Simulations of constant current images of open-shell systems

Mikaël Kepenekian[1], Richard Korytár[1,2], Roberto Robles[1] and Nicolás Lorente[1]

[1]CIN2, Consejo Superior de Investigaciones Científicas, 08193 Bellaterra, Spain

[2]Karlsruhe Institut of Technology, Karlsruhe, Germany

Open-shell systems are challenging objects. For scanning tunneling microscopy (STM) imaging, matters become complex when the open-shell structure involves a multiconfigurational electronic structure. In this contribution, we will show results on STM imaging simulations of a copper phthalocyanine (CuPc) on a Ag (100) substrate, as motivated by the experiments of Mugarza et al. [1]. CuPc is an open-shell molecule with a magnetic moment of S=½. It is thus a rather simple molecule. However, when adsorbed an extra electron is captured giving rise to a S=1 ground state configuration which is then open-shell and multiconfigurational. This leads to exotic behavior such as Kondo correlations.

We first study the system using typical theoretical tools such as density functional theory (DFT) and different levels of transport calculations. We compare the simple approach by Tersoff and Hamman [2] with the more involved of Landauer [3]. However, present forms of DFT imply a mean-field approach that erases the multiconfigurational study. For this we have developed a non-crossing approximation (NCA) approach [4] based on the Baym-Kadanoff formalism in the limit where we keep the full multiconfigurational Hilbert space but we reduced the Fock space to infinite correlation.

We find that while Tersoff-Hamman gives a good qualitative picture, DFT-based methods fail in reproducing the experimental conductance behavior and only NCA can give an account of the Kondo phenomena.

References:

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