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Highly selective covalent organic functionalization of epitaxial graphene

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Graphene functionalization with organics is expected to be an important step for the development of graphene-based materials with tailored electronic properties. However, its high chemical inertness makes difficult a controlled and selective covalent functionalization, and most of the works performed up to the date report electrostatic molecular adsorption or unruly functionalization. We show hereafter a mechanism for promoting highly specific covalent bonding of any amino-terminated molecule and a description of the operating processes. We show, by different experimental techniques and theoretical methods, that the excess of charge at carbon dangling-bonds formed on single-atomic vacancies at the graphene surface induces enhanced reactivity towards a selective oxidation of the amino group and subsequent integration of the nitrogen within the graphene network. Remarkably, functionalized surfaces retain the electronic properties of pristine graphene. This study opens the door for development of graphene-based interfaces, as nano-bio-hybrid composites, fabrication of dielectrics, plasmonics or spintronics.

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