Quantum Effects in Single Molecule Magnets

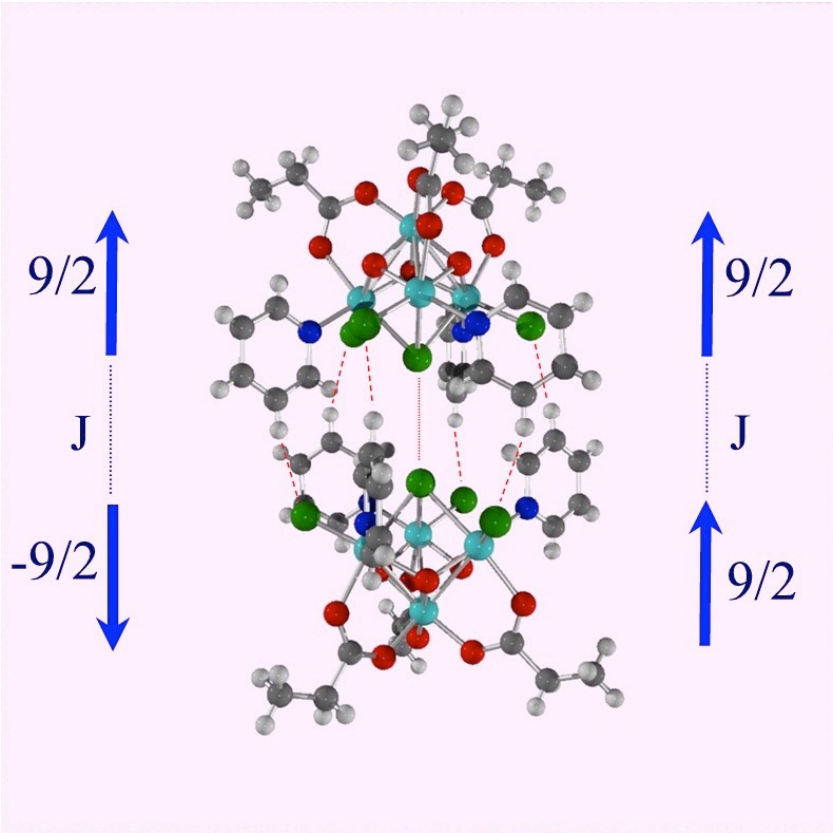
NSF-Nanoscale Interdisciplinary Research Team, DMR-0103290-01

Andrew Kent (NYU), George Christou (UF), Naresh Dalal (FSU), Stephen Hill (UF) and David Hendrickson (UCSD)

This project focuses on the synthesis and study of molecules that contain a small number of metal atoms, such as 12 manganese ions or 8 iron ions, that lock their individual magnetic moments together to form a larger and coherent "nanomagnet". The resulting moment of the molecule, unlike that of a single ion, is potentially large enough to detect and to use in an electronic device to process or store information. Further, crystalline arrays of such nanomagnets have been synthesized in which each element of the array is a magnetic molecule, also known as a single molecule magnet. SMMs enable fundamental studies of properties intrinsic to magnetic nanostructures that have previously been inaccessible. For example, recent studies of SMMs have revealed the quantum nature of the spin-dynamics in a nanomagnet [1], and raised basic questions about this behavior that are not yet understood. A metastable state of the magnetization, say “spin-up,” has been convincingly shown to decay by quantum tunneling through a magnetic anisotropy barrier to a “spin-down” state, in a process called quantum tunneling of magnetization (QTM). Remarkably, QTM in SMMs can be switched on and off with small applied fields or by exchange interactions between neighboring SMMs [2] enabling quantum control and potentially quantum information processing.

The molecules in this array are thought to be inherently "alike", just like all sugar molecules are alike. This route to arrays of nanomagnets may be contrasted with the alternative method that uses slow and complex electron beam lithographic techniques to create arrays of nanomagnetic metals, such as iron and cobalt–that are far from monodisperse in their magnetic characteristics. Until recently, the sameness of SMMs, like sugar molecules, was taken for granted. In addition, magnetic exchange forces between molecules in arrays were assumed to be small.

In research this past year by this NIRT team it has been shown that the molecules are not always magnetically identical and also that nearby molecules can interact via exchange forces with one another [2-6]. Magnetic differences and intermolecular interactions are detected as subtle features of the electron magnetic resonance spectrum, nuclear magnetic resonance, and magnetic properties of single crystals. Strong evidence has been found that the distribution of magnetic properties is associated with disorder in the solvent molecules that surround the core of transition metal ions. These effects, previously undetected, will need to be understood at a fundamental level if these molecules are to be used as device elements. Their study requires a team effort, involving chemists and physicists and state-of-the-art instrumentation at New York University, University of California at San Diego, University of Florida and Florida State University and the National High Magnetic Field Laboratory.



The structure of the $[Mn_4O_3Cl_4(O_2CEt)_3(py)_3]_2$ dimer, denoted $[Mn_4]_2$. Each Mn_4 has a spin of 9/2 and applies a large internal "exchange field" to its pair in the dimer via the hydrogen bonds (dashed lines in the figure). Red, oxygen; cyan, manganese; green, chlorine; blue, nitrogen; gray, hydrogen; black, carbon.

References and Recent Group Publications:

1. G. Christou, D. Gatteschi, D. N. Hendrickson, and R. Sessoli, MRS Bulletin, Volume **25**, No. 11, 66-71 (2000).
2. W. Wernsdorfer, N. Aliaga-Alcalde, D. N. Hendrickson, G. Christou, Nature **416**, 406-409 (2002).
3. R. S. Edwards, S. Maccagnano, E.-C. Yang, S. Hill, W. Wernsdorfer, D. Hendrickson and G. Christou, J. Appl. Physics (submitted 2002)
4. K. Park, M. A. Novotny, N. S. Dalal, S. Hill and P. A. Rikvold, Phys. Rev. B **65**, 14426 (2002); J. Appl. Phys. **91**, 7167 (2002); Phys. Rev. B, October 2002 (to be published); also, cond-mat/0204481 (April 24. 2002); S. Hill, S. Maccagnano, K. Park, R. M. Achey, J. M. North and N. S. Dalal, Phys. Rev. B **65**, 224410 (2002); K. Park, N.S. Dalal and P.A. Rikvold, J. Chem. Phys. (to be published, 2002);
5. E. del Barco, A. D. Kent, E. Rumberger, D. N. Hendrickson and G. Christou, Europhysics Letters in press, cond-mat/0209167
6. R. M. Achey, P.L. Kuhns, A. P. Reyes, W. G. Moulton, and N.S. Dalal, Phys. Rev. B 64, 64420 (2001); Solid St. Comm. 121, 107 (2002).