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Low-temperature specific heat of $LaAl_2$:Gd in magnetic fields up to 4900 Oe*

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The specific heat of $LaAl_2$:Gd was measured between 1.3 and 6 K for Gd concentrations of 0, 2.0, 3.0, and 6.0 at.% in various applied magnetic fields up to 4900 Oe. Broad peaks associated with spin glass transitions in zero field were shifted to higher temperatures by the applied field. The field dependence of the magnetic specific heat follows a law of corresponding states and exhibits several characteristics predicted by molecular-field theories based on the Ruderman-Kittel-Kasuya-Yosida interaction. For no value of applied field were the results consistent with the absence of magnetic order as suggested by nuclear relaxation measurements.

INTRODUCTION

In zero and small applied fields (a few hundred Oe or less) the onset of magnetic order in dilute alloys of Gd in $LaAl_2$ has been observed by magnetization,[1] specific-heat,[2] and nuclear-quadrupole-resonance[3] techniques. This ordered state, called the magnetic or spin glass state,[4] is due to the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction between the randomly distributed solute spins, and molecular-field theories based on this interaction[5] have successfully accounted for a number of the features of such systems at low temperatures.

While certain aspects of the low-field problem are reasonably well understood, there is evidence that the magnetic order in $LaAl_2$:Gd may be anomalously affected by the presence of larger applied fields. NMR measurements by McHenry *et al.*[6] taken in fields greater than 3.5 kOe were consistent with the absence of magnetic order, even down to 1.2 K for samples with Gd concentrations up to 10 at.%. On the other hand, low-field magnetization measurements by Maple[1] indicate an ordering temperature of about 10 K for a 10-at.% sample. McHenry *et al.* offered two possible explanations for this apparent discrepancy: one that NMR and magnetization measurements may sample the ordered spin system quite differently; the other that the applied field itself may alter the nature of the magnetic order.

Nuclear-quadrupole-resonance (NQR) measurements by MacLaughlin and Daugherty[3] taken in zero and small fields verified the magnetization data by a method which presumably samples the spin system in the same way as does NMR. Thus, the NQR measurements seem to confirm the speculation that the applied field significantly alters the ordered state, but do not rule out the possibility that the sensitivity of the resonance technique to ordered spins is field dependent.

It should be mentioned that unusual behavior with respect to small applied fields has been observed in susceptibility measurements on La:Gd and La_3In:Gd, in which the height of the susceptibility peak at the transition temperature T_m can be drastically reduced by a small field H, even when $\mu H \ll k_B T_m$.[7,8]

To help clarify how the magnetic order in $LaAl_2$:Gd is affected as applied field is increased, we report here the results of specific-heat measurements on this system in various fields up to 4900 Oe.

EXPERIMENTAL

The samples used for this study had Gd concentrations of 0.0, 2.0, 3.0, and 6.0 at.%. They were prepared from rare earths of 99.9% purity and Al of 99.999% purity by a combination of arc melting and induction heating which has been described previously.[2]

Specific heats were measured between 1.3 and 6 K using standard techniques described elsewhere.[9] Temperatures were measured with a germanium thermometer whose calibration was corrected for magnetoresistance effects. The calibration was checked by measuring the specific heat of 5.8 moles of 99.999% pure copper. For all values of applied field, the data deviated from published results[10] by 1% or less.

Demagnetizing fields for each sample were estimated from the magnetization data of Maple[11] for a sample of concentration 15 at.% by assuming that the magnetization obeys a law of corresponding states.[12] Demagnetization effects were found to be small enough such that the approximate equivalence of the external and internal fields could be assumed for all samples.

RESULTS AND DISCUSSION

Over the temperature range studied the total specific heats of the alloys can be written as

$$C = \gamma T + \beta T^3 + (\Delta\gamma) T + C_M ,$$

where the first two terms represent the matrix specific heat, with $\gamma = 10.08 \pm 0.05$ mJ/mole K^2 and $\beta = 0.124 \pm 0.001$ mJ/mole K^4 as previously reported.[2] The term $(\Delta\gamma)T$ is an enhancement of the linear term due to the addition of Gd atoms and is approximately proportional to solute concentration ($\Delta\gamma \approx 1.0$ mJ/mole K^2 at.% Gd). The magnetic contribution C_M, which represents the major portion of the total specific heat, is due to the interactions between the solute moments. In making this separation we have assumed that γ, β, and $\Delta\gamma$ are all unchanged in the presence of an applied field. This is consistent with the work of Luengo and Maple,[13] which showed no appreciable changes in these terms up to 3400 Oe for Gd concentrations up to 0.4 at.%.

