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Spin and charge correlations in molecule-based materials — Physical properties, chemistry, and materials aspects

Foreword

The phenomenon of magnetism has fascinated humanity for centuries. Historically, the preparation of novel magnetic materials was almost exclusively the domain of solid-state chemists. However, in recent years, an exciting new development was sparked by the idea to use discrete molecular spin carriers and to assemble them into ordered one-, two-, or three-dimensional arrays by exploiting non-covalent interactions, like electrostatic attraction, Lewis acid/base pairing, or hydrogen bonding. The advantages of this 'molecule-to-material' approach are immediately clear: rather than having to follow a metallurgical synthesis protocol, one is permitted to prepare these novel materials from solutions of the constituent building blocks already at ambient temperature. Consequently, highly sophisticated functional groups can be employed, which otherwise would not be compatible with the extreme conditions usually required for classical solid-state syntheses. Moreover, systematic variations of the ligand spheres of the spin carriers (i.e. paramagnetic organometallic complexes or Werner-type coordination compounds), as well as of the bridging units, allow a fine tuning of the materials' properties. However, a major challenge arising from the 'molecule-to-material' approach still remains, i.e. the problem of how to maintain efficient spin−spin coupling between the individual paramagnetic centres in the crystal lattice and thus to bring about correlation effects.

In 1988, a landmark proof-of-principle work was published by J.S. Miller and A.J. Epstein, in which they showed that the charge-transfer salt $[(C_5Me_5)_2Fe]^+[TCNE]^-$ exhibits a ferromagnetic behaviour at low temperatures. Since then, research towards the targeted synthesis of improved molecular magnets has gained tremendous attention.

Because of the inherently interdisciplinary nature of the subject, success in the area of molecular magnetic materials heavily depends on close collaborations between chemists and physicists, experimentalists and theoreticians. Six years ago, the J.W. Goethe University of Frankfurt am Main (Germany) launched a joint research effort to study spin and charge correlations in molecule-based organometallic solids. The initiative, which was financially supported by the German Science Foundation (DFG), joins the forces of 16 research groups from the departments of chemistry, physics, and mineralogy. The main emphasis of our research is placed on interesting many-body effects resulting from strong quantum fluctuations that suppress long-range magnetic order. Of particular interest is the possibility to generate materials with small magnetic exchange coupling constants, weak enough for laboratory fields to drive the systems into novel states of matter near a zero-temperature quantum-phase transition.

Our results were discussed with international colleagues at a conference held from 17th to 19th October 2005 in Königstein / Taunus, near Frankfurt am Main. The present issue of the *Comptes rendus Chimie* contains a collection of the conference contributions in their written form and thus provides a comprehensive overview of the most recent developments in the field of molecule-based magnetic materials and gives a flavour of the diversity of quantum many-body effects that these systems show.

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