Title:	**Development of a first principles force field for metal-organic [Fe4] cages**
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Metal-organic cages (MOCs) containing four Fe(II) metal centres ($[Fe_4]$) represent a novel class of molecules that have attracted a lot of interest due to, among others, its efficiency in encapsulating greenhouse gases such as SF_6. On the top of that, the $[Fe_4]$ molecules exhibit spin-crossover (SCO) behaviour, which can be tuned as a function of the guest molecule encapsulated in the system. Due to their size, conventional electronic structure calculations are not suited to study the origin of such interaction and its effect on the SCO behaviour and different computational methodologies need to be developed in order to study the nature of the host-guest interaction.

First, by performing DFT calculations, the Fe-N bond will be modelled using a Morse potential. This data will be adjusted simultaneously against an energy scan along the totally symmetric normal mode in order to generate an *ab initio* force field for the molecule. Also, a series of electronic structure calculations will be carried out at CASSCF/NEVPT2 level to extract the relevant Angular Overlap Model parameters for a very simple molecule, $[Fe(NH_3)_6]^{2+}$, representative of the Fe(II) environment in $[Fe_4]$ MOCs; necessary to construct the force field.

In order to see if the results of $[Fe(NH_3)_6]^{2+}$ can be extrapolated to more complex systems like the $[Fe_4]$ cages, the ammonia results will be compared with systems of increasing complexity yet closely related to the actual Fe environment in the $[Fe_4]$ system: $[Fe(py)_6]^{2+}$ and $[Fe(C_6H_6N_2)_3]^{2+}$. Once validated, then the complete force field for the $[Fe_4]$ system will be assembled and tested.

Finally, LFMM simulations will be run in the NVE, NVT or NPT/NσT ensemble to study the interaction between the $[Fe_4]$ system and the SF_6 molecule in a large enough system so the crystal packing effects can be properly modelled.

Keywords: force field, electronic structure, transition metals, molecular mechanics, Ligand-Field Molecular Mechanics, ab initio ligand field theory, angular overlap model, density functional theory, LFMM, AILFT, AOM, DFT