Welcome

Il Dipartimento di Scienze Chimiche accoglie il prof. Alessandro Soncini che terrà un seminario dal titolo:

Single Molecule Toroics: What theory can do for you

Martedì 14 febbraio 2023, ore 16.45 Aula H, Dipartimento di Scienze Chimiche, Via Marzolo, 1.

Molecular nanomagnets (MNMs) based on transition metal and lanthanide ions are strong contenders in the quest for the ultimate miniaturization of electronic devices for both classical[1] and quantum[2] computing technologies, due to the possibility of encoding and processing information in their giant spin states. However, achievement of long magnetic-relaxation times (pivotal to develop molecular memories), and long spin decoherence times (pivotal to develop molecular qubits), are hampered by the strong dipolar fields produced by neighboring molecules. A spin-based quantum degree of freedom, topologically non-equivalent to the MNMs giant spin, consists of the *toroidal moment*, arising in the ground state of special spin rings, where local spin moments couple into collective magnetic vortex states[3] featuring a vanishingly small magnetic moment (hence vanishing dipolar fields), in Single Molecule Toroics (SMTs)[3-6]. Here we discuss various SMTs we identified in polynuclear 4f[5,6], hybrid 3d-4f[7-9], and 3d metal complexes[10], in both strong and weak spin-orbit coupling limits. Especially for the strong spin-orbit coupled case of rare earth compounds, scalar relativistic multiconfigurational ab initio calculations play a key role in the identification of SMTs. We discuss how models based on such calculations are set up, including ab initio methods developed in our group[11,12]. We present applications to 3d-4f polynuclear complexes Dy_3MDy_3 (M = Cr^{3+}, Fe^{3+}, Co^{3+}, Al^{3+}) displaying con-rotating (*ferrotoroidic*) and counter-rotating (*antiferrotoroidic*) coupled toroidal units[7–9], and to a large $Fe_{10}Dy_{10}$ non-planar ring featuring a manifold of hybrid toroidal-magnetic excitations. Limitations of current ab initio approaches are discussed, as exposed by our work on 4f charge density maps[13,14], and by our recently developed ab initio non-covalent crystal field approach based on non-orthogonal group functions[12].

1. Guo, F.-S. et al. Magnetic hysteresis up to 80 kelvin in a dysprosium metallocene single-molecule magnet. *Science* **2018**, 362, 1400
2. M. Atzori et al., Room-Temperature Quantum Coherence and Rabi Oscillations in Vanadyl Phthalocyanine. *J. Am. Chem. Soc.* **2016**, *138*, 2154-2157
3. Soncini and L. F. Chibotaru, Toroidal magnetic states in molecular wheels *Phys. Rev. B* **2008**, *77*, 220406
4. N.A. Spaldin et al. Toroidal moment in condensed matter physics and its relation to the magnetoelectric effect *J. Phys: Cond. Matt.* **2008,** *20*, 434203.
5. L. Chibotaru, L. Ungur, A. Soncini The origin of nonmagnetic Kramers doublets in the ground state of dysprosium triangle: Evidence for a Toroidal Magnetic Moment. *Angew. Chem. Int. Ed.* **2008**, *47*, 4126.
6. J. Luzon et al. Spin Chirality in Molecular dysprosium triangle: The archetype of a the noncollinear Ising model. *Phys. Rev. Lett.* **2008**, *100*, 247205
7. Vignesh K, Soncini A. et al., Ferrotoroidic ground state in a $CrDy_6$ complex displaying slow magnetic relaxation. *Nature Comm.* **2017**, *8*, 1023.
8. J. Ashtree et al. Tuning the ferrotoroidic coupling and magnetic hysteresis in double-triangle complexes {Dy_3MDy_3} via the M-linker. *Eur. J. Inorg. Chem.* **2021**, 435.
9. K. Hymas, A. Soncini, Ch. 6 and K. Murray, A. Soncini et al. Ch. 2, in *Single Molecule Toroics*, *Ed.* K. Murray, Springer Nature, Switzerland AG **2022**
10. J. Crabtree, A. Soncini, Toroidal quantum states inspin-frustrated triangular nanomagnets with weak spin-orbit coupling, *PRB* **2018**, *98*, 094417
11. S. Calvello, M. Piccardo, S. V. Rao, A. Soncini, "CERES: An ab initio code dedicated to the calculation of the electronic structure and magnetic properties of lanthanide complexes" *J. Comp. Chem.* **2018**, 39, 328-337
12. Soncini A., Piccardo M., Ab initio non-covalent crystal field theory for lanthanide complexes. *Phys. Chem. Chem. Phys.* **2022**, *24*, 18915-18930
13. C. Gao, A. Genoni, S. Gao, S. Jiang, A. Soncini, J. Overgaard, *Nature Chemistry* **2020**, *12*, 213-219
14. N. F. Chilton, D. Collison, E. J. L. McInnes, R. E. P. Winpenny, A. Soncini, *Nature Communications* **2013**, 4, 2551

Il Direttore del Dipartimento

Michele Maggini