Charge Transport in van der Waals Thin Films, the case of Reduced Graphene Oxide from single flake to thin film

Alessandro Kovtun, Andrea Candidi, Alex Boschi, Andrea Liscio, Vincenzo Palermo
1 Istituto per la Sintesi Organica e la Fotoreattività (ISOF-CNR), via Gobetti 101, 40129 Bologna, Italy
alessandro.kovtun@isof.cnr.it

Large area van der Waals (vdW) thin films are assembled materials consisting of a network of randomly stacked nanosheets. The multiscale structure and the two dimensional (2D) nature of the building block mean that interfaces naturally play a crucial role in the charge transport of such thin films. While single or few stacked nanosheets (i.e., vdW heterostructures) have been the subject of intensive works, little is known about how charges travel through multilayered, more disordered networks. Here, we report a comprehensive study of a prototypical system given by networks of randomly stacked reduced graphene oxide 2D nanosheets, whose chemical and geometrical properties can be controlled independently, permitting to explore networks ranging from a single nanosheet to some billions forming thin films, with room temperature resistivity spanning from $10^{-5}$ to $10^{-1}$ Ω m.

Most studies give an ambiguous interpretation of charge transport phenomena by qualitative plot of resistance vs temperature, while a univocal interpretation of the charge transport phenomenon is still object of debate: Efros-Shklovskii variable-range hopping (ES-VRH) or 2D Mott variable-range hopping (2D-VRH). Based on analysis of the reduced activation energy $W(T)$, we systematically observed a clear transition between two different regimes: Efros–Shklovskii variable-range hopping (ES-VRH) below a critical temperature $T^*$ and power law behavior above.

First, we demonstrate that the two regimes are strongly correlated with each other, both depending on the charge localization length $\xi$, calculated by the ES-VRH model, which corresponds to the characteristic size of overlapping sp² domains belonging to different nanosheets. Thus, we propose a microscopic model describing the probability that charges circumvent the hopping barriers increases with film thickness, with a corresponding increase in the effective delocalization of the electronic states up to the micron scale.

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References


Figures

Figure 1: Resistivity vs Temperature of single sheet rGO (a) and thin film (b). Charge transport scheme(c).