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Inelastic neutron scattering study of the spin-gap behavior in $Cu_2Te_2O_5Br_2$

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Abstract

We present direct observational evidence of a quasi-spin-gapped phase in the spin-tetrahedral system $Cu_2Te_2O_5Br_2$ using inelastic neutron scattering. Our data show a magnetic peak centred at 4.20 ± 0.05 meV, which falls in intensity with increasing temperature in consistence with a thermal depopulation of spin from a non-magnetic ground state to an excited state. However, the magnetic peak also shows a dispersive character, which is associated with the magnetic order in this system.

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The quantum spin system $Cu_2Te_2O_5Br_2$ shows intriguing ground state properties, and has received considerable attention in recent years. It consists of weakly interacting tetrahedra of Cu^{2+} ions, which are believed to order at low temperatures while co-existing with a spin-gapped phase [1]. Magnetization measurements [2] show a large drop in the magnetization of $Cu_2Te_2O_5Br_2$ at low temperatures, which is attributed to a singlet–triplet gap in the magnetic excitation spectrum. Johnsson et al. modelled the system as isolated spin-tetrahedra, which gives a good fit to the magnetization data, resulting in a singlet–triplet gap energy ~43 K. More recent studies [3–7] have investigated the possibility of different inter-tetrahedral exchange paths and the relative strength of their coupling. In addition, an anomaly is observed in the heat capacity at $T_N^{Br} \sim 11$ K [1], but the nature of the transition is unclear [8].

We report the direct observation of a quasi-spin-gap in the excitation spectrum of $Cu_2Te_2O_5Br_2$ using inelastic neutron scattering (INS). With this

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technique we are able to probe the momentum dependence of the magnetic excitations as well as their magnitude. We have performed the measurements using the HET time-of-flight spectrometer at the ISIS neutron facility, Rutherford Appleton Laboratory, UK. A polycrystalline sample of $Cu_2Te_2O_5Br_2$ was prepared according to the method described by Johnsson et al. [2].

Fig. 1 shows a 2D energy transfer ($\hbar\omega$)–momentum transfer ($|Q|$) contour plot for $Cu_2Te_2O_5Br_2$ at 7 K, with an incident energy of $E_i = 18$ meV. The grey scale denotes the scattering intensity, $S(|Q|, \hbar\omega)$, which here is the powder average of the spin–spin correlation function. The data show a band of high intensity centred at ~5 meV, which extends to lower energies in the momentum transfer region around ~0.7 Å^{-1}. The peak shows a clear drop in scattering intensity with increasing $|Q|$, indicating that it is a magnetic excitation. Indeed, it falls far more quickly than the Cu^{2+} magnetic form factor [9] due, presumably, to the structure factor of a spin-cluster arrangement. One would also expect S to drop to zero as $|Q| \to 0$ for a spin-cluster, suggesting that a peak in $S(|Q|)$ occurs below $|Q| = 0.4$ Å^{-1}, which is the lowest value we are able to measure in the energy region of interest. In addition, a narrow, dispersive-looking vertical band of intensity is observed for $|Q| \sim 0.7$ Å^{-1}, extending downwards from the excitation at 4.2 meV towards the elastic peak. The intensity of this band increases with an initial increase of temperature, but gradually falls beyond ~25 K. Therefore, on the one hand, we identify the magnetic peak as an excitation across the 'spin-gap' in the energy spectrum that has previously been predicted by models invoked to explain magnetic susceptibility [2] and Raman scattering [3] measurements. However, we believe that below the ordering temperature in this system, additional magnetic states within the 'gap' allow for the dispersive nature of the excitation observed.

Fig. 2 depicts the energy dependence of the magnetic excitation, taken over a $|Q|$ range of 0.6–0.8 Å^{-1}. The peak is fitted to a double Lorentzian function, giving a best fit width of 3.66 ± 0.09 meV and position of 4.20 ± 0.05 meV. This peak width is noticeably larger than the resolution width of the instrument (~0.5 meV), which may be due to the presence of multiple peaks arising from several closely lying states separated by a gap energy. A broad peak may also be due to dispersion of the excitations in the

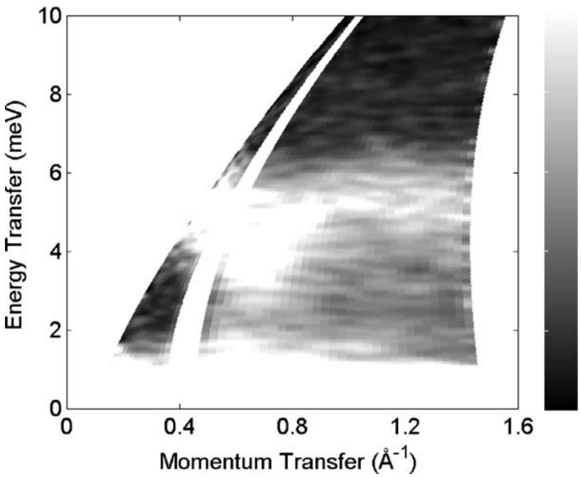

Fig. 1. 2D contour map of the magnetic scattering in $Cu_2Te_2O_5Br_2$ as a function of energy transfer ($\hbar\omega$) and momentum transfer ($|Q|$), obtained at 7 K, with an incident energy of 18 meV. The grey scale denotes the scattering intensity (S), measured in mb sr^{-1} meV^{-1} f.u^{-1}.

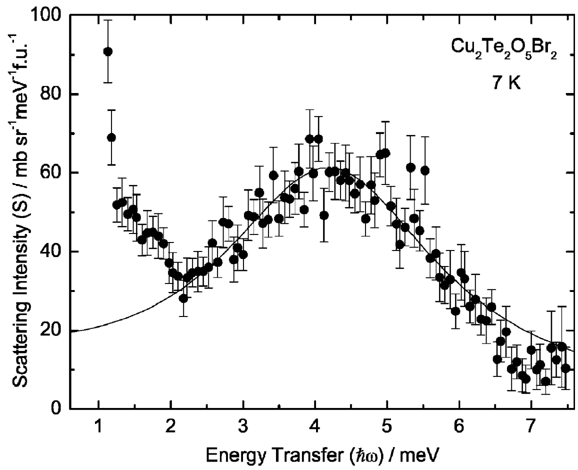

Fig. 2. Scattering intensity (S) versus energy transfer ($\hbar\omega$) summed over the $|Q|$ range 0.6–0.8 Å^{-1} at $T = 7$ K. The solid line represents the least-squares fit of the peak to a double Lorentzian function.

ordered state. The theoretical model of isolated tetrahedra contains two low-lying singlets separated from higher triplets, with further splitting possible when inter-tetrahedral coupling is taken into account. With temperature, the relative population of the possible multiple states would alter, causing a relative change in the intensity of the respective magnetic peaks. Whilst we cannot resolve these individual peaks in our data, we observe a decrease in the intensity of the broad magnetic peak with increasing temperature (not shown) as would be expected for an overall thermal depopulation from a non-magnetic ground state to an excited state. The intensity observed in the 1–2 meV region in Fig. 2 is attributed to the dispersive component of the scattering (centred around $|Q| \sim 0.7\,\text{Å}^{-1}$) discussed previously.

In conclusion, our neutron scattering investigation of $Cu_2Te_2O_5Br_2$ reveals a magnetic excitation with magnitude ~4.2 meV, providing evidence of at least one, possibly multiple, energy gap(s) in the spin excitation spectrum. In addition, a dispersive component to the excitation is observed in connection with the magnetic ordering in this system.

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