Neutron Scattering Studies in the Quantum Magnet $CuMoO_4$

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$CuMoO_4$ is a magnetic insulator made up of a complicated network of quantum s = 1/2 magnetic moments residing at the Cu^{2+} site. The Mo^{6+} site is non-magnetic. At room temperature and below, this material exhibits [1] two triclinic phases known as alpha and gamma $CuMoO_4$. At ambient pressure, there is a very strong 1st order structural phase transition between these two structures at about $T_c \sim 190$ K. Both the alpha (high-T) and gamma phases (low-T) phases are triclinic, but the structural phase transition between the two shows a remarkable 13% volume change. This phase change is accompanied by a change in colour of the material from green (alpha) to red-ish brown (gamma). For this reason, this material is referred to as displaying piezo or thermal chromism, and is of considerable current interest.

We are primarily interested in this material for its low temperature magnetic properties, related to networks of interacting s = 1/2 Cu^{2+} moments. Asano *et al.* has conducted [2] high field magnetization studies on this material, and has observed very interesting 1/3 magnetization plateaus occurring at about 9 T (see figure 1). This low temperature, high field magnetization behaviour can be modeled based on networks of six s = 1/2 moments, which arrange to form two dimers and two free spins (shown in the inset of figure 1). However, variation in the form of the M vs. H curves as a function of temperature suggest that at least a subset of the networks of s = 1/2 moments may enter a magnetically ordered state at temperatures below 4 K.

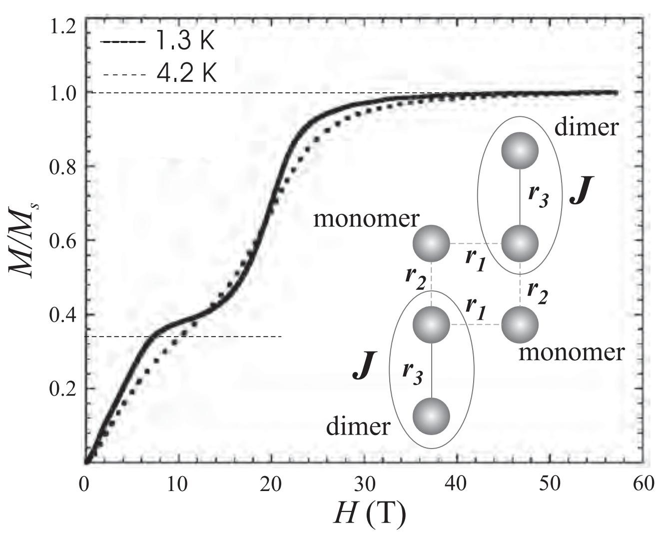

Fig 1. Magnetization plateaus at the 1/3 of saturation magnetization observed in CuMoO4 by high field magnetometry [2]. The plateaus can be modeled on the basis of a network of six spin-½ moments, organized into two dimers and two free spins shown in the inset.

In order to investigate the magnetic behaviour of the low temperature ground state of the complex $CuMoO_4$, we carried out elastic and inelastic neutron scattering measurements on polycrystalline samples as a function of temperature and magnetic field on C5 Spectrometer. We used a vertically focusing PG-002 monochromator and a flat PG-002 analyzer with a fixed final energy of $E_f = 3.52$ THz. Two PG filters were used in the scattered side to eliminate higher order wavelength neutrons from the beam. A nitrogen cooled sapphire filter was used in the main beam to minimize the fast neutron background. For both experiments we used a collimation setting of [none, 0.48°, 0.56°, 1.2°] achieving an energy resolution of ~ 1meV.

