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FIELD-INDUCED MAGNETIC PHASE TRANSITION IN A SINGLET GROUND STATE DIMER SYSTEM

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Abstract. – AC-susceptibility measurements on the singlet-ground state system Cs₃Cr₂Br₉ are extended to very low temperatures (46 mK) and high applied magnetic fields (12 T). The resulting phase diagram is shown to exhibit features at variance with recent soft-mode and molecular-field models for this system.

The conditions under which singlet ground state systems order magnetically is a topic of continuing interest. Compounds typified by Cs₃Cr₂Br₉ form a class of materials where trivalent transition metal ions couple strongly in pairs by exchange interactions to form magnetic dimers of high symmetry (D₃h). The Cr-Cr separation is 3.3 Å within the dimers and 7.4 Å (same sublattice), 7.5 Å (different sublattices) between nearest neighbour dimers. Antiferromagnetic coupling between ground Cr³⁺ ions leads to a ground manifold for the dimer consisting of singlet, triplet, quintet and septet levels at 0, 6.3, 17.5 and 31.0 cm⁻¹ respectively [1]. Previous experimental work on this material may be broadly divided into the two main themes of optical [1-3] and neutron scattering measurements [4, 5].

Optical studies have centred on intra-dimer interactions, in particular exchange interactions in those excited dimer levels where the normal Heisenberg-Dirac-Van Vleck hamiltonian is not appropriate. In general these studies have explained most of the features of the absorption, Zeeman and MCD spectra by considering the dimer as an isolated unit. Inelastic neutron scattering measurements have studied the ground manifold and the effect of interdimer interactions. This has demonstrated that the singlet-triplet excitations exhibit significant dispersion and although no evidence for magnetic order has been observed down to 1.5 K, the dispersion suggests that a transition to magnetic order might be induced by a magnetic field. Attempts to observe ordering effects in field by neutron measurements have proved inconclusive [5]. That such a transition does exist has recently been shown by an AC-susceptibility study over a restricted range of temperature (1.5-1.9 K) and field (0-5 T) [6]. From the limited nature of the data, it has not proved possible from these measurements to test the models suggested to describe the transition to order or to distinguish between models.

In this paper, we present an extension of AC-susceptibility measurements to much lower temperature (down to 46 mK) and to higher applied fields (up to 12 T) in an effort to map out the magnetic phase diagram. Experiments were carried out on polycrystalline samples mounted under helium gas. For lower temperature readings, fields up to 10 T were applied to the sample cooled in a helium dilution refrigerator. For higher temperatures, a separate apparatus equipped with a 12 T magnet was used. The form of the observed transition in the susceptibility as a function of field is shown in figure 1 at several temperatures. For temperatures above 1.3 K, the transition exhibits hysteresis between up and down fields which becomes more marked with increasing temperature. In addition the character of the transition becomes more difficult to ascertain. The transition was therefore followed into the higher temperature regime by using a slow temperature sweep at fixed field. The resulting phase diagram is shown in figure 2. The increase in the uncertainty of the transition position with increasing temperature may be related to the small anisotropy reported by Leuenberger et al. [6] in orientated single crystal measurements.

Fig. 1. – The measured AC-susceptibility of polycrystalline Cs₃Cr₂Br₉ as a function of applied magnetic field for various temperatures.

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Several theoretical models have been suggested to predict the phase diagram in this system. One is a soft-mode theory in the random phase approximation which has been used to interpret the singlet-triplet excitations measured by inelastic neutron scattering [5] and has been extended to include applied magnetic field [9]. From zero-field results, this theory predicts that a component of the acoustic branch of the singlet-triplet excitation near the K point will be driven towards zero energy by applied magnetic field giving rise to an ordered phase at $T = 0$ between fields of 2.5 T and 15.5 T. For higher temperatures the field range of the ordered phase would diminish and no order should be observed above a temperature of 3 K.

The predicted behaviour of the soft-mode model is shown in figure 2 for the range of temperature and field of this experiment. An alternative approach involves a static molecular field approximation in which the two lowest levels of the dimer are treated as a pseudospin of $S = 1/2$ [7, 8]. The resulting phase diagram is qualitatively similar to that given by the soft-mode theory, but for reasonable parameter values the ordered region is expected to be smaller in temperature and field range.

It is clear from figure 2 that, although the predicted qualitative trends are present, detailed features of the soft-mode theory, such as the approximate temperature independence of the transition below 1.5 K, are not observed. Also the onset of order at a field of 1.5 T as $T \to 0$ is much lower than predicted. Further, there is little evidence for a decrease in the region of order at high fields. It has been suggested by Leuenberger [9] from his earlier susceptibility results in the region of 1.5 K that the observed discrepancy between theory and experiment in this temperature range may be accounted for qualitatively by a renormalisation procedure. This has the effect of raising the energy of the soft mode and thus shifting the ordering field to higher field values. Although this may improve the fit in the limited temperature region around 1.5 K, it can be seen from results reported here that it will produce a considerably poorer fit in the very low temperature regime. It would appear that further progress requires a more detailed review of theory, possibly with the inclusion of correlation effects. The deviation from theory at high fields is almost certainly related to the neglect of the quintet level at 17.5 cm$^{-1}$. The lowest component of this level will become appreciably populated at fields above about 8 T and should actually become the ground state of the system at field above 12 T.

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