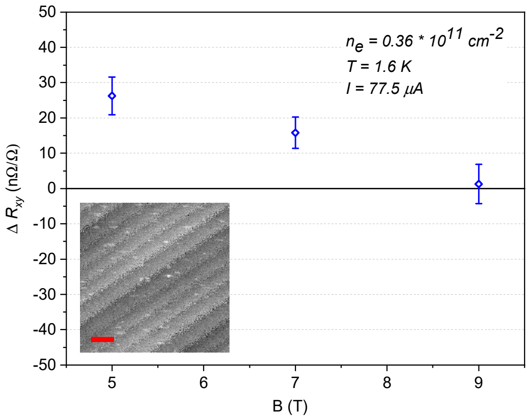

Figure 2. Precision characterization of a graphene Hall bar with multilayer graphene domains. Graphene carrier density $n \approx 0.36 \times 10^{11}$ cm^{-2}. The error bar represents type A uncertainty. Inset: Reflective intensity images of epitaxial graphene in the Hall bar region.

Figure 2 shows the quantum Hall resistance deviation of a 400 µm wide Hall bar from nominal value $\Delta R_{xy} = (R_H - R_K/2)/(R_K/2)$, where R_H is the measured Hall resistance at $\nu = 2$ plateau and R_K is the van Klitzing constant. At 1.6 K and 9 T and with working current of 77.5 µA, the Hall resistance of this device agrees with $R_K/2$ to $\approx 1 \times 10^{-9}$. The multilayer graphene domain coverage rate in the Hall bar area for this sample is ≈ 1.67 % (inset of Fig. 2). We will report the QHE measurement of a number of graphene devices and the correlation between their performance and the epitaxial graphene quality.

III. CONCLUSION

We are investigating the influence of morphology-related defects in epitaxial graphene, including the substrate terrace and multilayer graphene domain, on the performance of graphene QHRS. Quantum Hall effect in epitaxial graphene may break down due to the anisotropic scattering at the terrace edges and the multilayer graphene domains. We will analyze the characteristic parameters of these defects and correlate them with the quantum Hall effect measurements. This work will help us further improve the quality of epitaxial graphene to enhance the QHRS performance.

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REFERENCES

[1] T. Schumann, *et al.*, “Anisotropic quantum Hall effect in epitaxial graphene on stepped SiC surfaces” *Phys. Rev. B* **85**, 235402, June 2012.
[2] M. K. Yakes, *et al.*, “Conductance anisotropy in epitaxial graphene sheets generated by substrate interactions,” *Nano Lett.* **10**, 1559 – 1562, April 2010.
[3] M.A. Real, *et al.*, “Graphene epitaxial growth on SiC(0001) for resistance standards,” *IEEE Trans. Instrum. Meas.* **62,** 1454-1460. June 2013.
[4] Y. Yang, *et al.*, “Epitaxial graphene homogeneous and quantum Hall effect in millimeter-scale devices,” *Carbon.* **115,** 229, 2017.
[5] M. Kruskopf, *et al.*, “Comeback of epitaxial graphene for electronics: large-area growth of bilayer-free graphene on SiC,” *2D Mater.* **3** 041002, 2016.
[6] M. Kruskopf, *et al.*, “A morphology study on the epitaxial growth of graphene and its buffer layer,” *Thin Solid Films* **659**, 7-15, 2018.
[7] Y. Yang, *et al.*, “Low carrier density epitaxial graphene devices on SiC,” *Small* **11**, 90, 2015.
[8] V. Panchal, *et al.*, “Confocal scanning microscopy for rapid optical characterization of graphene,” *Commun. Phys.* **1**, 83, 2018.