Understanding molecule-surface interactions with DFT calculations

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The adsorption of molecules on solid surfaces is a problem of great importance and difficulty. From catalysis to molecular devices, the interactions between molecules and surfaces are determining. In order to understand them, many degrees of freedom need to be accounted for, some of which need complex quantum calculations. The mean-field approach of present implementations density functional theory (DFT) limits the applicability of DFT when many-body effect and quantum fluctuations become important, but it permits us to perform calculations in realistic systems.

I will summarize our research in three systems that we find interesting: the first one is the presence of different adsorption patterns in CO on Cu (111) as revealed by the scanning tunneling microscope. In this research, the modelling of the patterns were complicated by the difficulty to understand the topology of the images plus the complicated interactions between molecules and between molecules and substrate. We showed that the electric field of the STM induced a phase transition between the rationalized patterns [1].

The second topic will be the study of the role of adatoms stabilizing molecular structures. From some rare cases studied in the early 2000's, to now, the role of adatoms seem to be increasingly ubiquituous in many molecular structures on noble metal substrates. I will briefly explain our research on the supramolecular patters appearing during the adsorption of biphenyl molecules on Au (111), and how the interplay of charge transfer and van der Waals interactions become relevant in understanding the dynamics of these molecules on the surface [2,3].

Finally, I will present the spontaneous formation of molecular chains when nickelocene molecules are adsorbed on Au (111). This is an example of how DFT-based simulations and experimental data can be matched to understand a quite surprising effect [4].

References:
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