Covalently grafted iron (II) phthalocyanine to graphene as a system for scalable molecular spintronics

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Hybrid systems consisting of transition metal phthalocyanine (TM-Pc) grafted to graphene constitute promising platforms for densely packed single-molecule magnets (SMMs). The most of the studies dealing with such hybrid systems considers the TM-Pc adsorbed to the graphene in such a way that the planes of two species are parallel to each other. Here, we selected iron(II) phthalocyanine (FePc) and investigated theoretically the possibility of grafting the molecule vertically to the graphene plane, which would guarantee much larger molecule packing density in the hybrid systems. We study the interaction of FePc with pristine and defective graphene layers employing density functional theory. Our calculations indicate that through proper dehydrogenation of the benzol rings in the FePc molecule, its adsorption to graphene is thermodynamically favorable. In general, the presence of anchoring sites on the graphene layer, *i.e.*, point defects, additionally facilitates the adsorption of FePc, allowing one to achieve high density of SMMs per unit area. Using the combination of group theory, ligand field splitting, and the calculated PBE0 Kohn-Sham eigenvalue spectrum, we resolve the electronic structure and predict the spin states of both isolated FePc and FePc-graphene hybrid systems. Regardless of the adsorption site and the number of removed hydrogen atoms from the benzol rings of FePc, the magnetic moment of the SMM remains unchanged with respect to the free FePc. These results should mediate a successful synthesis of densely packed TM-Pc systems and may open up a new avenue in designing scalable graphene-SMM systems for spintronics applications. The part of the results has been already published [1].

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