APPLICATION OF ADVANCED ATOMIC-SCALE TOOLS TO MULTI-FUNCTIONAL HYBRID ORGANIC-INORGANIC MATERIALS

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Hybrid organic-inorganic materials are at the forefront research in the field of multi-functional systems [1]. Among these materials, layered inorganic systems can be be organically modified by intercalation, grafting, exfoliation, etc.. These modifications, in turn, allow to build hybrid nano-structures with tuneable physico-chemical properties. The experimental team at IPCMS is particularly interested in layered hydroxides and oxides in view of their peculiar opto-electronic and magnetic properties. Yet, the chemical modification of inorganic layered structures by large flexible adducts induces disorder and the precise structure of these materials is often largely unknown. For instance, in hydroxides, the structure of the cobalt hydroxy-acetate phase ($Co_2(OH)_3(OAc)\cdot H_2O$) is only deduced on the basis of its analogy with the zinc hydroxy nitrate [1,2]. Given this scenario, a precise understanding of chemical bonding among all constitutive units involved is essential to guide the synthesis of new materials and to tune magnetic properties. Also, experiments do not always have access to the full set of properties (structural, electronic, magnetic) calling for contribution from theory. In recent works, in synergy between the experimental and computational groups at DCMI department, density functional theory (DFT) based molecular dynamics simulations have been done on the copper hydroxide acetate [3]. This has shown how different spin topologies can be realised and controlled by applying an external.

To get a deeper insight into the structure and reactivity of these solids, in this thesis, we shall focus on methodological issues and on applications related to **a)** accuracy and performance of exchange-correlation (XC) functionals and **b)** linear scheme approaches. This implies, the need of controlling the XC in systems for which DFT XC approaches are not fully satisfactory to make them more quantitative. Furthermore, the search of the stable structure of hybrid organic-inorganic materials and the characterization of their electronic and magnetic properties is still a challenge. Simulations of large systems (500-1000 atoms) [4] is required to describe global properties of the hybrid system having different chemical compositions and morphology. Particular attention will be given to the functionalization reaction pathway [5].

This PhD thesis project will be exploit the local HPC cluster at IPCMS, computational resources of the University of Strasbourg (EQUIP@MESO) and national centers. Moreover, the experience of our team in the development and use of the most advanced techniques and codes will provide a solid background for the formation of the candidate.

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