

## SLS Symposium on

### Novel Materials

Tuesday, November 5, 2013

10:00 to 12:15, WBGB/019

#### 10:00 Chemically programmable 2D electron spin array

*J. Nowakowski, Ch. Wäckerlin, S.-X. Liu, M. Jaggi, J. Girovsky, D. Siewert, A. Shchyrba, T. Hählen, A. Kleibert, P.M. Oppeneer, F. Nolting, S. Decurtins, T. A. Jung and N. Ballav*

#### 10:30 Temperature-dependent incommensurability in orthorhombic LuMnO<sub>3</sub> epitaxial films

*Y. W. Windsor, S. W. Huang, L. Rettig, A. Alberca, V. Scagnoli, U. Staub, Y. Hu, T. Lippert and C. W. Schneider*

#### 11:00 Coffee

#### 11:15 Tunneling, Remanence and Frustration in Dysprosium based Endohedral Single-Molecule Magnets

*R. Westerström, J. Dreiser, C. Piamonteze, R. Stania, M. Muntwiler, S. Weyeneth, K. Krämer, S.X. Liu, S. Decurtins, A. Popov, S. Yang, L. Dunsch, and T. Greber*

#### 11:45 Peculiar Magnetic and Orbital Excitations in Cuprates Captured by Resonant Inelastic X-ray Scattering

*V. Bisogni, C. Monney, K. Zhou, V. Strocov and T. Schmitt, K. Wohlfeld, S. Kourtis, S. Nishimoto, J. Trinkauf, R. Kraus, C. Sekar, B. Buechner, M. Daghofer, J. van den Brink and J. Geck*

## Chemically programmable 2D electron spin array

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Dorota Siewert,<sup>[a]</sup> Aneliia Shchyrba,<sup>[c]</sup> Tatjana Hählen,<sup>[a]</sup> Armin Kleibert,<sup>[d]</sup> Peter M.  
Oppeneer,<sup>[e]</sup> Frithjof Nolting,<sup>[d]</sup> Silvio Decurtins,<sup>[b]</sup> Thomas A. Jung,<sup>\*,[a]</sup> Nirmalya Ballav,<sup>\*,[f]</sup>

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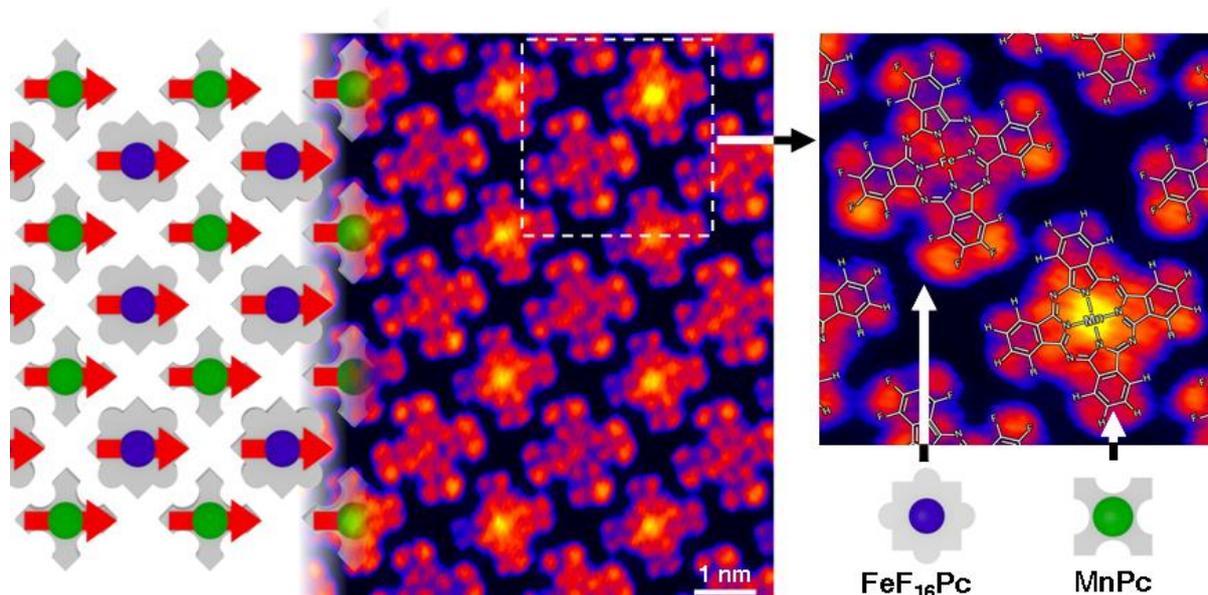
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In 2005 Scheybal et al. showed that metallorganic molecules (e.g. MTPP, MPc) can be magnetically coupled to a ferromagnetic substrate [1]. Magnetization of such molecules can be switched off [2], switched on [3], tuned or changed from ferromagnetic (FM) to antiferromagnetic (AFM) [4] by means of a chemical stimulus (i.e. coordination of a ligand). All of those modifications can be reversed by desorbing the ligand [2-4]. In the presented work [5] a 2D magnetic chessboard self-assembled by MnPc and FeFPC molecules is presented and the spin state of the system is changed from ON/ON to ON/OFF by an external ligand (NH<sub>3</sub>) and restored by its controlled thermal desorption (Fig. 1). The process of spin-state switching was studied by an element-specific, synchrotron technique – X-ray Magnetic Circular Dichroism (XMCD) at the Surface/Interface Microscopy (SIM) beamline of the SLS.



