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# Rational Design of Molecular Nano-Architectures on Complex Metallic Surfaces

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Self-assembly of 2D molecular films on metal surface templates presents major applications in the fields of molecular sensors/electronic, self-lubrication and photovoltaics. In this context, **the expansion of self-assembly on intermetallic complex surfaces with regards to usual simple metallic surfaces should help to achieve a better understanding of this process, and lead to the rational design of new systems with potentially innovative physical properties.** We therefore propose, in this thesis, to investigate these questions by numerical simulations, using 2D molecular films self-assembled on quasicrystalline-related substrates. Indeed, only a few attempts to formally refine the predictions of self-assembly have been made in materials science [1]. One major obstacle is that the corresponding thermodynamic equilibrium structure is the result of a subtle interplay between the energy gained upon molecular adsorption and association and the entropy lost during the surface confinement. To tackle this problematic, we will consider **model systems** constituted by **binary quasicrystalline approximants as substrates**, like  $\text{Al}_9\text{Co}_2(001)$  and  $\text{Al}_{13}\text{Co}_4(100)$ . The set of molecules will be  **$\text{C}_{60}$  molecules** as a primary objective since they are an archetypal model system for the proposed approach of self-assembly [2]. The method that will be implemented is based on **multi-scale approach** using the following tools: the Density Functional Theory (DFT) for electronic structure calculations, off-lattice Monte Carlo (MC) for simulating large 2D molecular patterns and Molecular Dynamic (MD) to have access to atomic trajectories. DFT calculations will be performed to set appropriate energy models both for the molecule-substrate and molecule-molecule interactions. The interatomic potentials will be developed at different degrees of simplification within the Tight-Binding (TB) method [3-5] to obtain a set of modular energy models while keeping a significant influence of the electronic structure. A preliminary result for the template surface  $\text{Al}_{13}\text{Co}_4(100)$  is shown in Fig. 1. The interatomic potentials have been fitted on DFT calculations for metal-metal interactions and used in classical molecular dynamics (MD). This preliminary work, funded in 2020 by the LABEX NIE, has been performed in collaboration with E. Gaudry (IJL Nancy). This close cooperation will naturally continue as part of the thesis.

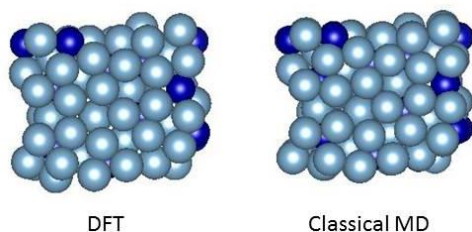


Fig. 1 : Illustration of the comparison between DFT and MD for one of the most favorable obtained structures of an  $\text{Al}_{13}\text{Co}_4(100)$  surface (top view).

## References

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