Grain Boundary Resistivity in Polycrystalline Graphene

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Abstract

In recent years, graphene has emerged as a favorable material for a wide range of applications [1]. While single-crystal graphene would be ideal, the most promising approach for the mass production of graphene is chemical vapor deposition (CVD), which results in a material that is polycrystalline [2,3]. This polycrystallinity arises due to the nucleation of growth sites at random positions and orientations during the CVD process. In order to accommodate the lattice mismatch between misoriented grains, the grain boundaries in polycrystalline graphene consist of a variety of non-hexagonal carbon rings, which can serve as a source of carrier scattering [4,5]. Indeed, several experimental works have demonstrated that grain boundaries add an extra resistance compared to single-grain samples [6-9]. In addition, a variety of studies have demonstrated the high chemical reactivity of the grain boundaries, compared to pristine graphene [6,10]. This has led to the possibility of using polycrystalline graphene as efficient chemical sensors [11]. Thus, in order to understand the large-scale transport properties of polycrystalline graphene, it is important to understand charge transport through the grain boundaries, and how this is altered by chemical functionalization.

In this work, we use numerical simulations to examine the role that grain boundaries play in charge transport through polycrystalline graphene. We find that grain boundaries increase the sheet resistance of graphene samples, and with a simple scaling law we extract the intrinsic grain boundary resistivity. The calculated grain boundary resistivity is 1-2 orders of magnitude smaller than what is obtained from most measurements, suggesting that scattering due to the non-hexagonal structure of the grain boundaries is relatively small, and that another mechanism must be responsible for most of the experimentally-measured grain boundary resistivity. We examine this by progressively adding chemical adsorbates to the grain boundaries, as depicted in Fig. 1. We find that functionalization can tune the grain boundary resistivity by more than one order of magnitude, as shown in Fig. 2, bringing the simulations within the range of experimental measurements [6-9,12,13]. These results have strong implications for CVD-grown graphene, as they indicate that chemical functionalization can play a strong role in the electrical properties of this material. We also present recent experimental measurements that demonstrate the impact of chemical functionalization on an individual grain boundary.

References

[1] K.S. Novoselov *et al.*, Nature, **490** (2012), 192-200.
[2] P. Y. Huang *et al.*, Nature **469** (2011), 389-392.
[3] K. Kim *et al.*, ACS Nano **5** (2011), 2142-2146.
[4] O.V. Yazyev and S.G. Louie, Nat. Mater. **9** (2010), 806-809.
[5] D. van Tuan *et al.*, Nano Lett. **13** (2013), 1730-1735.
[6] D.L. Duong *et al.*, Nature **11** (2012), 235-240.
[7] Q. Yu *et al.*, Nat. Mater. **10** (2011), 443-449.
[8] Jauregi *et al.*, Solid State Commun. **151** (2011), 1100-1104.
[9] A.W. Tsen *et al.*, Science **336** (2012), 1143-1146.
[10] P. Nemes-Incze *et al.*, Appl. Phys. Lett. **99** (2011), 023104.
[11] A. Salehi-Khojin *et al.*, Adv. Mater. **24** (2012), 53-57.
[12] Clark *et al.*, ACS Nano **7** (2013), 7956-7966.
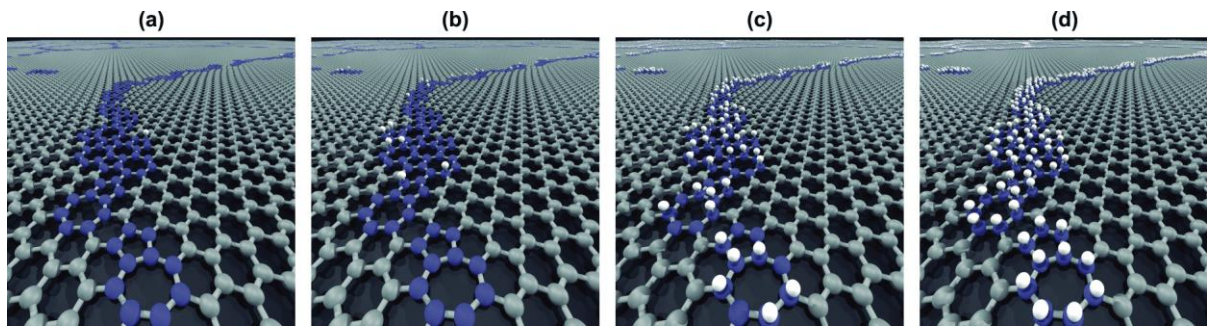[13] Vlassiouk *et al.*, Nanotechnology **22** (2011), 275716.

Figures

Fig. 1. Schematic representation of H adsorbates on a graphene grain boundary. Gray atoms belong to the graphene grains, blue atoms belong to the grain boundaries, and white atoms are the H adsorbates. Adsorbate concentrations are (a) 1%, (b) 10%, (c) 50%, and (d) 100%.
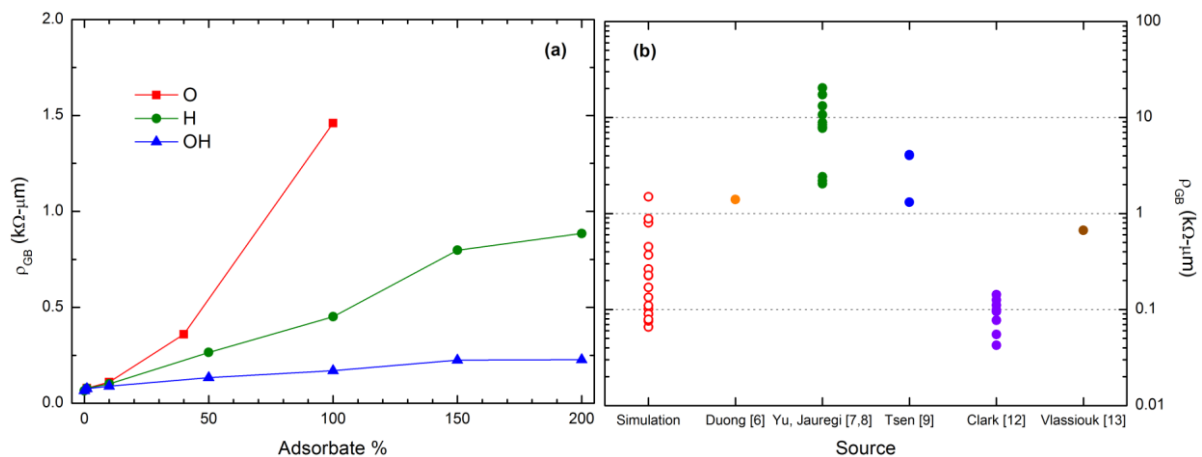

Fig. 2. Effect of chemical functionalization on grain boundary resistivity. Panel (a) shows the grain boundary resistivity as a function of adsorbate concentration. Red squares are for epoxide, green circles are for hydrogen, and blue triangles are for hydroxyl adsorbates. Panel (b) shows the grain boundary resistivity from a variety of sources. The first set of data (red open circles, far left) gives our numerical simulations, and the rest are the experimental measurements of other groups.