**Figure 1** Scanning Tunneling Micrograph of the supramolecular magnetic chessboard with a puzzle-pieces representation identifying the different species

- [1] A. Scheybal, et al., *Chem. Phys. Lett.*, **411** (2005), 214.  
[2] C. Wäckerlin, et al., *Nat. Commun.* **1** (2010), 61.  
[3] C. Wäckerlin, et al., *Angew Chem, Int. Ed.* **52** (2013), 4568.  
[4] C. Wäckerlin, et al., *Chem. Sci.* **3** (2012), 3154.  
[5] C. Wäckerlin, et al., *Adv. Mat.* **25** (2013), 2377.

# Temperature-dependent incommensurability in orthorhombic LuMnO<sub>3</sub> epitaxial films

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Recent years have seen an increasing interest in Type-II multiferroics, where the electric polarization is intrinsically coupled to the magnetic order [1]. The very large magnetoelectric coupling in such materials makes it feasible to manipulate the magnetic order by electric fields, a key capability for future spintronic applications due to the higher efficiency in applying voltages rather than currents. In this context, it is interesting to study thin films due to the ability to manipulate and control the electronic and magnetic properties by varying the strain (e.g. via the substrate). Orthorhombic *REMnO<sub>3</sub>* perovskites (*RE* = rare earth) are a family of magnetic oxides. Using the heaviest *RE* ions (*RE*=Ho, Tm, Yb, Lu) these become type-II multiferroics with an E-type antiferromagnetic spin structure ( $\uparrow\text{-}\uparrow\text{-}\downarrow\text{-}\downarrow$ ). The E-type structure produces a very large polarization due to symmetric exchange striction, with *RE*=Lu having the largest value and the highest  $T_C$  in this family [2]. Unfortunately, these heavier *RE* systems cannot be synthesized under ambient pressures so high-quality single crystals are not available in bulk form.

In this talk I will present our ongoing study of orthorhombic LuMnO<sub>3</sub> epitaxial films, a material of very recent interest [3]. Using resonant soft x-ray diffraction (RSXD) at the SIM beamline we find that the magnetic ordering differs from the commensurate E-type structure expected in bulk, and instead exhibits an incommensurate and highly T-dependent *quasi-E-type* spin structure. Further experiments varying thickness and substrate have been conducted, as well as x-ray diffraction (XRD) at the Materials Science beamline, in which variations in strain and signatures of a ferroelectric distortion were identified. I will discuss how these findings fit together, and how these results may have implications on other E-type systems or other *REMnO<sub>3</sub>* perovskites (such as the well-known case of TbMnO<sub>3</sub>).