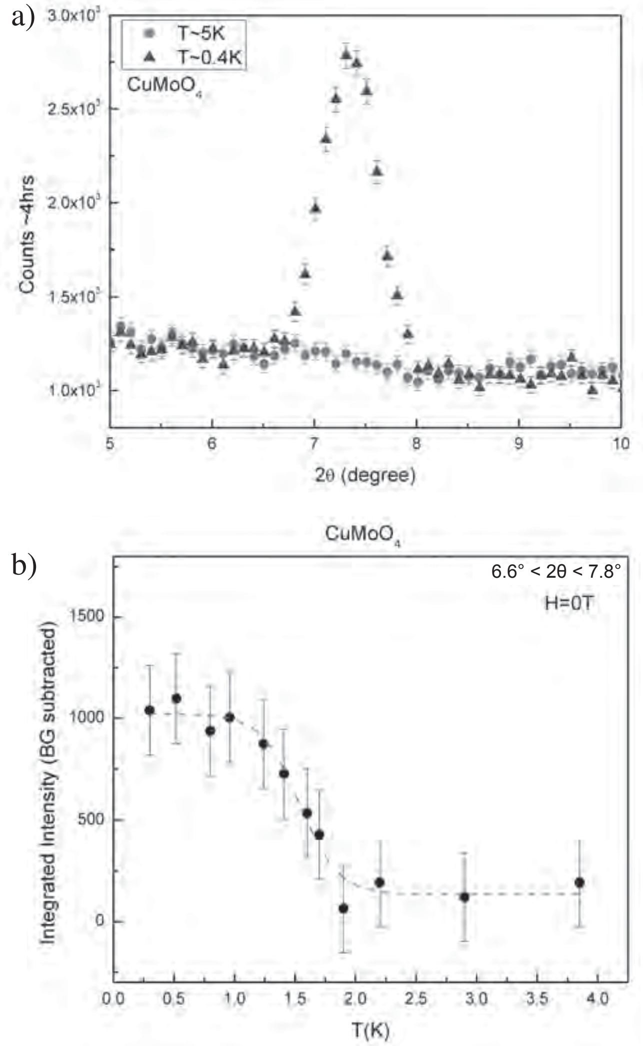

Fig 2. Elastic neutron scattering results show that the low temperature ground state of this quantum magnet is magnetically ordered. (a) The observed intensity as a function of scattering angle at 0.4 K compared to 5 K. (b) The order parameter (integrated intensity of the neutron scattering observed at 6.6° < 2θ < 7.8°) as a function of temperature.

Our elastic neutron scattering measurements clearly show that a peak at the scattering angle of 2θ ~ 7.2° appears below ~ 1.7 K on cooling (see figure 2a). From the observed scattering angle (2θ value) of this magnetic Bragg peak and the incident neutron wavelength (λ = 2.37 Å), we propose that the free spins of neighbouring sites order antiferromagnetically with the wavevector Q ~ 0.3 $Å^{-1}$. Based on these results, we conclude that the low temperature ground state of this quantum magnet is indeed magnetically ordered with the two free spins in the unit cell ferromagnetically coupled to each other while antiferromagnetically coupled to the ones in the neighbouring sites. In addition, the temperature dependence of this peak shown in figure 2b clearly resembles the magnetic phase transition that was observed with specific heat measurements [2], see the zero-field curve in figure 3a. Also our study of the field dependence of this peak is in full agreement with the observed field dependence of the specific heat across the transition [2] as shown in figures 3a and 3b.

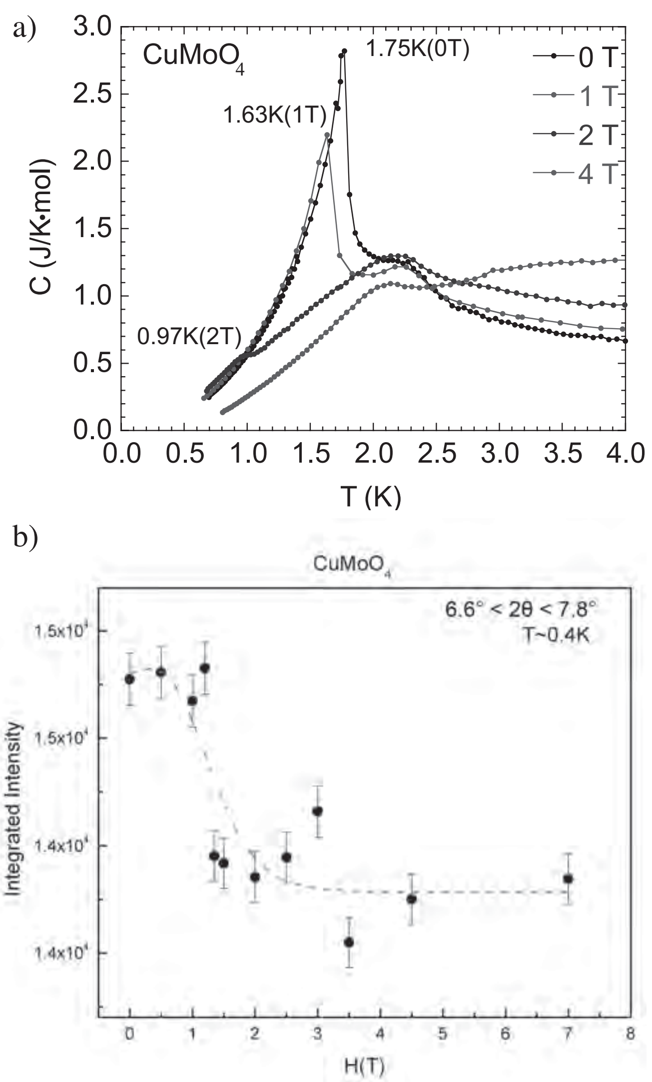

Fig 3. *(a) Temperature dependence of the specific heat observed in zero-field as well as in magnetic fields up to 4 T. From Ref. [2]. (b) The order parameter (integrated intensity of the neutron scattering observed at 6.6° < 2θ < 7.8°) as a function of magnetic field. As seen the field dependence of the peak intensity that we observe clearly follows that of the specific heat.*

