

Epitaxial growth of polycrystalline C₆₀ assemblies on metallic substrates

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Growth of buckminsterfullerenes (C₆₀) on metallic substrates has been extensively studied because of its potential applications for molecular electronics [1]. Knowing the details about the interaction and interface between C₆₀ and metals is a key aspect to control electrode-to-molecule charge injection. As a consequence, many structural and electronic studies of single C₆₀ on metallic substrates and monocrystalline layers of C₆₀ have been performed [2-4]. Most of these studies focus on microscopic details about how fullerenes interact with the metallic substrate: charge transfer, orbital hybridization, adsorption site, molecular orientation, rearrangement of metallic atoms, etc.

Here we concentrate on the epitaxy of the molecular layer during the first steps of growth, that expectedly determine the quality of the interfaces and quality of the film. When depositing less than a monolayer of fullerenes on metal surfaces polycrystals form, showing phases with different crystallographic orientations, lattice parameters and complex superstructures with their substrate. On several metallic surfaces, for instance Cu(111), a puzzling coexistence of metastable phases with different structural parameters is observed, including ordered and disordered phases [5,6]. Two questions remains open. Why several molecular phases can coexist on the same sample? What is the relation between the crystallographic orientation and the more or less complex molecular height patterns?

To address these questions we studied C₆₀ molecules on Cu (111) and deposited them by molecular beam epitaxy under ultra high vacuum. We used scanning tunneling microscopy to inspect the different phases (five in total, cf Fig. 1 for a few of them). We built a molecular structural phase diagram, using a simple model that considers only elastic energy stored in the molecular layer and the degree of molecule-to-substrate structural coincidence. Our approach is general, and reveals that the observed phases are indeed stable or metastable molecular phases, not only on copper but also on other metal surfaces. With the input of density functional calculations, we manage to identify the main parameters that govern the formation of superstructure patterns. Competition between biaxial strain of the molecular layer and C₆₀-metal interaction explains the existence of uniform, periodic and disordered phases.

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[4] Sakurai et al., Prog. Surf. Sci., **51**, 263-408 (1996)

[5] Pai et al., Phys. Rev. B, **69**, 125405 (2004)

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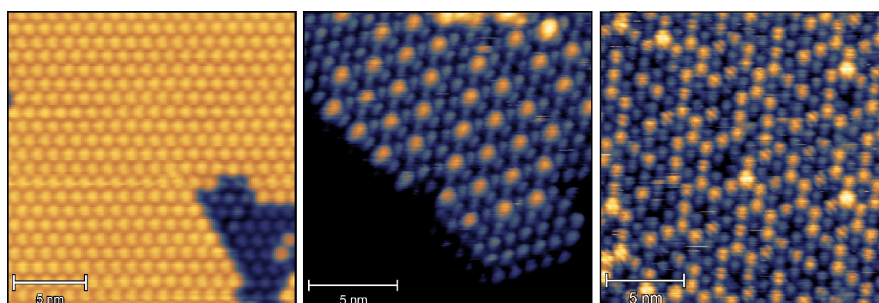


Fig. 1 : Scanning tunneling microscopy images of fullerenes on Cu(111). Each colored dot corresponds to one molecule. From left to right: uniform-height phase, superstructure phase and disordered phase. On each image, the dense molecular rows have a specific orientation.