Quantum oscillations of the total spin in a heterometallic antiferromagnetic ring

T. Guidi
HMI, Berlin, Germany

S. Carretta, P. Santini and G. Amoretti
University of Parma, Italy

R. Caciuffo
EC, JRC-ITU, Karlsruhe, Germany

A. Hiess, ILL

J.R.D. Copley and Y. Qiu
NIST Center for Neutron Research, USA

G. Timco and R.E.P. Winpenny
University of Manchester, UK

ABSTRACT

The identification of systems displaying quantum coherence at the mesoscopic scale is of great interest, both from a fundamental scientific perspective, and in the context of potential technological applications in the field of quantum computation. Antiferromagnetic molecular rings are very interesting in this respect. Using inelastic neutron scattering (INS), with applied magnetic fields B up to 15 T, we have studied the spin dynamics of an important member of this class of materials, Cr_7Ni. We demonstrate that several avoided crossings (ACs), involving states with different total-spin quantum numbers, occur with increasing B. This corresponds physically to quantum oscillations of the total spin of the ring.

Antiferromagnetic (AF) molecular rings are cyclic clusters with dominant nearest neighbour AF coupling. When organised in a crystal lattice, they provide an ensemble of identical, almost non-interacting magnetic units [1] and represent very promising systems for the observation of quantum coherent phenomena.
Among the wide class of AF rings synthesised so far, the Cr_7Ni heterometallic ring has been identified as a model system for the implementation of qubits [2]. This cyclic cluster has a S = 1/2 doublet ground state, resulting from the dominant AF exchange interaction between seven Cr(III) ions (s = 3/2) and one Ni(II) ion (s = 1) (figure 1).
A doublet ground state, well separated from the first excited level, is one of the prerequisites for a system to be suitable for encoding a qubit. Another crucial condition is the persistence of quantum coherence during the time needed for the elementary computational operations to be performed. It is thus important to know in great detail the quantum spin dynamics of these systems to better understand to what extent their properties match the desired ones.
The application of an external magnetic field B enables the energies and the composition of the eigenstates from the ring's Hamiltonian to be tuned. In particular, in Cr_7Ni several "avoided crossings" (AC) between different spin states are induced by a field with appropriate value and direction. When AC conditions are met, two different spin states are superimposed, and the total spin of each ring oscillates in time between S and S+1 [3].
Using the Disk Chopper Spectrometer at NIST, we have observed this phenomenon around an AC that involves the spin ground state [3].
The three-axis spectrometer IN14 was then used to study the spin dynamics at AC conditions involving excited spin states, and to investigate the effects of the AC on the composition of the spin wavefunctions. For this experiment, we used a 0.4 g single crystal sealed in a preservative HF/MeCN(1:2) atmosphere within an aluminum sample holder. A 15 Tesla vertical-field cryomagnet was used to apply a magnetic field parallel to the ring plane (figure 1). With fixed final neutron wave vector k_F = 1.15 $Å^{-1}$, and a base temperature of 1.5 K, we collected inelastic constant Q scans, at different magnetic fields around the

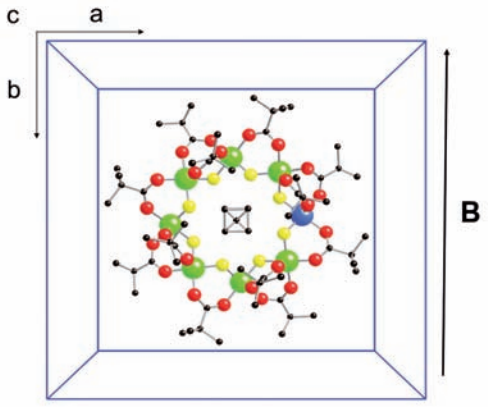


Figure 1. Schematic view of the Cr_7Ni molecular structure in the unit cell (view direction parallel to the crystallographic c axis). Cr: green spheres, F: yellow, O: red, C: black; Ni, blue, disordered over the 8 sites of the ring. H atoms are omitted for clarity. The experiment on IN14 was performed with the magnetic field lying in the plane of the ring.

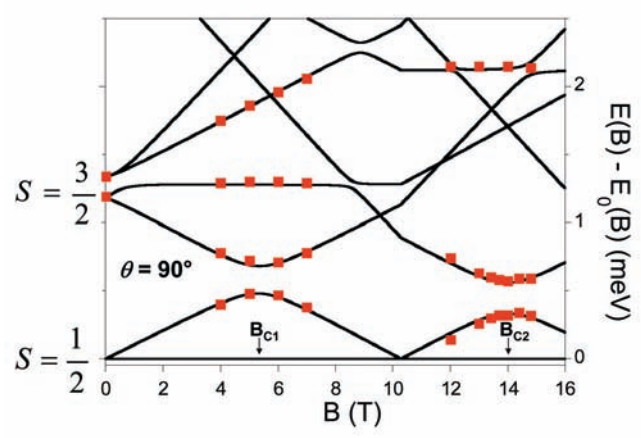


Figure 2. Calculated field-dependence of the low-lying energy levels of Cr_7Ni, relative to the ground-state energy, for an angle θ=90° between the applied magnetic field and the anisotropy axis c. Points indicate the positions of the observed INS peaks. AC conditions occur in correspondence of magnetic field amplitudes B_{C1} and B_{C2} (arrows).

