

Lord of the rings

Magnets built of molecular rings of magnetic ions are fundamental model systems for studying the complex correlations and dynamics of quantum spins at the atomic scale. A new generation of neutron spectrometers can reveal complete four-dimensional maps of the spin correlations in spin rings.

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Molecular magnets are exceptionally beautiful chemical structures, in which magnetic metal ions combine with matching ligands to form rings, chains, horseshoes, clusters or cages of spins. These magnetic units are potential building blocks of future devices for information storage and quantum information processing¹ — for which the anisotropy terms are especially important. Writing in *Nature Physics*, Michael L. Baker and co-workers² now report complete four-dimensional maps of the spin correlations in rings of eight magnetic chromium Cr^{3+} ions. This provides crucial input for the theoretical modelling of these systems, the chemical optimization of their properties and, more fundamentally, leads to a better understanding of magnetism at the atomic scale.

Intelligent chemical design allows direct control of the number of metal ions and their exchange Hamiltonian in molecular structures^{3,4}. Recent synthetic efforts have produced new molecular magnets consisting, for example, of homo- or heterometallic rings with variable size, spin values, anisotropy and exchange interactions. These molecular magnets have been intensely investigated by a number of experimental techniques including neutron scattering. Nanoscale arrays of magnetic ions are also studied using magnetization measurements, optics and local probes such as scanning tunnelling microscopy (STM). Custom-made structures can be assembled by STM and

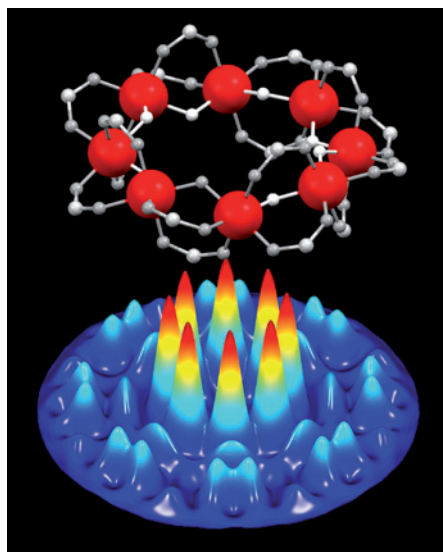


Figure 1 | Schematic of the intensities measured by neutron TOF spectroscopy and the structure of the Cr_8 molecular ring. The momentum dependence of the intensities determine the Fourier components of two-spin correlation functions. In the simplified structure model of the spin ring, the Cr^{3+} ions with a spin of $3/2$ are red. Image courtesy of Michael L. Baker.

their excitations studied by spectroscopy^{5,6}. Whereas STM offers local control of the interaction geometry, other techniques permit direct measurements of the spin states

in the time domain. However, with some of these techniques it is more challenging to determine complex exchange interactions and spin correlations.

Baker *et al.* use neutron time of flight (TOF) spectroscopy to determine the spin excitations in a single-crystal sample of the prototypical Cr_8 antiferromagnetic ring. The energy levels of the ring were already known from previous high-resolution experiments on powder samples^{7,8}, and now the full momentum and energy dependences of three characteristic excitations have been recorded; a weak splitting due to the anisotropy of the lowest level has also been observed.

Experimental neutron intensity maps, such as the one presented in Fig. 1, together with the Cr_8 structure, were used to extract the Fourier components of the two-spin dynamical correlations, which were then compared with model calculations. Such exact calculations are possible for the molecular ring of just eight spins, whereas, due to the size of the Hilbert space, computations for much larger systems are limited by the amount of memory and computing power. The agreement between the experimental data and the model calculations is excellent for all energies and moment transfers. The experimentally determined Fourier components and linear response led to the conclusion that tunnelling of the Néel vector is not responsible for the low-temperature spin dynamics in such a Heisenberg-like

molecular ring, contrary to the case of larger, more anisotropic systems.

At least three important recent developments led to these exciting results. First, chemists are now able to grow large, high-quality single crystals of molecular magnets. Second, modern neutron TOF spectrometers — such as the IN5 spectrometer at the Institut Laue-Langevin used by Baker and co-workers or the pioneering MAPS, MERLIN and LET spectrometers of the ISIS facility at the Rutherford Appleton Laboratory — now have the level of detector coverage, energy and momentum resolution, neutron flux and signal-to-noise ratio to provide unprecedentedly detailed information about spin correlations. Third, sophisticated software has been developed for managing and analysing complex, four-dimensional spectroscopy data. Large arrays of

position-sensitive neutron TOF detectors, such as the one used by Baker *et al.*, have already led to important breakthroughs in a number of fields including studies of superconductors, strongly correlated electron systems, and now molecular magnets.

The model molecular spin ring chosen by Baker and co-workers is well defined — that is, the Hamiltonian has been determined and all quantities can be calculated. The cross-section for neutron inelastic scattering is known exactly. Tremendous progress has been made in neutron TOF spectroscopy in the past decade. These points underlie the demonstration of the potential of the experimental approach used and its application to more general studies of molecular magnets. The study by Baker and co-workers is therefore a benchmark and an important step towards the understanding of the complex spin correlations in molecular

magnets. Rings with an odd number of homo- or heterometallic spins or with frustration, Fe₁₈ as a candidate for Néel-vector tunnelling, and entanglement control by a magnetic field are potentially exciting future directions of study. □

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