In order to study the singlet-triplet excitation spectrum associated with the s = 1/2 dimers, we also carried out inelastic neutron scattering measurements on this polycrystalline material at C5 spectrometer. The results of these measurements as a function of temperature and magnetic field are shown in figures 4-6. As shown in figure 4, in the ordered state and in zero magnetic field, we observe a broad peak at an energy transfer of ~ 3 meV which is about the same size as the expected singlet-triplet splitting of ~ 26 K proposed [2] based on the high field magnetization studies. The temperature and Q-dependence of this ~ 3 meV excitation confirm the magnetic nature of this excitation. As shown in figure 5, with the application of a magnetic field in the range of zero to 7.5 T, the 3 meV peak broadens and an additional field-dependent feature develops at a lower energy of ~ 1.75 meV.

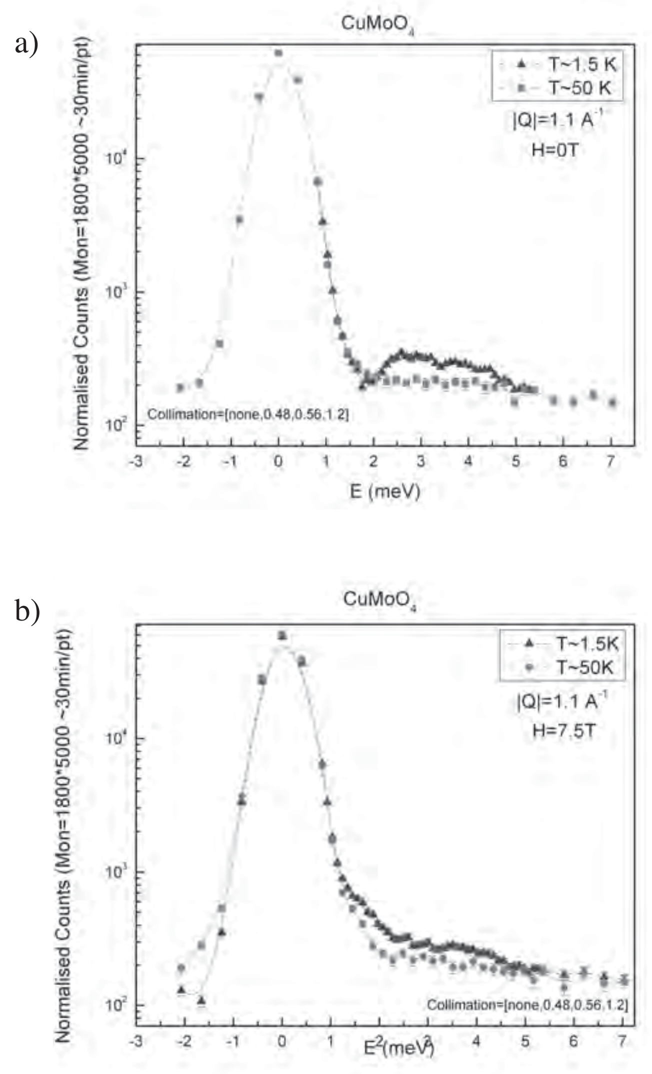

Fig 4. *Inelastic neutron scattering measurements were carried out at the C5 triple-axis spectrometer to study the singlet-triplet excitation spectrum related to the s = 1/2 dimers and thereby to confirm their existence. (a) In the ordered state and in zero magnetic field, we observe a broad peak at an energy transfer of ~ 3 meV. (b) At the largest applied field of 7.5 T, we observe a splitting of the broad ~ 3 meV excitation. A better energy resolution is needed to resolve the structure in the excitation peak in presence of the magnetic field.*

