Polycrystalline graphene as a raw material for gas sensors

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The design of graphene-based gas sensors involves three fundamental components: the sensitive graphene-based material, the device design and the selectivity performance. In the sensitive material context, graphene synthesized by mechanical exfoliation has been proven as a suitable sensor material after achieving gas detection at concentrations of one part per billion [1]; however, mechanical exfoliation is not scalable to mass-production of devices. In contrast, highly-damaged graphene like reduced graphene oxide can be synthesized at low cost and large scale, but the sensitivity is diminished to less than one part per million [2]. The most promising way to synthesize large-area high-quality graphene is chemical vapor deposition (CVD), which usually results in a polycrystalline material. This material consists of a variety of misoriented graphene grains with grain boundary interfaces. These grain boundaries are made up of non-hexagonal carbon rings with high chemical reactivity, which can increase the sensitivity up to one part per trillion [3]. In addition, ozone-treated polycrystalline graphene demonstrated significant enhancement of sensing performance compared with the untreated material [4]. Nevertheless, despite the promise of polycrystalline graphene, the sensing mechanisms of this material are not fully understood.

In this work, we perform numerical simulations of quantum transport in oxygen-doped polycrystalline graphene samples. To carry out the transport calculations we use a real-space order-N quantum wave packet approach to compute the Kubo-Greenwood electrical conductivity, which we then relate to the mobility as a function of carrier concentration in order to match with experimental measurements [5,6]. It has been shown that adsorbed oxygen in graphene brings about epoxide functionalization, triggering strong intervalley scattering and a metal-insulator transition [7]. Additionally, in polycrystalline graphene the grain boundary sites are highly reactive, making those sites likely for epoxide functionalization. Our calculations suggest that this functionalization has a lower impact on the mobility than functionalization within the pristine graphene grains. Thus, the electrical transport behavior of functionalized polycrystalline graphene depends on the spatial extension and distribution of the grain boundaries. This behavior serves as an important design consideration for the optimization of gas sensors based on CVD-grown graphene.

References

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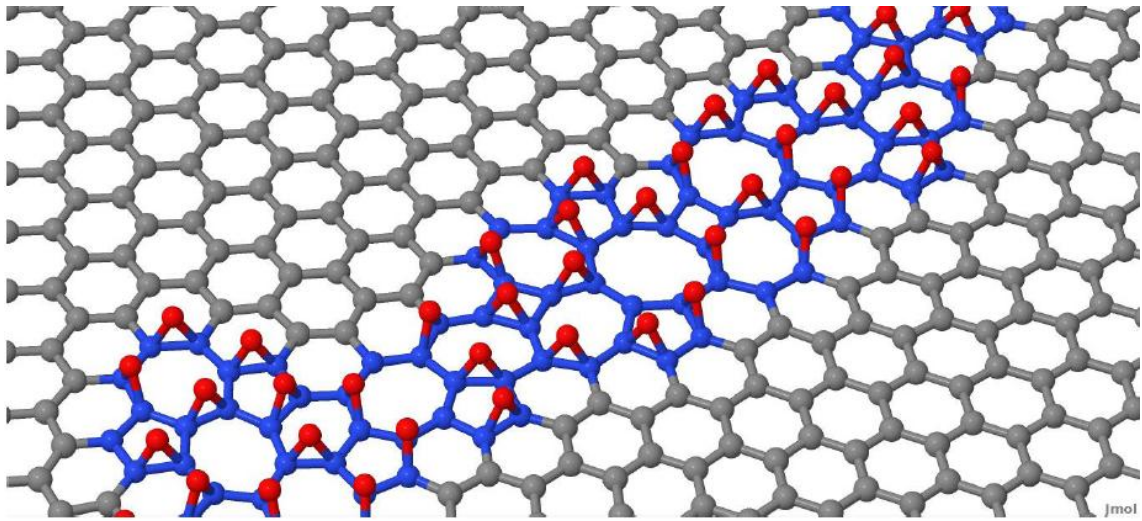
Figure 1. Polycrystalline graphene sample with epoxide functionalization. Carbon atoms are schematically colored in gray, the carbon atoms in the grain boundary are colored in blue, and the Oxygen atoms are colored in red.