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V.S. Prudkovskiy, K.P. Katin, M.M. Maslov, Pascal Puech, R. Yakimova, et al.. High robustness of epitaxial 4H-SiC graphene to oxidation processes. *Journal of Physics: Conference Series*, 2018, 1124, pp.081020. 10.1088/1742-6596/1124/8/081020 . hal-02080621

HAL Id: hal-02080621

<https://hal.science/hal-02080621>

Submitted on 18 Jun 2019

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To cite this article: V.S. Prudkovskiy *et al* 2018 *J. Phys.: Conf. Ser.* **1124** 081020

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High robustness of epitaxial 4H-SiC graphene to oxidation processes

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Abstract. We present an experimental prove of high robustness of epitaxial 4H-SiC graphene to oxidation processes. During a post-fabrication cleaning procedure we noticed that epitaxial graphene is extremely stable to ozone treatment. We analyse graphene properties using both electron transport measurements and numerical calculations. We ascribe this effect to the substrate topography, which significantly affects the graphene stability under UV/ozone treatment.

1. Introduction

Great attention has been attributed to graphene as a perceptive material for new generation of electronic devices [1, 2]. Main obstacle of utilizing graphene for nanoelectronic is its significant sensitivity to contamination. Different polymers routinely used for nanofabrication leave residual traces on graphene. Majority of the research devoted to graphene-based electronics highlights the necessity of a cleaning procedure. The most efficient graphene decontamination procedure in our days is ozone cleaning [3], which is aggressive enough to clean graphene surface without destroying graphene crystal lattice for a proper chosen timescale of treatment.

2. Results

The experiment was carried out on graphene grown by sublimation process on 4H-SiC Si terminated substrate. Devices were fabricated in four-probe Van der Pauw configuration (Fig.1).



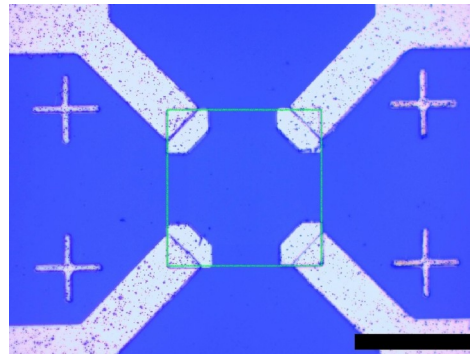


Figure 1. Optical image of a typical device, the scale bar is 50 μm . Graphene area is outlined by green line.

After fabrication all measured devices demonstrate electron doping. We associate this doping with contamination by polymers' radicals remaining on the graphene surface after fabrication process. The first two minutes of ozonation lead to a significant rise of the mobility, followed by the mobility saturation region that corresponds to 2 to 4 minutes of ozone exposure (Fig. 2). A large mobility drop after fifth minute of ozone exposure is observed. The increase of the mobility is most noticeable for the most contaminated devices. Initially low contaminated devices samples conserve their high initial mobility values and remain almost unaffected by the ozone treatment up to the fifth minute of ozonation, after which all the samples show a drop of the mobility to a range around $\sim 250 \text{ cm}^2/(\text{V}\cdot\text{s})$.

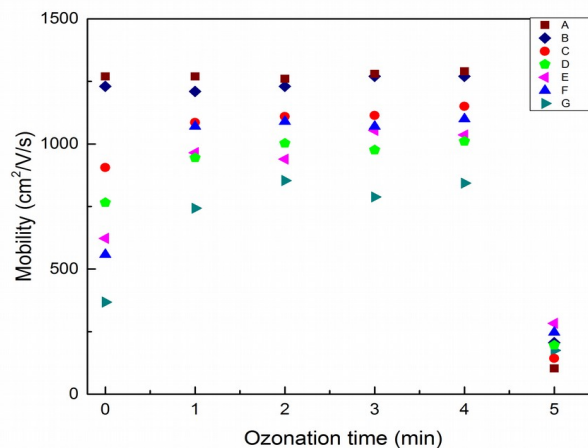


Figure 2. Graphene charge-carrier mobility as a function of ozonation time for seven devices with different doping level. Devices are named from A to G.

In order to understand this behavior we ask reader to focus on dependence of the carrier density on ozonation time (Fig. 3). The initial devices are electron doped with charge-carrier density in the range of $-1.5 \cdot 10^{12}$ to $-0.5 \cdot 10^{12} \text{ cm}^{-2}$. The carrier density of all devices after two minutes of ozonation reaches level of $-4 \cdot 10^{11} \text{ cm}^{-2}$.

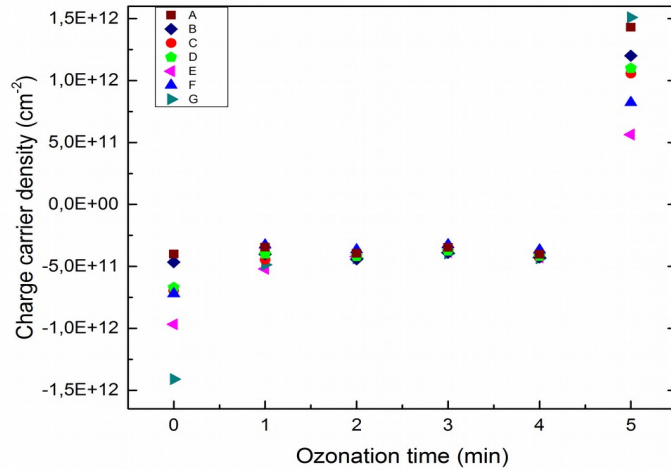


Figure 3. Graphene charge-carrier density as a function of ozonation time for seven devices with different doping level.

