Abstract Submitted
for the MAR12 Meeting of
The American Physical Society

Chemical reactivity imprint lithography on graphene: Controlling the substrate influence on electron transfer reactions

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The chemical functionalization of graphene enables control over electronic properties and interactions with other materials. Graphene's chemical reactivity is strongly influenced by the underlying substrate. In this paper, we show a stark difference in the rate of electron transfer chemistry with aryl diazonium salts on monolayer graphene supported on a broad range of substrates. Reactions proceed rapidly when graphene is on SiO_2 and Al_2O_3 (sapphire), but negligibly on alkyl-terminated and hexagonal boron nitride (hBN) surfaces. The effect cannot be explained by the overall graphene doping levels alone, and can instead be described using a reactivity model accounting for substrate-induced electron-hole puddles in graphene. Raman spectroscopic mapping is used to characterize the effect of the substrates on graphene. Reactivity imprint lithography (RIL) is demonstrated as a technique for spatially patterning chemical groups on graphene by patterning the underlying substrate, and is applied to the covalent tethering of proteins on graphene.

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Date submitted: 05 Dec 2011 Electronic form version 1.4