

Spin and Charge Correlations in Molecule-Based Materials Physical Properties, Chemistry, and Materials Aspects

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Humanity has been captivated by the magnetic phenomenon for millennia. In the past, solid-state chemists were nearly completely responsible for creating new magnetic materials. However, in recent years, the concept of using discrete molecular spin carriers and assembling them into organised one-, two-, or three-dimensional arrays by utilising non-covalent interactions, such as electrostatic attraction, Lewis acid/base pairing, or hydrogen bonding, has sparked an exciting new development. The benefits of this "molecule-to-material" approach are right away apparent: instead of needing to adhere to a metallurgical synthesis protocol, one is allowed to manufacture these unique materials from solutions of the constituent building blocks that are already at room temperature. As a result, more complex functional groups can be used that would not be possible under the severe conditions required for traditional solid-state syntheses. Additionally, systematic modifications of the bridging units, paramagnetic organometallic complexes, or Werner-type coordination compounds' ligand spheres enable fine-tuning of the materials' characteristics. A significant obstacle to the "molecule-to-material" method, namely the issue of how to maintain effective spinespin coupling between the various paramagnetic centres in the crystal lattice and so produce correlation effects, still exists.

J.S. Miller and A.J. Epstein demonstrated in a seminal proof-of-principle paper published in 1988 that the charge-transfer salt $[(C_3Me_5)_2 Fe][TCNE]$ shows ferromagnetic behaviour at low temperatures. Since then, work on the selective synthesis of better molecular magnets has attracted a lot of attention.

Success in the field of molecular magnetic materials depends significantly on tight cooperations between chemists and physicists, experimenters and theoreticians due to the subject's intrinsic interdisciplinary nature. A collaborative research project to examine spin and charge correlations in molecular-based organometallic solids was started at the J.W. Goethe University of Frankfurt am Main (Germany) six years ago. The project brings together 16 research groups from the chemistry, physics, and mineralogy departments. It was financially supported by the German Science Foundation (DFG). Our research focuses mostly on fascinating many-body events arising from powerful quantum fluctuations that suppress long-range magnetic order. Particularly intriguing is the potential to produce materials with magnetic exchange coupling constants that are modest enough for laboratory fields to induce novel states of matter close to a zero-temperature quantum phase transition.

On October 17–19, 2005, a meeting was organised in Königstein/Taunus, close to Frankfurt am Main, where our findings were reviewed with worldwide colleagues.

The current issue of the *Comptes rendus Chimie* includes a collection of the conference contributions in written form, giving a thorough overview of the most recent advancements in the field of molecule-based magnetic materials and providing a taste of the variety of quantum many-body effects that these systems exhibit.