Neutron scattering group

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# Energy levels of the giant Keplerate magnetic molecule $\{Mo_{72}Fe_{30}\}$: Inelastic neutron scattering and theory

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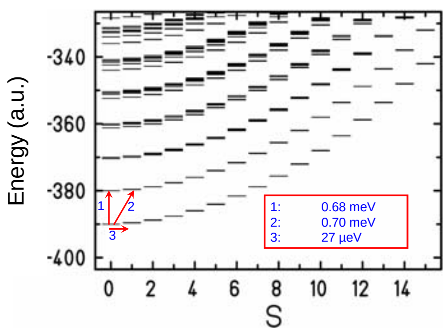
## Background and Motivation

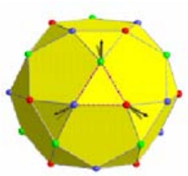
Materials consisting of magnetic molecules are ideal prototypes for the study of fundamental problems in magnetism at the nanoscale level. This work was undertaken to test theoretical predictions of Luban and co-workers on a solvable three-sublattice approximation of a 30 spin ($s$ = 5/2) system interacting antiferromagentically via a Heisenberg exchange interaction. The 30 spins are located at the vertices of an icosidodecahedron, shown below.

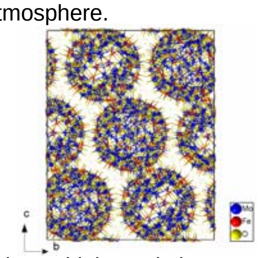



In the approximation of three sublattices (each designated a color), the theory predicts that a sequence of "rotational bands" with an energy gap of E = 5J (J = 1.57 K, is the exchange coupling , E = 0.67 meV) separating the lowest two bands. The theory also gives detailed predictions for the Zeeman shift of the energy levels in a magnetic field.

## Materials and Approach

The deuterated $\{Mo_{72}Fe_{30}\}$ polyoxomolybdate cluster was isolated as $[Mo_{72}Fe_{30}O_{252}(CD_3COO)_{12}\{Mo_2O_7(D_2O)\}_2\{D_2Mo_2O_8(D_2O)\}(D_2O)_{91}] \cdot$ *ca.* 140 $D_2O$, (space group *R -3* (no. 148) Unit cell dimensions: *a* = 55.1306(14) Å *c* = 60.1932(22) Å; Cell volume: 158439.52(812) Å$^3$), and handled under helium atmosphere.





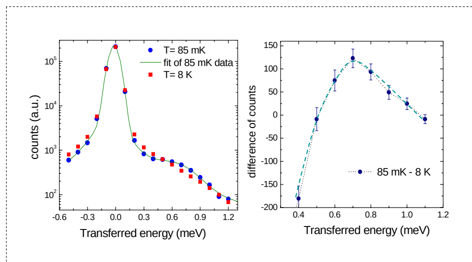
Inelastic neutron scattering at high resolution spectrometers (0.02 – 0.2 meV) were carried out to determine the magnetic excitations. Neutron scattering measurements were performed on three different types of instruments:

- RITA2, a cold-neutron triple-axis spectrometer at PSI (Switzerland), using a fixed final neutron energy $E_f$ =3.7meV.
- Disc Chopper Spectrometer, at NIST Center for Neutron Research, a time of flight technique with a fixed initial energy $E_i$ =2.272 meV.
- OSIRIS time of flight spectrometer, at the ISIS facility (UK), using a fixed final neutron energy $E_f$ =1.845 meV.

The spectra are collected at very low temperatures (60 – 200 mK) using dilution refrigerators and compared with those at higher temperatures (8 – 30 K). The effect of external magnetic field is also explored.

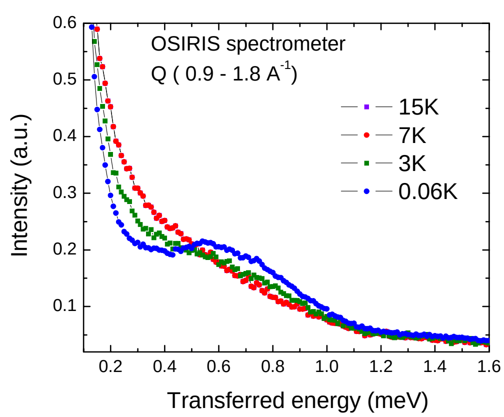
## Results and Discussion

### A. Inelastic Measurements at Zero Magnetic Field



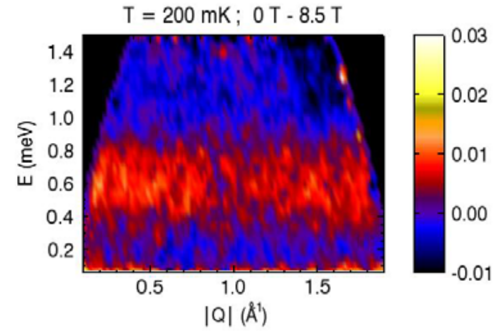

Data collected on RITA II instrument at 85mK indicate an excitation at approximately 0.63meV. Similar results were obtained using the Disc Chopper Spectrometer (NIST) and OSIRIS spectrometer (ISIS). These observations are consistent with theoretical predictions of a ~0.67meV energy gap between the lowest two rotational bands. Inelastic scans performed at PSI at T=85mK and T=8K, Q=1A$^{-1}$ and the difference of these two scans are shown.

### Temperature dependence

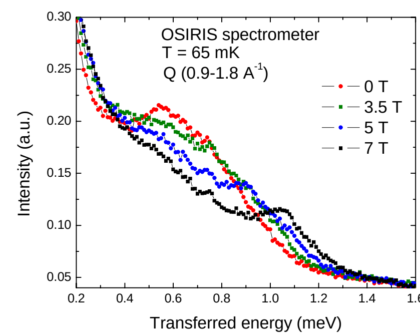



Energy cuts of slices collected at OSIRIS (ISIS) at zero magnetic field at various temperatures down to 60mK show an excitation at 0.65 meV consistent with the theoretical predictions, and with the measurements at NIST and at PSI.

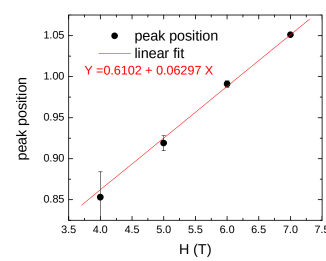
### B. Inelastic Measurements under Magnetic Field




Energy versus momentum transfer slice of the difference between data collected at zero magnetic field and in 8.5 Tesla. (DCS-NIST experiment), at T=200mK. The red/orange band shows the excitation.




Cuts along energy from data collected on OSIRIS spectrometer (ISIS) at T=60mK and various magnetic fields up to 7 Tesla.




Linear behavior of excitation energy that appears when magnetic field is applied. This behavior is predicted by the theory.

## Conclusions and Future Work

- Magnetic excitations determined by inelastic neutron scattering are consistent with theory for the Fe30 Keplerate.
- Magnetic field dependence of the excitations is still a challenge
- Experiments on a variety of novel magnetic molecules are planned for the near future.

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