Single-Molecule Magnets on Two Dimensional Materials: insights from ab initio calculations*

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Single-molecule magnets (SMMs) continue to be attractive candidates as building blocks for a variety of quantum technologies. They possess high spin and their magnetic properties may be tuned. They may also be assembled as dimers for further applications. The Mn_3 dimer is such an example in which the Mn_3 triangles are connected via ligands to form either the ferromagnetic (FM) or the antiferromagnetic (AFM) ground state.[1] One challenge for SMMs is finding a suitable substrate that keeps their magnetic properties intact, helps stabilize their assembly, and allows access to probes. In this talk, after presenting results of our spin-polarized density functional theory based calculations of the electronic structure and magnetic properties of the Mn_3 dimers themselves, we examine the modifications to these properties when the dimers are adsorbed on graphene and monolayer hexagonal boron nitride (*h*-BN), as well as when they are sandwiched between two layers of *h*-BN. Our calculations for the total spin of the FM (S = 12) and AFM configurations of the isolated dimers agrees with the experimental results [1] only when the dimers are charged (+2) (similar to molecules in solvent) and these values are retained on both supports. We find that the calculated magnetic exchange interactions to be in reasonable agreement with those deduced from experiments, except for the case of the charged FM dimer. Furthermore, both the FM and AFM configurations of the $[Mn_3]_2$ dimer retain their spin localization about the Mn^{3+} ions when deposited upon graphene. The binding energy for the system lies in the range 0.8 to 1.6 eV. We find very little charge transfer between the dimers and *h*-BN when the dimers are neutral and about 0.28 *e* when they are charged (+2). Interestingly, for the dimers on graphene, the charge transfer is about 0.3 *e* for the neutral dimes and about 1.58 *e* when they are charged. We compare the effect of these two substrate on the magnetic properties and electronic structure of the Mn_3 dimers and comment on the viability of the sandwiched structure for proposed experimental examinations of transport through the SMMs.

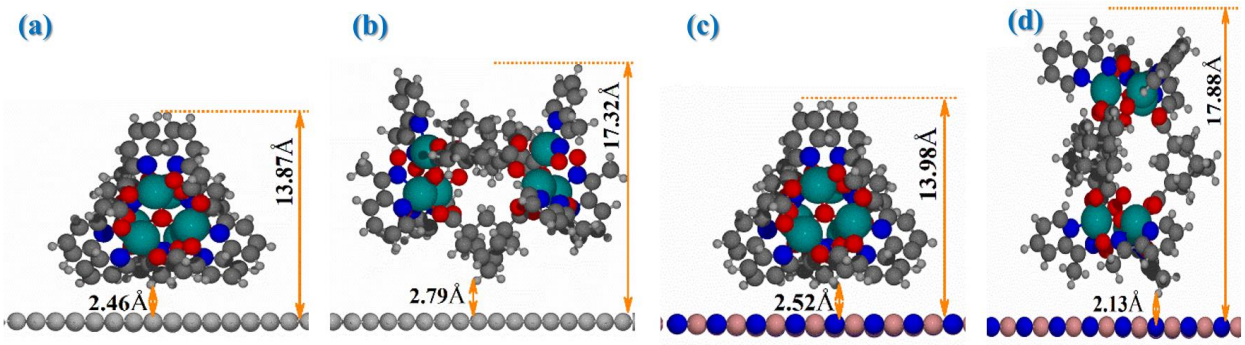

Fig. Supported Mn_3 dimers: a) FM on graphene; b) AFM on graphene; c) FM on *h*-BN; d) AFM on *h*-BN

[1] Nguyen et al. *J. Am. Chem. Soc.* 2015, 137, 7160−7168.

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