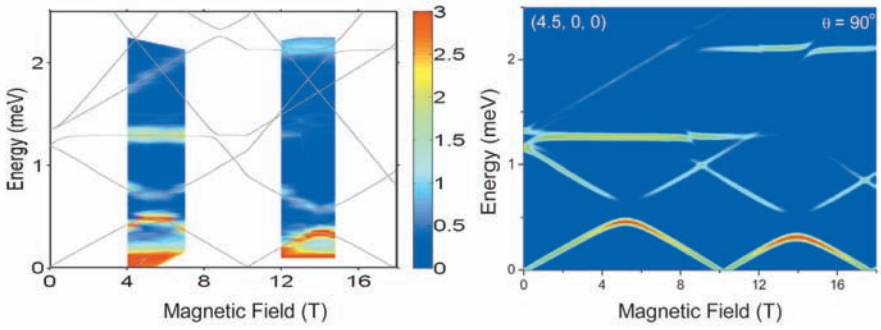
ACs. The modulus of Q was chosen close to the value corresponding to the maximum of the magnetic intensity as a function of Q [4].

In figure 2 the energies of the observed excitations are compared against the calculated energy level scheme as a function of the magnetic field. The occurrence of ACs between the first and second excited spin states at B_{C1}=5.3T and B_{C2}=14T is evidenced by the field dependence of the observed excitations and by the very good agreement with the theoretical calculations.

The spectrum at 0 T in figure 3b shows the transition from the S = 1/2 ground state to the S = 3/2 first excited state, split by anisotropic spin-spin-interactions and uniaxial local crystal fields. Increasing the field, we were able to follow the Zeeman shift of the low temperature excitations in the field interval corresponding to the AC fields B_{C1} (figure 3b) and B_{C2} (figure 3a).

The mixing of different spin states at the AC condition also affects the intensities of the observed excitations. Figure 4 shows a plot of the INS cross section as a function of magnetic field and energy transfer. For magnetic fields approaching B_{C1} and B_{C2}, a redistribution of the INS intensity is observed, providing additional insight into the readjustment of the spin wavefunctions in the vicinity of the AC condition.

In conclusion, high field INS measurements on a Cr_7Ni single crystal have demonstrated the occurrence of avoided crossings involving states with different total-spin quantum numbers. The INS cross section shows an enhancement of the effect of the superposition of different states at the AC condition,



where quantum oscillations of the total spin of the molecule take place. The very good agreement for the positions and intensities of the observed excitations with calculations gives us confidence in the validity of the model spin Hamiltonian and permits further speculations and predictions as to the feasibility of quantum information processing using this class of molecules.

Figure 3. Constant Q scans for Q=(4.5, 0, 0) r.l.u. (reciprocal lattice units), varying the incident energy (fixed k_F=1.15 $Å^{-1}$) at different magnetic fields near the AC conditions (T=1.5 K). The data are vertically offset for clarity. The modulus of Q was chosen close to the value where the magnetic intensity has its maximum [4].

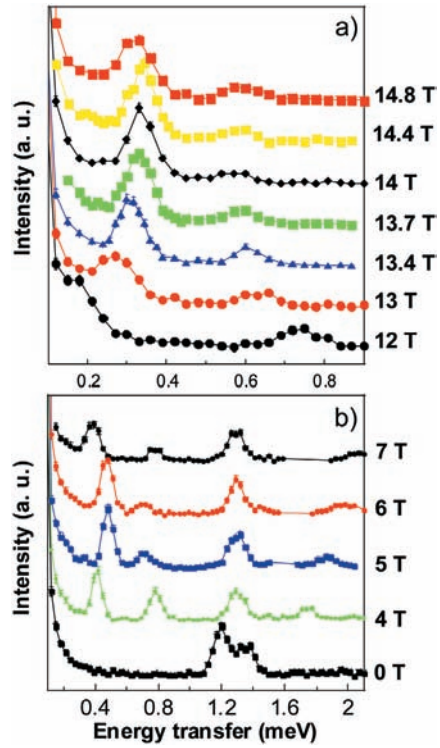


Figure 4. Intensity plot showing energy and field dependence of the measured (left) and calculated (right) INS cross sections for θ=90° and Q=(4.5, 0, 0) r.l.u.; T=1.5 K.

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