
HYBRID FUNCTIONALS AS A TOOL TO INVESTIGATE STRUCTURE, ELECTRONIC AND MAGNETIC PROPERTIES IN LAMELLAR ORGANIC-INORGANIC SOLIDS

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Lamellar organic-inorganic solids represent a prototype class of systems featuring a wealth of interesting correlations between structural organization, chemical properties and magnetic character. In a recent work, issued from a synergy within atomic-scale modeling team of the DCMI department, density-functional-theory (DFT) based molecular dynamics simulations have been done on the copper hydroxide acetate. This has shown that to different spin topologies, corresponding to the macroscopic magnetic states of copper hydroxide acetate, can be obtained, along with an estimate of the magnetic exchange coupling. More recently, pressure effects could be taken into account. Given these premises, it is of interest to pursue this research line not only by considering other systems (among those studied within the DCMI department at the IPCMS) but also by seeking methodological improvements, capable of bridging the gap between the still incomplete predictive power of DFT-based calculations and experiments. A possible workaround to improve the description of the exchange and correlation (XC) interactions of standard DFT, the stumbling block preventing a more quantitative description, is the use of a new class of XC functionals. It appears that functionals (termed hybrid functionals) combining the ordinary XC, often based on the homogeneous electron gas model, and the Hartree-Fock exchange are better suited to provide a quantitative description of the magnetic coupling in lamellar organic-inorganic materials and, as a general statement, can help when strong electronic correlation effects are present. Quite recently, the first-principles molecular dynamics program CPMD has been extended, by our and other groups, to make possible the use, within a computational strategy allowing for structural optimization, the use of hybrid exchange-correlation functionals (PBE0, B3LYP, HSE06, etc.) in a first-principles molecular dynamics algorithm.

In this thesis, we wish to pursue our research on organic-inorganic solids possessing interesting magneto-structural correlations by taking advantage of hybrid functionals on systems encompassing up to 500 atoms. For hybrid functionals, the computational effort becomes prohibitive in the case of the consideration of a real temporal trajectory, but it can be made more accessible when considering structural optimization only. This will be exactly our strategy, to be applied to selected systems among those currently under investigation at the IPCMS, for which structural data might or might not be available.