Spin-Density Interactions in Metal-Organic Networks/Graphene Heterostructures

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Abstract

The influence of a graphene substrate on the electron interaction within two-dimensional metal-organic networks (2D-MON) has been studied with the help of first principle DFT and DFT+U calculations. We have investigated a class of 2D-MON where magnetic Co atoms are bonded to polyphenyl-dicarbonitrile (PPDCN) linkers with different polyphenyl lengths (C_n, n = 1-3) to modulate the distance between Co atoms from 11.4 (C1) to 19.9 Å (C3). The freestanding 2D-MONs show a ferromagnetic (FM) behaviour that slowly turns to paramagnetic (PM) from C1 to C3 as the Co-Co distance increases. In contrast, the 2D-MON with short linker (C1) adsorbed on graphene demonstrates an antiferromagnetic (AFM) character that also gradually turns into a PM character for longer linkers (C2, C3). The presence of graphene drastically improves the spin interaction between Co atoms of the 2D-MON, and we can clearly observe a magnetic signature in the graphene substrate with spin-polarized scanning tunnelling microscopy (STM) simulations (Figure 1). Moreover, the Co atoms of the 2D-MON are strongly coupled to graphene and contribute to modify its electronic and magnetic properties. This work finally shows that 2D-MON can be used to introduce a magnetic character into graphene and to modify its electronic properties through a non-invasive process.

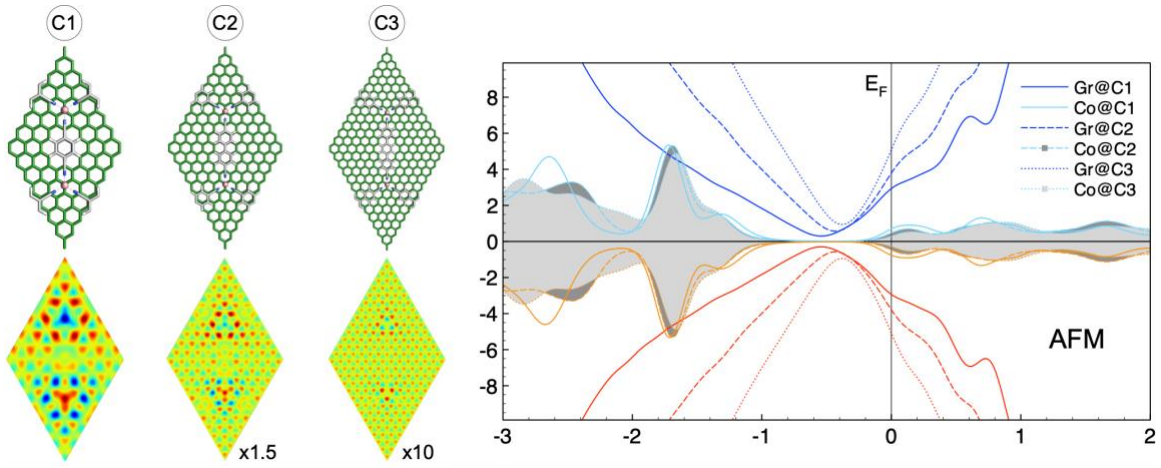

Figure 1. Top view of the unit cell (upper panels) used for the C1 to C3 models, and corresponding simulated STM images of the backplane of graphene (lower panels). Projected DOS of Co atoms and graphene for the AFM phase of C1 to C3 complexes.