Multiscale Charge Transport in van der Waals Thin Films: Reduced Graphene Oxide as a Case Study

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Large area van der Waals (vdW) thin films are assembled materials consisting of a network of randomly stacked nanosheets. The multi-scale structure and the two-dimensional nature of the building block mean that interfaces naturally play a crucial role in the charge transport of such thin films.[1] While single or few stacked nanosheets (i.e. vdW heterostructures) have been the subject of intensive works,[2] 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,[3] permitting to explore percolated networks ranging from a single nanosheet to some billions with room temperature resistivity spanning from 10^{-5} to 10^{-1} $\Omega\cdot$m. We systematically observe a clear transition between two different regimes at a critical temperature T*: Efros-Shklovskii variable range hopping (ES-VRH) below T* and power law (PL) behavior above. Firstly, we demonstrate that the two regimes are strongly correlated with each other, both depending on the charge localization length ξ, calculated by ES-VRH model,[4] which corresponds to the characteristic size of overlapping sp2 domains belonging to different nanosheets. Thus, we propose a microscopic model describing the charge transport as a geometrical phase transition, given by the metal-insulator transition associated with the percolation of quasi-1D nanofillers with length ξ,[5,6] showing that the charge transport behaviour of the networks does neither depend on geometry nor on the defects of the nanosheets, ultimately suggesting a generalized description on vdW and disordered thin films.[7]

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FIGURE

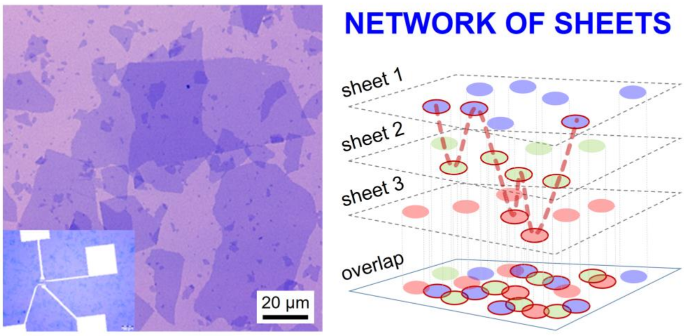

Figure: (Left) Optical image of a sparse network composed by few 2D RGO sheets in partial contact. Inset: Optical image showing the metal pad geometry used to measure CT in such samples; inset size: 1.7×1.2 mm^2.(Right) Three-layer RGO thin film. Each plane is represented as a patchwork of isolated sp2 domains (circles) separated by domain border defects. For sake of clarity, we distinguish each layer with a different colours. Dashed red line corresponds to a random path connecting overlapped disks.