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January 1966

HYPERFINE AND ELECTRONIC HEAT CAPACITIES OF AuMn*

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The low temperature heat capacity of a gold-manganese alloy near the composition AuMn has been measured. The hyperfine field at the Mn nuclei is found to be 320 kOe. The electronic heat capacity gives a Fermi level density of states of 0.445/eV-AuMn molecule, which is remarkably low for a transition metal compound with an unfilled d-shell. This is interpreted as showing a negligible contribution of the d-band to the density of states and suggests a $3d^5$ configuration for the Mn ion cores.

Near the composition AuMn the gold-manganese alloy crystallizes in the ordered CsCl structure, transforming martensitically to a tetragonal lattice with the onset of antiferromagnetic ordering at about 200°C.^{1,2} For gold concentrations of greater than 50 at.%, such as in the sample reported on here, the structure has $c/a \sim 0.97$ at the Néel temperature. The manganese moments are aligned parallel in the a - a planes with adjacent planes antiferromagnetically aligned with respect to each other.¹ Torsion measurements by one of us (W.B.) on a twinned single crystal show

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that the moments are directed along an a axis. The symmetry is strictly orthorhombic, however the lattice distortion from tetragonal is extremely small¹ and has not been measured.

The sample used was spark cut from a twinned single crystal which was grown by induction heating in a high purity graphite mold. Manganese of 99.98% purity and gold of 99.999% purity were used in preparing the alloy. The crystal was heat treated for several hours at both 1000°C and 600°C to insure homogenization and complete chemical ordering. The gold concentration of the final sample used was 51.5 at.% as found by chemical analysis. X-ray analysis of the twinned crystal and of annealed powder ground from the same crystal were in agreement with the known structure and lattice constants of the room temperature phase at this concentration. No evidence of chemical disordering of the CsCl structure could be found from X-ray powder patterns.

The heat capacity measurements were made between 0.14 and 1.18°K with an apparatus similar to that previously described.³ The data below 0.8°K fit the equation

$$C(\text{mJ mole}^{-1} \text{ deg}^{-1}) = 1.05T + 3.21T^{-2} - 0.00156T^{-4}. \quad (1)$$

The T and T^{-2} terms were determined by the slope and intercept of a plot of CT^2 versus T^3 for the temperature region between 0.4 and 0.8°K as shown in Fig. 1, and the T^{-4} term was determined as explained below.

Above 0.8°K the lattice heat capacity becomes observable. This contribution is only several percent of the total heat capacity for the highest temperatures measured, and it is not possible to determine the coefficient of the T^3 term accurately.

Since the Au nucleus has a very small magnetic moment ($\mu_{\text{Au}}^2/\mu_{\text{Mn}}^2 = 0.0015$), the observed hyperfine heat capacity can be considered to come from Mn nuclei only. (A recent Mössbauer effect study on the same sample used in this work⁴ showed that there was no significant hyperfine field at the Au nuclei. This can be understood by symmetry considerations.) A hyperfine field H_e of 320 kOe at the Mn nuclei is thus calculated from the T^{-2} term by comparison with the first term of the equation

$$\frac{C_N}{fR} = \frac{1}{3} \left(\frac{I+1}{I} \right) \left(\frac{\mu H_e}{kT} \right)^2 - \frac{1}{30} \frac{(I+1)(2I^2+2I+1)}{I^3} \left(\frac{\mu H_e}{kT} \right)^4 + \dots \quad (2)$$

where R is the gas constant, k is Boltzmann's constant, and f , I , and μ are the atomic fraction, the nuclear spin, and the nuclear magnetic moment of Mn. This value is used to calculate the expected T^{-4} term of C_N , which is that given in Eq. (1).

The coefficient in the electronic heat capacity, corresponding to a Fermi level density of states of 0.445/eV-AuMn molecule, is remarkably low for a transition metal compound with an unfilled d-shell. On the assumption of divalent Mn and monovalent Au (three electrons per unit cell) one has a thermal effective mass ratio $m^*/m = 0.68$. In view of the expected high density of states for the d-band levels of the manganese atoms, this result indicates a negligible contribution from the d-band to the Fermi level density of states. This could be understood in terms of a band picture for the Mn d electrons in which the band for electrons with spin up at a particular Mn site is entirely below the Fermi level, and the other half of the d-band for electrons with spin down is entirely above the Fermi level. Such a description suggests a $3d^5$ configuration and a localized moment of $5\mu_B$ for the Mn ion cores. Neutron diffraction measure-

ments¹ give a value of $4.2\mu_B$, while magnetic susceptibility measurements in the paramagnetic region⁵ indicate a moment near $5\mu_B$.

In conclusion we wish to thank Lawrence Welsh for assistance in the preparation of the alloy.

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FIGURE CAPTION

Fig.1 The heat capacity of AuMn. For the upper curve the T^{-4} term makes a negligible contribution.

Fig. 1