Synthesis of millimeter-size monolayer epitaxial graphene with uniform strain and record magnetotransport

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Abstract

We present a novel synthesis process that produces at least 99% coverage of high-quality, monolayer epitaxial graphene (EG) with uniform strain over a 27 mm^2 area of SiC (0001) substrate. This method is demonstrated to be repeatable and scalable, and provides the size and quality of graphene needed for future electronic applications.

Today, most EG is grown leveraging two advances in the field. The first was in 2009 when Emtsev and his colleagues showed that EG quality is significantly improved by annealing SiC under an argon atmosphere[1]. The second uses confinement controlled sublimation to approach the desired equilibrium growth conditions[2]. Still, production of monolayer EG free of bilayer inclusions remains very challenging, mainly due to the extended micron-wide terraces on SiC(0001) where bilayers grow along the step edges.

In this work, we approach this problem using an innovative face-to-graphite (FTG) geometric constraint[3] at high temperatures (1900 °C) after a H_2-treatment of the substrate. The H_2 treated samples develop unique terrace morphology which inhibits bilayer formation. Specifically, Raman mapping shows at least 99 % coverage of monolayer graphene with unprecedented strain uniformity determined from over 1100 data points collected from three large regions on a 27 mm^2 device.

We also demonstrate for the first time a high mobility of 5800 cm^2/Vs in a very large device (27 mm^2), more than 3000 times larger than devices with similar performance[4]. Moreover, metrological accuracy at a record current level of 0.72 mA for quantum Hall resistance (QHR) is reported in the same device. This higher measurement current provides unprecedented sensitivity for graphene-based QHR metrology, allowing direct adoption in industrial settings.

Our method will directly impact the fundamental research community by enabling the simple and effective production of high-quality, monolayer graphene and the industrial community by providing an exceptional electronic material at the millimeter-scale and beyond.

References

[1] K. V. Emtsev, A. Bostwick,K. Horn, J. Jobst, G. L. Kellogg, L. Ley, J. L. McChesney, T. Ohta, S. A. Reshanov, J. Röhrl, E. Rotenberg, A. K. Schmid, D. Waldmann, H. B. Weber, T. Seyller, Nature Mater., **8** (2009), 203-207.

[2] W. A. de Heer, C. Berger, M. Ruan, M. Sprinkle, X. Li, Y. Hu, B. Zhang, J. Hankinson, E. H. Conrad, Proc. Natl. Acad. Sci. USA. 108 **(2011)**, 16900-16905.

[3] M. A. Real, E. A. Lass, F.-H. Liu, T. Shen, G. R. Jones, J. A. Soons, D. B. Newell, A. V. Davydov, R. E. Elmquist, IEEE Trans. Instrum. Meas. **62** (2013), 1454-1460.

[4] E. Pallecchi, F. Lafont, V. Cavaliere, F. Schopfer, D. Mailly, W. Poirier, A. Ouerghi, Sci. Rep. **4** (2014), 4558.

Figures

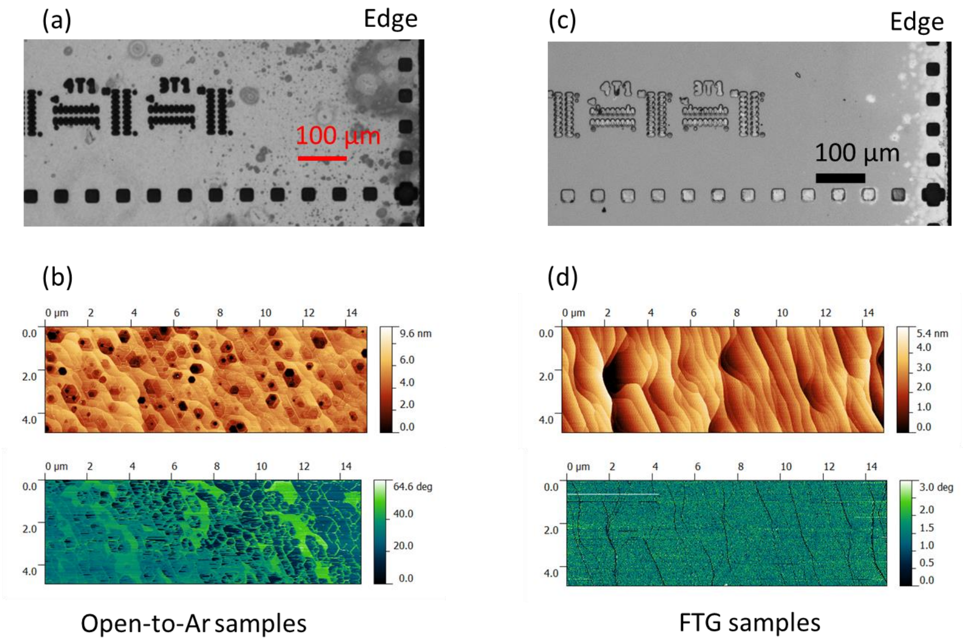

Figure 1. (a-b) Transmission optical image (a) and AFM images (b) of epitaxial graphene on SiC annealed with Si-face open to Ar, without H_2-treatment. (c-d) Transmission optical image (c) and AFM images (d) of epitaxial graphene on SiC annealed with FTG method, with H_2-treatment. The optical images are taken at the edge of the samples, while the AFM images are taken in the center of the samples.

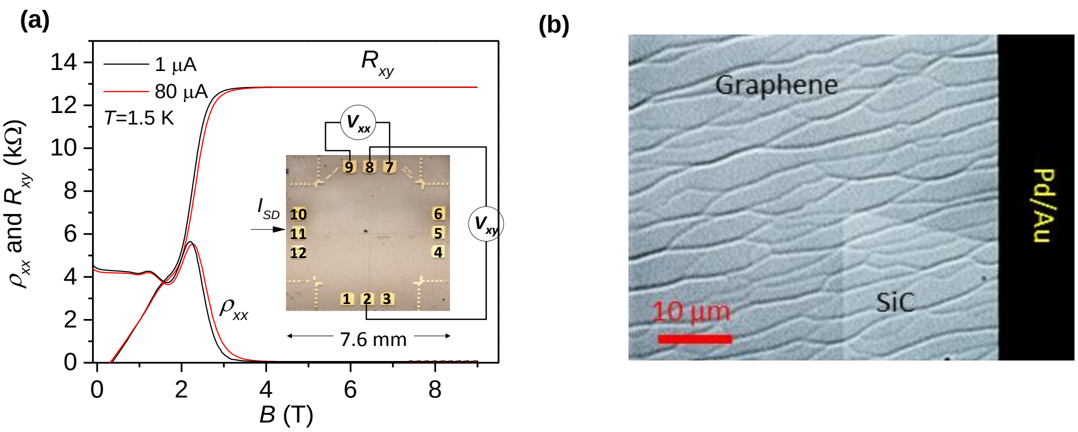

Figure 2. (a) Magnetotransport measurements of a 27 mm^2 octagonal sample at 1.5 K for AC currents of 1 µA and 80 µA. Inset shows a photograph of the device and the AC transport measurement configuration for longitudinal voltage V_{xx} and for Hall voltage V_{xy}. (b) Optical image of a contact region of the device in (a), showing a Pd/Au contact and the boundary between monolayer graphene and bare SiC.