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Investigation of Doping Mechanism in Graphene Decorated with Copper Nanoparticles

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Graphene thin films are promising material for integration as transparent conductors in optoelectronic devices, due to their unique electronic and optical properties. Graphene is highly transparent in the visible photon energy region and has superb carrier mobility. However, due to its zero-gap band structure, graphene has a very low electrical carrier density. The carrier density in graphene can be enhanced by applying a gate voltage, or by appropriate doping. Here we present a combined experimental and theoretical investigation of large-area graphene thin films decorated with copper nanoparticles (Cu-NPs). The nanoparticles have been nucleated on graphene by radio frequency sputtering of Cu at room temperature and subsequent thermal annealing (Fig (c) and (e)). We observe a significant decrease in sheet resistance of graphene thin films by a factor of ~20 when doped with Cu-NPs (Fig (f)). Such a dramatic decrease is too large to be accounted for through the introduction of conduction pathways due to the presence of Cu-NP as depicted in Fig (b). Concurrent Scanning Kelvin Probe Microscopy (SKPM) (Fig (d)) investigations corroborated by computer simulations of the electronic density of states for Cu-NP decorated graphene flakes indicate that the local work function of graphene in the presence of overlayed Cu-NPs moves the Fermi level into the an electronic band (Fig (h)), well away from the canonical point. These shifts in the Fermi level of graphene are imaged by SKPM measurements and are also accompanied by changes in the work function of Cu-NPs, which depend on their size and concentration. Theoretically predicted shifts of the Fermi level in graphene thin films are in excellent agreement with the measured decreases in sheet resistance as a result of the increased charge carrier density. In particularly the shift in the Fermi level of doped graphene affecting the edge electronic states in finite graphene flakes.

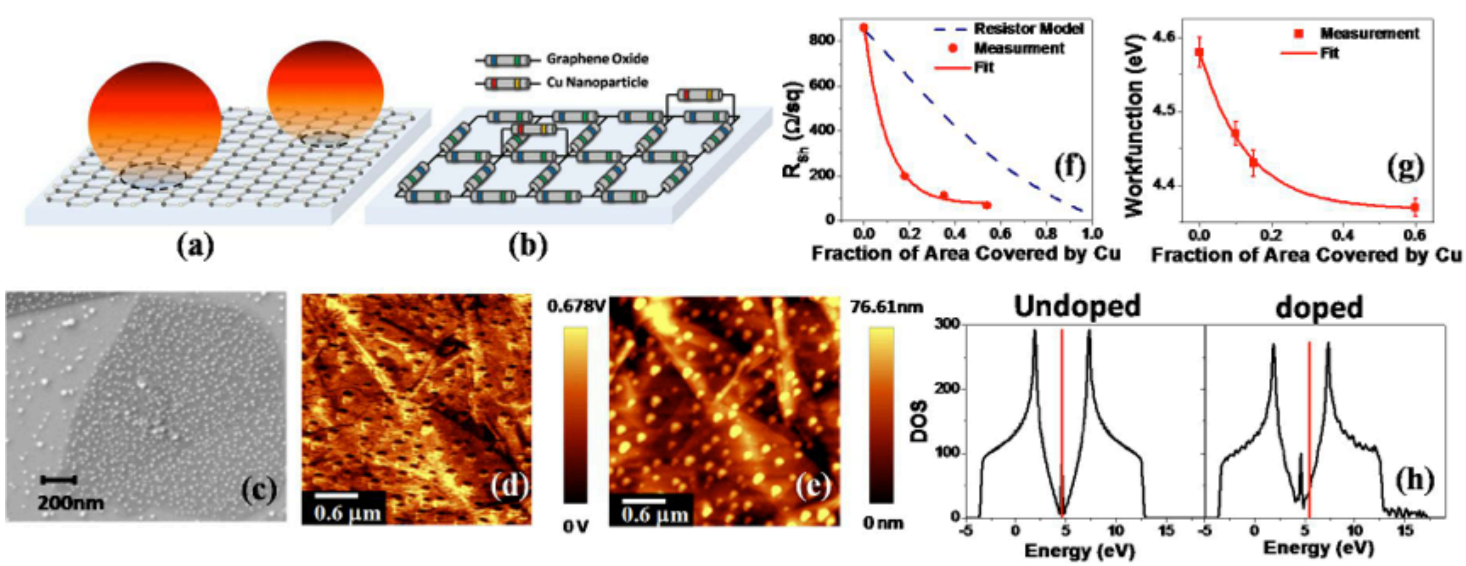

Figure: (a) schematic of Cu-NPs on graphene showing contact region. (b) Schematic of the equivalent resistor circuit. (c) Scanning electron micrograph, (d) Kelvin probe micrograph, and (e) atomic force micrograph of graphene decorated with Cu-NPs. (f) Measured sheet resistance vs. fraction of area covered by Cu-NPs and trend for the equivalent resistor model. (g) Work function of graphene vs. fraction of surface covered by Cu-NPs. (h) Electronic density of states for undoped and doped graphene calculated using a tight binding model (The vertical line is the calculated Fermi energy for the structure).

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