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LOW TEMPERATURE SPECIFIC HEAT OF THE MAGNETIC SUPERCONDUCTOR Gd$_{1.0}$Mo$_6$Se$_8$


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Abstract. - Measurements of the specific heat of the superconductor (T$_c$=5.8 K) Gd$_{1.0}$Mo$_6$Se$_8$ are reported for the temperature range 0.1 K < T < 4.0 K with applied magnetic fields of 0 < H < 18 kOe. The entropy associated with a large peak at 0.75 K for H = 0 indicates that the Gd$^{3+}$ spins become ordered even though the material remains superconducting. The specific heat is strongly affected by an applied field.

INTRODUCTION. - There has been much recent interest in the question of the coexistence of superconductivity and ordered magnetism /1,2,3/. In this paper we report the low temperature specific heat of a compound in which coexistence does occur, Gd$_{1.0}$Mo$_6$Se$_8$ (superconducting transition temperature T$_c$ = 5.8 K). The main result of this work is that the entropy associated with the specific heat indicates that the Gd$^{3+}$ spins become substantially ordered as the temperature is decreased from 1 K to 0.2 K.

EXPERIMENTAL DETAILS. - The method of preparation and sample characteristics were the same as for those used in earlier work /4/. X-ray phase analysis of the sintered pressed pellets at room temperature showed that the only impurity phase present in the otherwise single phase Chevrel structure was ~ 2-3% of SiO$_2$. The specific heat apparatus and techniques used will be described elsewhere. Measurements were made on 82 mg of the sample, which had been crushed and mixed with Apiezon N grease for thermal contact. We estimate absolute errors in the reported specific heat to be less than 15% at 0.1 K and on the order of 5% or less above 0.3 K. Temperature errors are estimated to be 2% or less, and the uncertainty in the magnetic field less than 1%. Measurements of the resistive transition show that the material remains superconducting down to 50 mK.

RESULTS AND DISCUSSION. - The specific heat (C) over the temperature (T) range 0.1 K < T < 4.0 K with applied magnetic fields H = 0, 2.5, 5, 10, and 18 kOe are shown in figure 1. The behaviour above 2 K is detailed in the inset. In all cases, the addenda contribution has been subtracted.

![Figure 1: Specific heat of Gd$_{1.0}$Mo$_6$Se$_8$ as a function of temperature for several values of the external magnetic field.](http://dx.doi.org/10.1051/jphyscol:19786162)

First, consider the result for H = 0. The main features are the rapid drop below 0.4 K, the large peak at 0.75 K, the valley at 1.3 K, and the
small lambda anomaly at 3.4 K. From prior work on this and related compounds /4/ it is known that below 4 K the contribution of phonons and itinerant electrons is negligible. Most of the entropy change below 4 K is associated with the peak in C at 0.75 K. This is seen in figure 2, where C/T and S are plotted as a function of temperature.

Fig. 2: Entropy and C/T of Gd$_{1.4}$Mo$_6$Se$_8$ as a function of temperature.

From figure 2 we estimate that nearly all of the entropy below 2 K belongs to the peak. But at 2 K the observed entropy is 90 % of the entropy expected from complete ordering by the Gd$^{3+}$ (J = 7/2) spins ($S_{\text{max}} = N_A k_b \ln (2J + 1) = 17.3$ J/mole K). Therefore we conclude that the Gd$^{3+}$ spins have become ordered at low temperatures. Furthermore, on the basis of measurements with H > 0 described below and the behaviour of the magnetic susceptibility /4/ of the similar (same T$_c$ /5/, same structure, same features in C above 1.4 K /5/) compound Gd$_{1.3}$Mo$_6$Se$_8$, we conclude that the ordering is antiferromagnetic.

If the data below 0.3 K are graphed on a semilog plot versus $T^{-1}$ (not shown) an energy gap ($\Delta$) behaviour is seen with $\Delta(H = 0) = (0.45 \pm 0.03)$ K. This gap may be an effect of anisotropy on the spin wave spectrum.

Now consider the results for H > 0. Below 0.3 K, the effect is to increase C up to 5 kOe. This means that the tendency to order is suppressed to lower temperatures, as expected of an antiferromagnet in more than one dimension. The effect of H < 5 kOe on the peak at 0.75 K is to smear it out, but not to shift it. At H = 10 kOe and above, the entire effect is to move ordering to higher T. This may mean that the system has gone into the spin-flop state. At 10 and 18 kOe, an energy gap behaviour for excitations is still observed, with $\Delta(10 \text{ kOe}) = (0.45 \pm 0.03)$ K and $\Delta(18 \text{ kOe}) = (1.11 \pm 0.08)$ K. On the basis of our observed effect of H on C, it is clear that the degrees of freedom involved are magnetic, and hence must be the Gd$^{3+}$ spins.

Finally we mention the lambda anomaly, which has been the subject of earlier work /5/. From the inset in figure 2, it is seen that the anomaly rides essentially unchanged on top of a smoother background of different origin. This indicates that it is not related to the magnetism in Gd$_{1.4}$Mo$_6$Se$_8$. Its origin remains a mystery.

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References