Decrease of electron concentration coincides with the mobility rise (Fig. 2) and corresponds to the phase of residual polymer decomposition. In following, we observe a saturation of charge density at this level. After the fifth ozonation minute the density sharply rises up to $1.5 \cdot 10^{12} \text{ cm}^{-2}$ along with change of charge carriers type from electrons to holes. This event coincides with the drop of the mobility (Fig. 2). The whole process of mobility and density evolution on ozonation time could be seen in Figure 4.

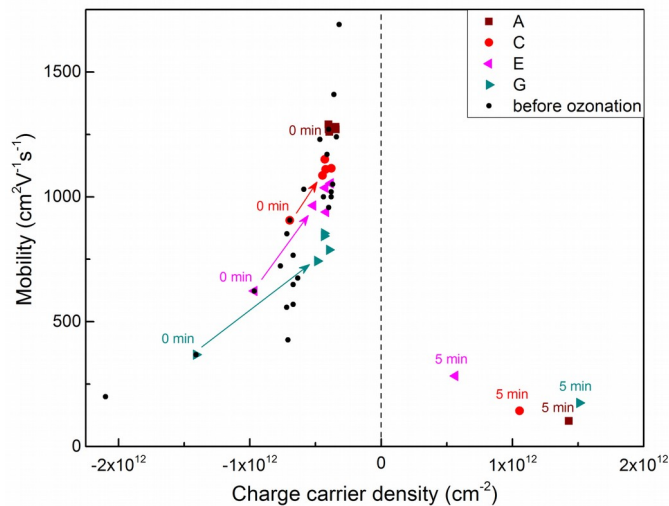


Figure 4. Graphene charge-carrier mobility μ versus density n . Black dots represent the $\mu(n)$ distribution of the all measured devices before ozonation. Colored markers correspond to devices: A (dark-red square), C (red circle), E (magenta triangle) and G (green triangle) after different ozonation time, labeled by corresponding color: “0 min” labels mark the initial states, arrows point toward the states after 1-4 min of ozonation, “5 min” labels mark the states after fifth minute of ozonation.

Thus the samples sustain up to 4 minutes of ozone cleaning procedure, which allows to increase the mobility and decrease the doping of contaminated samples. Recent experiments on exfoliated and

CVD graphene [4, 5] show that even one minute of ozonation leads to significant increase of the intensity of the D band in Raman spectra, originated from epoxy groups formation. This high SiC graphene stability is associated with the extremely flatness of epitaxial graphene contrary to exfoliated or transferred CVD graphene.

Our calculations revealed that surface roughness of graphene sheet could change the energy gain from epoxy group adsorption on a few tens of kilocalories per mole (Figure 5).

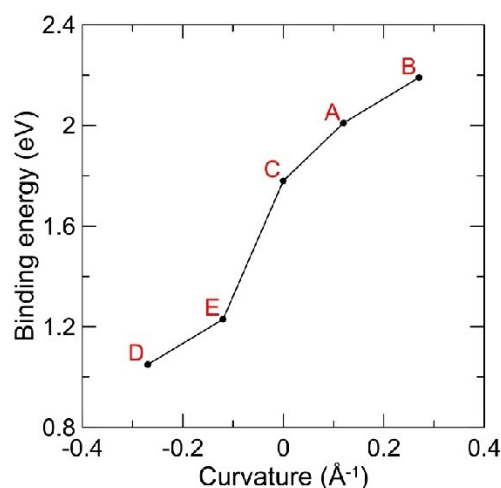


Figure 5. Dependence of the binding energy of the epoxy group on curvature of a graphene surface.

Such variations of energy are sufficient to do the two-epoxy groups adsorption more or less energetically favourable in comparison with the O₂ molecule formation depending on local sheet curvature. Thus, surface roughness can significantly affect the graphene stability under UV/Ozone treatment.

3. Conclusion

We found that graphene prepared by sublimation on 4H-SiC Si terminated substrate is extremely stable to radical oxygen atoms. Our measurements reveal the absence of defects in graphene lattice after four minutes of ozonation, while exfoliated and CVD graphene cannot sustain even a minute [4, 5]. This high stability is associated with an extremely flatness of epitaxial graphene contrary to exfoliated or transferred CVD graphene. Our calculations reveal that surface roughness can change the energy gain from epoxy group adsorption by a few tenths of electron volts.

Acknowledgments

The reported study was funded by RFBR, according to the research project No. 16-32-60081 mol_a_dk.

References

- [1] C. Berger, Z. Song, T. Li, X. Li, A.Y. Ogbazghi, R. Feng, Z. Dai, A.N. Marchenkov, E.H. Conrad, P.N. First, W.A. de Heer 2004 *J. Phys. Chem. B* **108** 19912
- [2] J. Baringhaus, M. Ruan, F. Edler, A. Tejada, M. Sicot, A. Taleb-Ibrahimi, A. Li, Z. Jiang E.H. Conrad, C. Berger, C. Tegenkamp, W.A. de Heer 2014 *Nature* **506** 349
- [3] V.S. Prudkovskiy, K.P. Katin, M.M. Maslov, P. Puech, R. Yakimova, G. Deligeorgis 2016 *Carbon* **109** 221
- [4] Y. Mulyana, M. Uenuma, Y. Ishikawa, Y. Uraoka 2014 *J. Phys. Chem. C* 2014 **118** 27372
- [5] M.G. Chung, D.H. Kim, H.M. Lee, T. Kim, J.H. Choi, D.K. Seo, J. Yoo, S. Hong, T.J. Kang, Y.H. Kim 2012 *Sensors and Actuators B* **166** 172