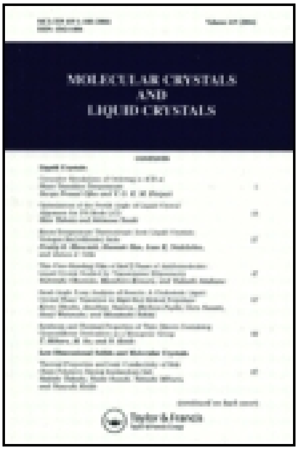

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Magnetic Field Dependent Susceptibility of $(TMTSF)_2ClO_4$

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MAGNETIC FIELD DEPENDENT SUSCEPTIBILITY OF $(TMTSF)_2ClO_4$

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Abstract The magnetic susceptibility (χ) of slowly cooled single crystals of $(TMTSF)_2ClO_4$ was measured for H//c* with fields up to 10 tesla and 1.7K < T < 300K. No anomaly in χ is observed at the anion ordering temperature 24K, with χ^{Pauli} constant for 4.2K < T < 30K. Between 4.2K and 2.2K, χ^{Pauli} increases threefold in aggreement with the high field specific heat data signaling a magnetic field induced phase transition. The magnetic field dependence of the χ at 4.2 and 2.0K shows the presence of ~1500 ppm Curie spins. Such localized spins and their associated Schottky anomaly can account for a large part of the 2 tesla specific heat anomaly reported earlier by Brusetti et al.

I. INTRODUCTION

The Bechgaard organic conducting salts of the form $(TMTSF)_2X$ have attracted a great deal of attention recently due to the rich variety of ground states that are available to the system depending of the anion used, temperature, pressure, and magnetic field.[1] In particular, upon slow cooling $(TMTSF)_2ClO_4$ undergoes an ordering of the anions at 24K and a transition at ambient presssure to a superconducting state for temperatures below 1.2K. A wide range of magnetotransport anomalies have been reported for this material at low temperatures (<4K) and magnetic field parallel to c*. These anomalies have been interpreted in terms of a magnetic field induced phase transition from a metallic, nonmagnetic state to a semimetallic, magnetic state.[2]

Direct probes of the variation of the density of states with magnetic field and temperature are important for detailed understanding of these myriad phenomena. Brusetti et al.[3] have reported the magnetic field dependence of the specific heat in $(TMTSF)_2ClO_4$ between 0.4K and 4K and magnetic field H < 6.3 tesla. Their data showed two key features: (a) At high fields (H ~6 tesla) the electronic contribution increased two fold as the temperature was lowered from 4K to 2K. This term was suggested to herald the onset of a spin density wave state at high fields and low temperature. (b) A local maximum in the electronic specific heat in the vicinity of 2 tesla, varying in magnitude and field with temperature. We have measured the static spin susceptibility, χ^S, of slow cooled $(TMTSF)_2ClO_4$ for H//c* and T > 1.7K. We find an anomalous threefold increase in χ^S as T is lowered from 4K to 2K, in agreement with the high field results of Brusetti et al. However, we also observed a sizeable density of Curie spins whose Shottky anomaly can, in large part, account for the local maximum reported for specific heat for H ~2 tesla.

II. EXPERIMENTAL TECHNIQUES

The temperature and field dependent χ was measured using a calibrated Faraday balance technique.[4] The samples, typically about 3-7 mg, were oriented with H//c* in a gold coated quartz bucket or a 99.999% Al bucket. Reproducibility to better than one percent was achieved.

III. RESULTS

The samples were slow cooled at less than 0.5K/minute through the anion ordering temperature and should be in their relaxed state. The spin susceptibility was obtained by subtraction of the diamagnetic core term -4.2×10^{-4} emu/mole[5] from the experimental data. The general features of $\chi^S(T)$ are in agreement with the earier results of Scott[5] and Miljak[6]. Figure 1 shows the data obtained at low temperatures on an expanded scale. There is no anomaly in the vicinity of the anion ordering temperature. The slow increase in χ^S as T decreases from 60K to 4K is consistent with 600-1000 ppm Curie spins in the sample. The χ^S predicted for 2K based on a constant χ^{Pauli} and 1000 ppm Curie spins is shown circled in Fig. 1. The experimental 7.6 tesla 2K data point is nearly a factor of three larger than predicted. The large increase is in agreement with the earlier specific heat data of Brusetti et al and supports a magnetic field induced transition at these temperatures and fields.

