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Magnetic Properties of Ultrathin Metal-Organic Layers on Metal Surfaces: Supramolecules and Macrocycles¹

PIETRO GAMBARDELLA, ICREA and Centre d'Investigacions en Nanociència i Nanotecnologia (ICN-CSIC, Barcelona, Spain

The design of molecular films with tunable electronic and magnetic properties has become a major goal of current research. Metal-organic complexes at surfaces provide versatile ways to achieve this goal by controlling the metal-ligand and metal-substrate hybridization. In this talk, we discuss two approaches to lay out the structure and magnetic properties of molecular layers on metallic substrates based on transition metal-directed supramolecular assembly [1] and the deposition of macrocycle molecular films [2]. Using a combination of scanning tunnelling microscopy, x-ray magnetic circular dichroism, ligand field and density functional calculations we study the interplay of molecular and metal states, addressing the spin and orbital moments of the molecules as well as their magnetic anisotropy. We show that, under certain conditions, these quantities can be controlled by the number and symmetry of the organic ligands independently from the substrate. We reveal unusual phenomena such as 500% giant orbital moment anisotropy in metal-phthalocyanine complexes and molecular Kondo states, which we contrast with measurements of metal atoms and nanoparticles at surfaces [3,4]. The charge state of the metal ions and their electronic coupling with the surface is analyzed in terms of multiplet simulations of x-ray absorption spectra and compared to density functional theory results.

[1] P. Gambardella et al., *Nature Mater.* **8**, 189 (2009).

[2] S. Stepanow et al., to be published.

[3] H. Brune and P. Gambardella, *Surf. Sci.* **603**, 1830 (2009)

[4] P. Gambardella et al., *Science* **300**, 1130 (2003).

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