X-ray Spectroscopy on Molecular Magnets: Magnetometry, Electronic Structure, and more

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The last decade of the past century witnessed the advent of both single molecule magnets (SMMs) and third generation synchrotron sources. The observation of quantum phenomena and the potential of SMMs to store information at the molecular level, boosted the interest of the scientific community on them. In parallel, X-ray scattering techniques have evolved to be irreplaceable tools in the study of magnetism of materials.

Many potential applications of molecular magnets imply its deposition onto a surface, and the preparation of small structures. Cyclic complexes are of particular interest because of their planar high symmetry. I will present recent work on wheel molecules of formula $\{Cr_{10}(OMe)_{20}(O_2CCMe_3)_{10}\}$ (Me = methyl), $[Cr_{10}]$, both in bulk and evaporated by direct sublimation in UHV on metallic single-crystal surfaces. A 2D self-organized quasi-hexagonal network monolayer of $[Cr_{10}]$ molecules, is formed on Cu(111) or Au(111). The magnetic characterization of monolayer, multilayer and bulk (powder and singlecrystal) samples of $[Cr_{10}]$ has revealed that this cluster exhibits an unusually high Sz-ground state.

Molecular overlayers on substrates have also a broad field of application in catalysis, sensing, molecular electronics, light-to-energy conversion, etc. I will present our recent work on Iron-phtalocyanines (FePc), which have been proposed as substitutes for precious metals in catalysis of the Oxygen Reduction Reaction. Sub-monolayer phases of FePc on Ag(110) are catalitically active. Remarkably, in oxygen-dosed phases, O₂ intercalates between the molecules and the surface, substantially changing the Fe magnetic moment [1]. XMCD allows to determine the distribution of non-oxygenated, and oxygenated species.

The path from the bulk to the surface in molecular magnetism implies a change in the available experimental techniques, in particular for the study of magnetic relaxation. In the bulk, relaxation is mostly studied by AC magnetic susceptibility, but when characterizing a thin-film sample, the signal from the substrate is orders of magnitude larger than the one from the sample itself. Due to its brilliance and chemical selectivity, XMCD has become the magnetometry of choice in surface-deposited molecular magnetism [2]. However, the only remaining witness of slow magnetic relaxation is bistability, which only appears in extremely slow relaxing systems, by opening the hysteresis loop. The information about the processes originating SMM behavior is mostly lost. The development of *user-friendly* X-ray Scattering techniques to study magnetic dynamics would be of great interest in the future. Hopefully, EBS may allow the inception of such experimental techniques at the ESRF.

References

- [1] J. Bartolomé, et al., J. Phys. Chem. C 2015, 119, 12488.
- [2] F. Bartolomé et al., Handbook of Magnetic Materials, edited by Ekkes Brück, vol. 26, pg. 1 (2017).