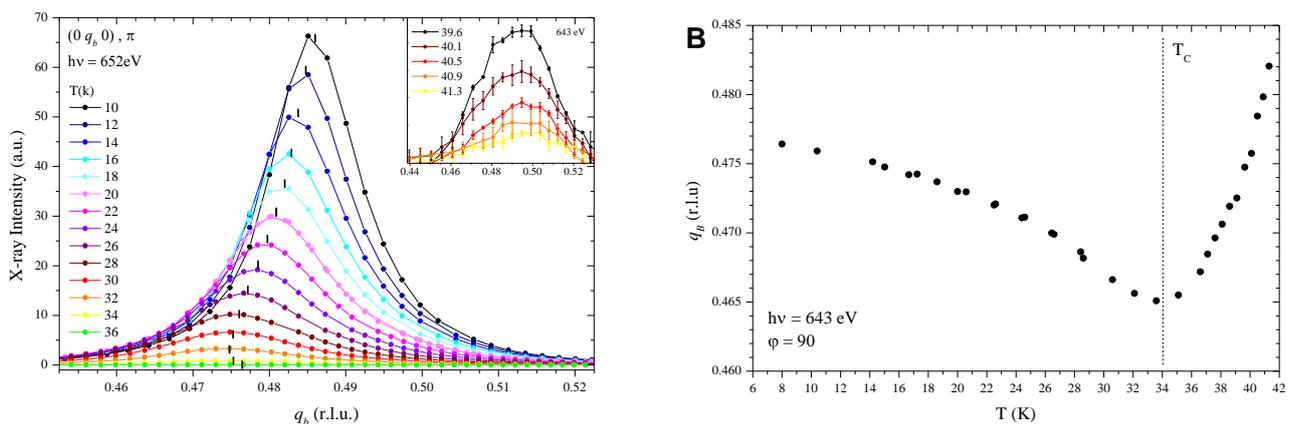


Fig. 1 (A) Magnetic  $(0 q_B 0)$  reflection with  $q_B \approx 1/2$  at varying  $T$ 's, exhibiting variations in intensity and peak position. Inset: high- $T$  peaks. (B) Incommensurability of  $q_B$  at all  $T$ 's, with a clear signature of  $T_C$ , below which electric polarization appears.

- [1] T. Kimura, et al., Nature 426, 55 (2003)
- [2] S. Ishiwata et al., Phys. Rev B 81, 100411(R) (2010)
- [3] J. S. White et al., Phys. Rev. Lett. 111, 037201 (2013)

# Tunneling, Remanence and Frustration in Dysprosium based Endohedral Single-Molecule Magnets

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Endofullerenes [1] represent a new family [2] in the class of lanthanide-based single molecule magnets (SMMs) [3]. The carbon cage offers a chemically protected nanometer sized environment where the magnetism of 4f ions can be systematically studied. Here we present results from the three SMMs Dy<sub>n</sub>Sc<sub>3-n</sub>N@C<sub>80</sub> (n=1,2,3) with one, two, or three magnetic moments. The three significantly different hysteresis curves (Fig. 1) demonstrate the decisive influence of the number of magnetic moments and their interactions. For **2** the intra-molecular interactions result in a blocking of the magnetization and a zero-field relaxation time of ~1 h at 2 K. In the case of **3** the magnetic coupling leads to a ground state which turns out to be one of the simplest realizations of a frustrated, *ferromagnetically*, coupled magnetic system.

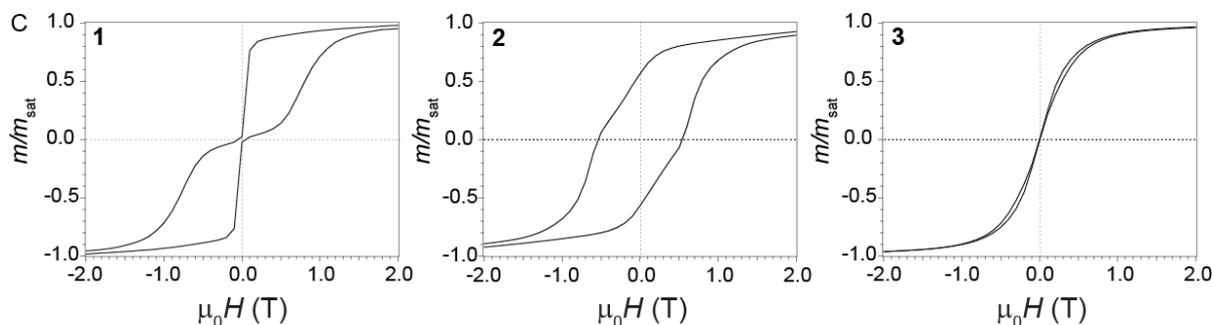
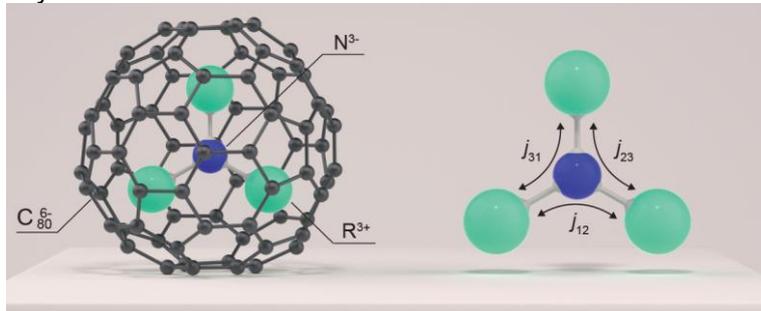


Figure: (A) Model of R<sub>3</sub>N@C<sub>80</sub> R=Rare earth (here Dy or Sc) (B) The endohedral R<sub>3</sub><sup>3+</sup>N<sup>3-</sup> unit and the corresponding couplings  $j_{i,k}$  that are partially mediated across the N<sup>3-</sup> ion. (c) Hysteresis curves recorded by SQUID at 2 K at a field sweep rate of 0.8 mTs<sup>-1</sup>.