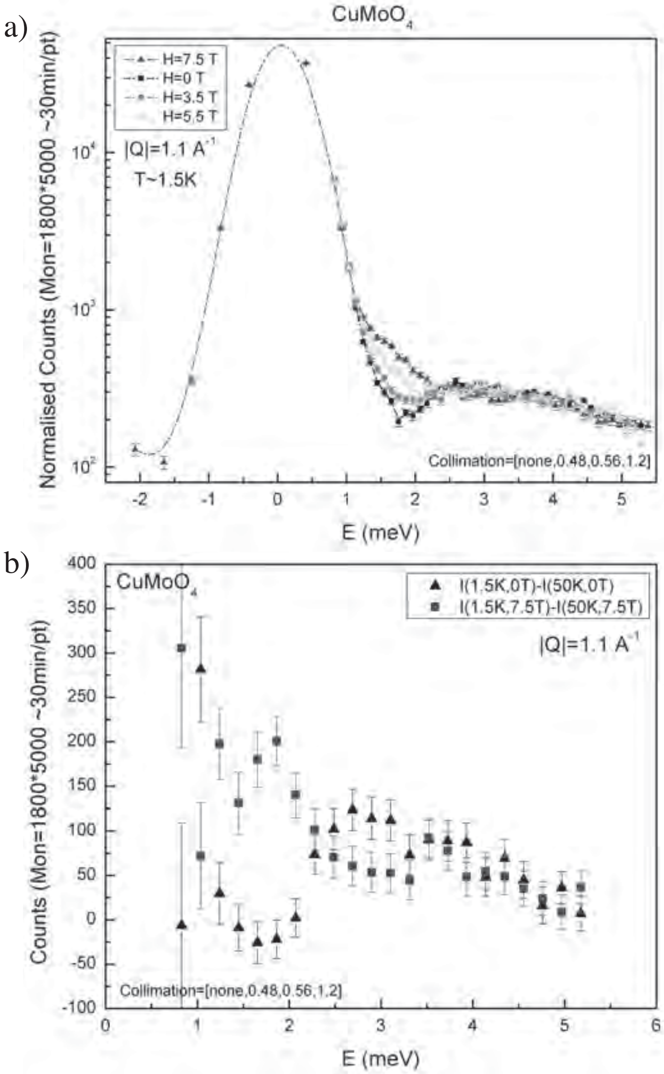

Fig 5. *(a) With the application of a magnetic field in the range of 0 T to 7.5 T, the 3 meV peak broadens and an additional field-dependent feature develops at a lower energy of ~ 1.75 meV. (b) High temperature background subtracted intensity at zero field and at an applied field of 7.5 T. The appearance of the additional feature is clearly seen here.*

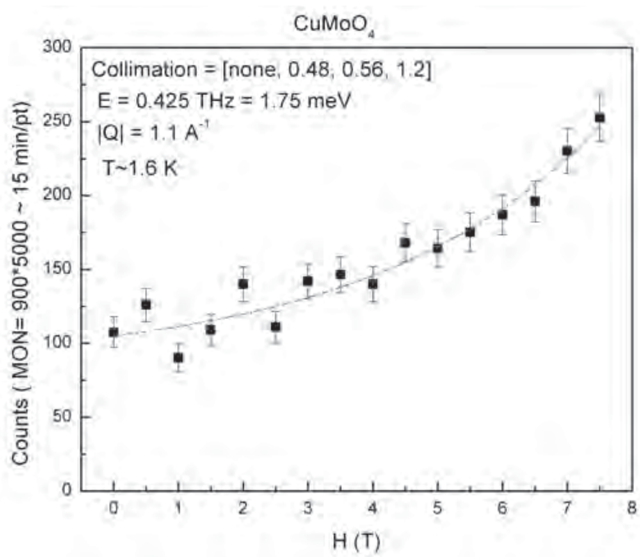

Fig 6. *The spectral weight at 1.75 meV as a function of magnetic field. As seen here and from figures 4 and 5, a clear but complicated field dependence is observed, an indication of the complexity of the ground state in this system.*

Our study confirms that the system undergoes a magnetic phase transition to a low temperature ground state below 1.7 K. In this ordered state the free spins in the unit cell are ferromagnetically coupled to each other while antiferromagnetically coupled to the ones in the neighbouring site. In addition our study confirmed the presence of the singlet-triplet excitations in the system. However the field dependence of this excitation appears to be complex. This is probably due to the fact that in addition to the magnetic dimers in the system there also two free spins in the unit cell. Further inelastic experiments in a magnetic field with better energy resolution as well as further theoretical work is required to fully understand the low temperature ground state of this complicated system.

References

[1] M. Wiesmann *et al.*, J. Solid State Chem., 132, 88 (1997).
[2] T. Asano et al., private communication.