The variation of susceptibility (magnetization/magnetic field) with magnetic field at 2K is shown in Fig. 2. The asterisks are the experimental data. The dashed curves indicate data taken on other runs, extending the range from 1 tesla to 10 tesla. Measurements between 1 and 2 tesla indicate that, to within the resolution of our instrument, the susceptibility is field independent. This is in agreement with Miljak et. al.[6] who reported no change in χ between 0.2 and 0.95 tesla. The apparent susceptibility decreases rapidly above 3 tesla and

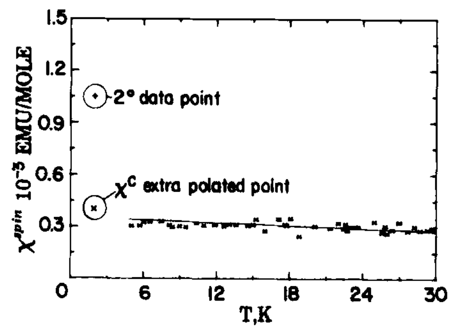

FIGURE 1. χ^S vs. T. The experimental 2K, 7.6 tesla data point ⊕ and the value from extrapolation of T > 4K data ⊗ are shown. See text.

then varies more slowly above 6 tesla. The total reduction in the apparent χ^S at 10 tesla is ~25 per cent. Similar data taken at 4.2K show a much smaller decrease in χ^S with increasing applied magnetic field.

IV. DISCUSSION

The presence of ~1000 ppm Curie spin detected in the 4-60K data and the detailed field dependence of χ^S at 2K support the role of spin 1/2 entities in determing the magnetic response. The solid curve in Fig. 2 is obtained by evaluating the Brillouin function for J = 1/2 at 2K. A very good fit to the field dependent contribution is obtained for ~1500 ppm spins. The predicted field behavior at 4K also accounts for the field dependence observed at that temperature. However, as indicated in Fig. 1, the net spin susceptibility still clearly indicated a transition to a different state between 4K and 2K at 7.6 tesla.

The change in electronic state at constant temperature with increasing magnetic field is the subject of considerable interest. The $\chi^S(H)$ data of Fig. 2 suggest no detectable change in the Pauli susceptibility with increasing field. This is in contradiction with the peak reported[3] for the electronic specific heat at ~3 tesla and 1.7K. However the presence of a sizeable number of Curie spins in our samples, and an even larger number in the earlier samples of Scott[5], suggest a significant contribution to the specific heat from the Schottky anomaly due to isolated spin 1/2's. A moderately good agreement was achieved to the field dependence of specific heat data of Brusetti et al assuming the presence of 4500 ppm Curie spins. The differences between the predicted Schottky anomaly in the specific heat and the reported data may signal a magnetically induced transition. Magnetic field dependent susceptibility and specific heat data on the same samples are necessary for an unambiguous analysis.

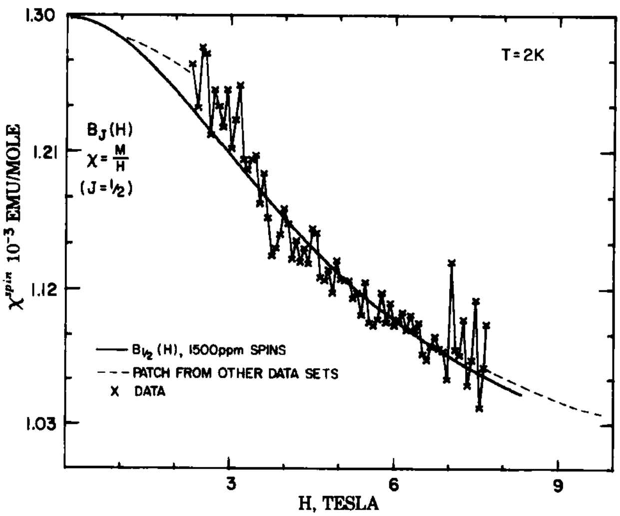

FIGURE 2. χ^S vs. H at 2K. See text.

V. SUMMARY

The magnetic susceptibility measured to high magnetic fields and low temperatures supports a transition to a spin density wave ground state. The increase in the Pauli susceptibility is nearly threefold. A sizeable contribution to the susceptibility from isolated s = 1/2 spins was reported. These spins can account for the observed magnetic field dependence of the susceptibiltiy at 2K and 4K. The presence of ~4500ppm spins can account for a large part of the low field peak reported earlier in the magnetic field dependence of the electronic specific heat.

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