

Preface to the JPCM Special Issue on Molecular Magnetism

Birgit Weber,¹ and Peter A. Dowben,²

¹ Inorganic Chemistry IV, University of Bayreuth, Universitätsstrasse 30, NW I, 95440 Bayreuth, Germany

² Department of Physics and Astronomy, University of Nebraska – Lincoln, Lincoln, NE 68588, U.S.A.

e-mail: weber@uni-bayreuth.de; pdowben1@unl.edu

This is a special issue on molecular magnets, and a special issue long awaited by many JPCM board members, past and present. The interest in molecular magnets is long standing and ranges from the fundamental aspects to the more practical implementations. The premise for pursuing molecular spintronics, utilizing the spin degree of freedom on the molecular scale, is because voltage control of the magnetic states enables nonvolatile device elements whose state may be switched without large current densities and power consumption. The successes with device fabrication from molecular magnets, combined with indications of success in manipulating the spin state by electrical means, suggest new approaches leading to developing novel molecular spintronics (a situation reviewed in [1]). Molecular systems have the very real possibility of providing devices that operate at room temperature, on a size scale less than 10 nm, while delivering low power GHz nonvolatile local magneto-electric logic or memory operations. As the expectations for novel printable molecular electronics grows, the design of flexible and high-density nonvolatile memory devices remains a not only hot topic, but a possible route to low power inexpensive flexible electronics.

The spin crossover (SCO) phenomenon, which relates to the transition between a low spin (LS) state of the metal ion to a high-spin (HS) state in 3d transition metal compounds, has potential applicability in molecular spintronic devices. While this is only one class of molecular magnets, the spin crossover molecules are the center piece of several of the articles highlighted in this special issue. There are several possible reasons for the increased interest in this material class. One attraction of the spin crossover molecules is the possibility of manipulating the switching progress around room temperature. Other reasons that these molecules garner attention are the multiple possibilities to switch and to read the different spin states, which makes those molecules especially attractive for device engineering. To understand the principles for achieving a practical device, using spin crossover molecular systems, it worthwhile reiterating the 9 requirements that need to be met (as partly discussed in [1]). (1) To be practical, the devices need to be amenable to manufacturing techniques, and this requires that the molecular devices can be fabricated as thin films. (2) A spin transition between different spin states is needed. (3) The ability to “lock” a molecule in a particular state and (4) isothermally “unlock” and switch the spin state at various temperatures, ideally room temperature. 5) The “write” operation needs to be accompanied by a read mechanism; this means that there is a property where the spin state is easily “sensed”, for example the spin state change is accompanied by a change in conductance. Conductance changes of the spin crossover molecular systems are well known, but if this is to be exploited in a device, then this should be for molecular films on a dielectric substrate, so that it is the spin crossover molecular system that dominates the device performance. (6) For practical operation, the device should be able to function at room temperature. (7) The “on” state resistance should be as low as

possible so as not to contribute the device delay time. (8) The voltage control of state switching needs to survive a million million switches, preferably a million billion times, without degradation. (9) the read mechanism for the "on" state should be significantly different from the "off" state.

To meet the challenges of implementing spin crossover and magnetic molecules into devices, investigators are looking at the manipulation of the ligands to both alter the "on" state conductivity and improve the on/off ratio [2-7]. Clearly a better understanding of the transport properties across molecular magnetic thin films is essential [6, 8]. Manipulation of the conductance, though an applied voltage [1,6,8] is likely key to device implementation. To realize a molecular device, contacts are necessary to any device, so investigations of the molecule-substrate interface are also essential (as in [4,9]). The ability to "lock" a molecule in a particular state can be influenced by intermolecular interactions (as reported in [3,10-12]). Some potential magnetic molecule devices depend on magnetic anisotropy [1], but the topic is of fundamental interest as well [13], as are cooperative effects [2,10-12,14]. Finally, yet importantly, there is the challenge to synthesize new switchable molecules for surface deposition, that show spin state switching and related property changes not limited to the bulk [2,3,5-7,10], but also on surfaces [4,9,10].

It is now clear that fabrication of molecular multiferroic devices is indeed possible as the evidence of isothermal electrical switching of spin crossover complexes accumulates. This voltage-controlled switching of the spin state is accompanied by a conductance change and can be engineered to be nonvolatile. But large local moment molecular magnets also offer a possible route to quantum computing [15,16], as a replacement to quantum dot schemes. Is it possible that the hyperfine interaction of the large local moment metal center molecular magnet (highlighted in the work of Wysoki and Park [16]) might be exploited ?

While the history of spin state manipulation, in molecular systems, is very long and dates at least as far back as the 1930's [17,18], there is both new and vibrant interest in the fundamentals and the possibilities. We hope you get a taste of the possibilities from the articles contained herein.

Birgit Weber, Bayreuth, Germany

Peter A. Dowben, Lincoln, Nebraska, U.S.A.

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