Scaling Properties of Charge Transport in Polycrystalline Graphene

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Abstract

Polycrystalline graphene is a patchwork of coalescing graphene grains of varying lattice orientations and size, resulting from the chemical vapor deposition (CVD)-growth at random nucleation sites on metallic substrates [1, 2, 3, 4, 5]. The morphology of polycrystalline graphene has become an important topic given its fundamental role in limiting the mobilities compared to mechanically exfoliated graphene monolayers [6]. The relationship between polycrystalline morphologies (grain sizes and grain boundary (GB) structures) and resulting physical properties is also a central aspect of the graphene roadmap in view of applications such as flexible electronics and high-frequency or spintronics devices [7]. Here we report new insights to the current understanding of charge transport in polycrystalline geometries. We first created realistic models of large CVD-grown graphene samples. Then, we used an efficient computational approach to compute charge mobilities within these systems as a function of the average grain size and the coalescence quality between the grains. Our results, which agree with recent experiments [8], reveal a remarkably simple scaling law for the mean free path and conductivity, correlated to atomic-scale charge density fluctuations (electron-hole puddles) along GBs. These findings establish quantitative foundations of transport features in polycrystalline graphene, thereby paving the way for improvements in graphene-based applications.

References

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Figures

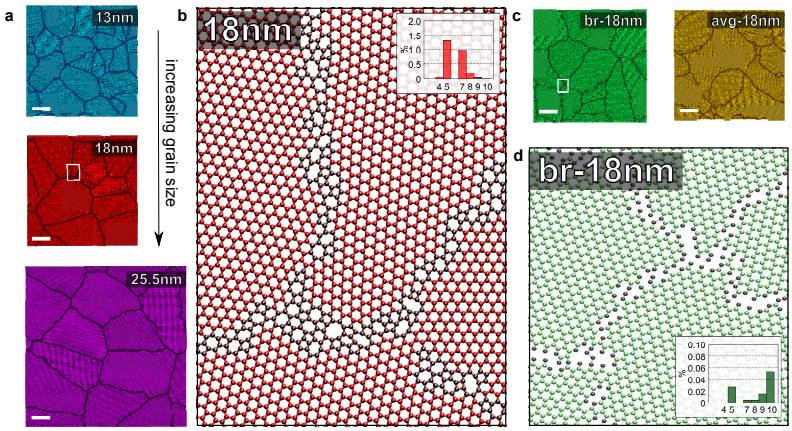

Figure 1: **Polycrystalline graphene samples.**

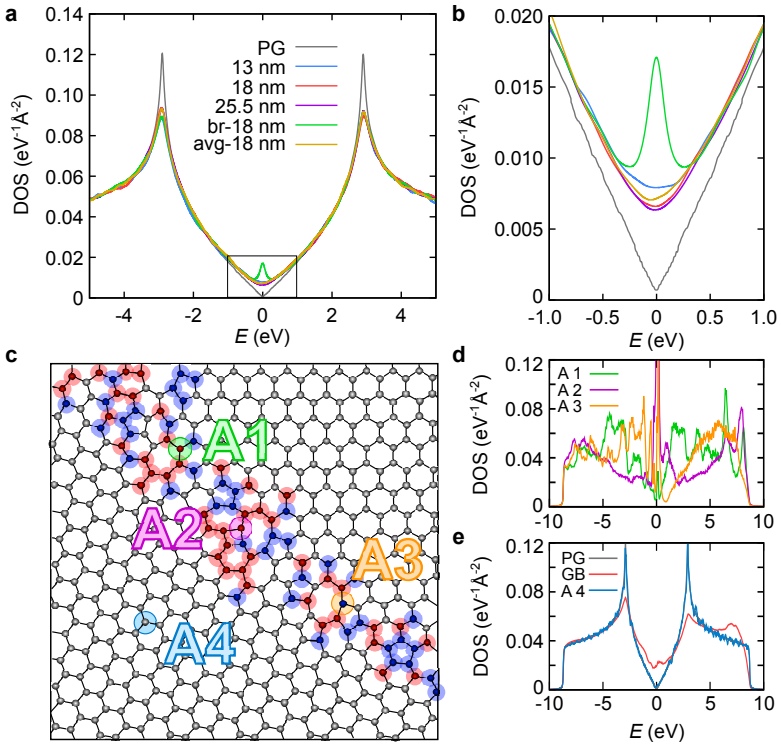

Figure 2: **Density of states (DOS)**. **a**. DOS for pristine graphene (PG) and the structures presented in Fig. 1. **b.** Higher magnification of the DOS close to the charge neutrality point ($E = 0$, area marked with a rectangle in panel **a**). **c**. Atomic structure of one of the boundaries in sample "18 nm", showing the electron-hole puddles at GB sites that develop due to local variations in the charge density δ_i: local electron doping ($\delta_i < -1 \times 10^{-4} e$/atom) is shown in blue and local hole doping ($\delta_i > 1 \times 10^{-4} e$/atom) in red. **d**. Local DOS for atoms A1, A2 and A3 marked in panel **c**. **e**. Local DOS for atom A4 marked in panel **c** as compared to the average DOS for pristine graphene (PG) and average LDOS for all atoms at GBs in the same sample (GB).

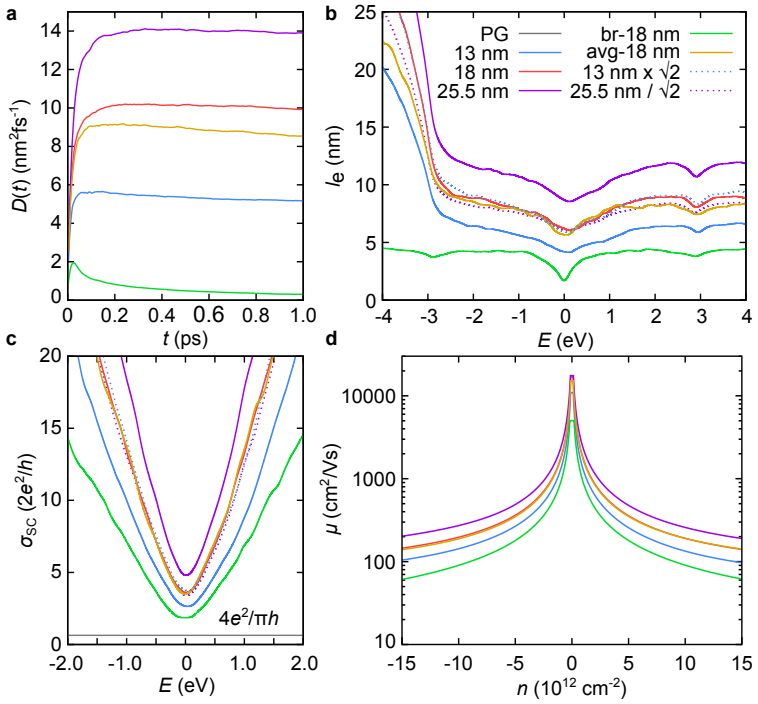

Figure 3: **Transport properties**. **a**. Diffusion coefficient ($D(t)$) for the samples presented in Fig. 1. **b**. Mean free path $\ell_e(E)$ for equivalent structures with scaled $\ell_e(E)$ for samples with $\langle d \rangle \approx 13$ nm and $\langle d \rangle \approx 25.5$ nm, showing the scaling law. **c**. Semi-classical conductivity ($\sigma_{sc}(E)$) for all samples and as scaled for the same cases as above. **d**. Charge mobility ($\mu(E) = \sigma_{sc}(E)/en(E)$) as a function of the carrier density $n(E)$ in each of the samples ($n(E) = 1/S \int_0^E \rho(E)dE$, S being a normalization factor).