[1] A. Popov *et al*, Chem. Rev., Chem. Rev. **113** 5989 (2013).; [2] R. Westerström *et al*, J. Am. Chem. Soc., (2012), **134**, 9840; [3] N. Ishikawa *et al*, J. Am. Chem. Soc., (2003), **125**, 8694

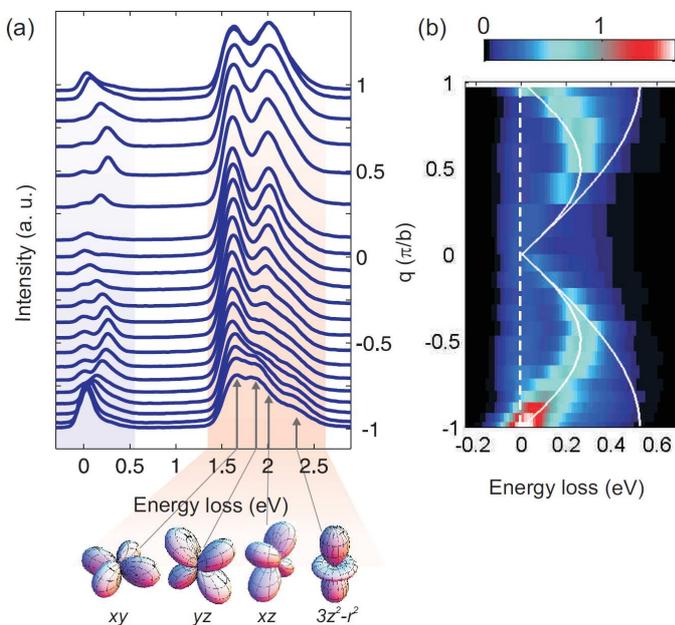
# Peculiar Magnetic and Orbital Excitations in Cuprates Captured by Resonant Inelastic X-ray Scattering

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Recently, resonant inelastic X-ray scattering (RIXS) on the 1D spin chain system  $\text{Sr}_2\text{CuO}_3$  has revealed an unprecedented and strong dispersion of  $dd$  orbital excitations. This result, observed in [1] for the first time, has been interpreted as the spin and orbital separation from the elementary electron in a 1D  $S=1/2$  system, similarly to the spin-holon separation observed in previous angle-resolved photoemission studies of the same system [2]. In this talk we report on the Cu  $L_3$  RIXS investigation of the two-leg ladder compound  $\text{CaCu}_2\text{O}_3$ , consisting of two coupled spin chains. Due to its buckled geometry, the inter-chain interaction of this system is one order of magnitude smaller than the in-chain interaction, but not negligible [3]. Therefore,  $\text{CaCu}_2\text{O}_3$  is an ideal model system to study the effect of a weak inter-chain interaction, i.e. the effect of a dimensionality higher than 1D, on both the high [ $dd$  excitations, Fig. (a)] and the low [spin excitation, Fig. (b)] energy scale.



The dispersion of the spinon continuum can be largely accounted for by neglecting the presence of the inter-chain interaction, as far as it concerns the energy range accessible to Cu  $L_3$  RIXS. A detailed analysis of the two spinons excitations will be also presented showing indeed how RIXS is sensitive to the time dynamics of the magnetic excitations [4].

In the high energy range typical of  $dd$  excitations, a dispersing  $3d_{xz}$  orbital excitation is found demonstrating that spin-orbital separation persists in a system with a dimensionality higher than 1D. With the support of theoretical results, we establish that such a fractionalization is far more robust than spin-charge separation [5].

## References

- [1] J. Schlappa, et al., Nature 485, 82 (2012).
- [2] C. Kim, et al., Physical Review Letters 77, 4054 (1996).
- [3] B. Lake et al., Nature Physics 6, 50 (2010).
- [4] V. Bisogni, et al., arXiv:1307.8393.
- [5] V. Bisogni, et al., Submitted to Physical Review Letters.