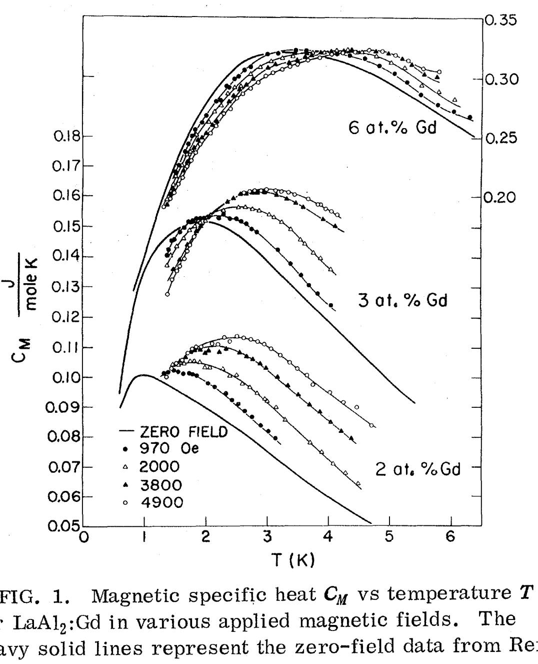

FIG. 1. Magnetic specific heat C_M vs temperature T for $LaAl_2$:Gd in various applied magnetic fields. The heavy solid lines represent the zero-field data from Refs. 2 and 9.

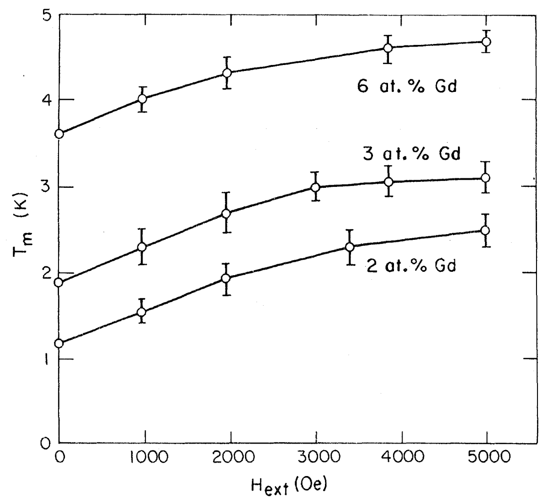

FIG. 2. Characteristic temperatures T_m, corresponding to maxima in C_M, vs applied field H_{ext} for $LaAl_2$:Gd. The vertical bars represent the uncertainty in T_m due to the broad nature of the transition. Solid lines are drawn for visual aid.

Figure 1 shows plots of C_M versus T for each sample, measured in various fields. The zero-field results, discussed elsewhere,[2] are characterized by large maxima occurring at temperatures T_m. Both T_m and $C_M(T_m)$ are proportional to Gd concentration in zero field. Below T_m, C_M becomes linearly proportional to T and independent of concentration. The principal effect of the applied field is an increase in T_m accompanied by a general broadening of the peak.

Figure 2 shows a plot of T_m versus H_{ext}, the applied field. The striking feature of this data is that the curves are nearly parallel, indicating that the effect of H_{ext} on T_m is concentration independent. Up to 2–3 kOe, ΔT_m varies linearly with H_{ext}, with $\Delta T_m / \Delta H_{ext} \approx 0.2$ K/kOe $\approx 0.4 g \mu_B S / k_B$, where $g = 2.0$ and $S = \frac{7}{2}$ are used for Gd.

Souletie and Tournier[12] showed that if the interaction between moments is due to an RKKY-like interaction, then the reduced specific heat, defined as C_M/c, where c is the concentration, is related to temperature and applied field by a law of corresponding states; that is, $C_M/c = f(T/c, H_{ext}/$

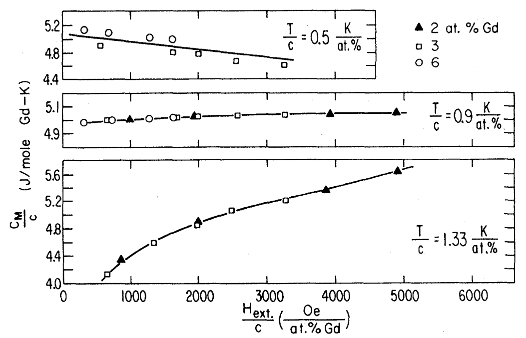

FIG. 3. Reduced magnetic specific heat C_M/c vs reduced applied field H_{ext}/c for different values of reduced temperature T/c, where c is Gd concentration.

c), where f is a concentration-independent function. For zero field, plots of C_M/c versus T/c show that $LaAl_2$:Gd does exhibit such behavior up to and somewhat above T_m.[2] In Fig. 3 plots of C_M/c versus H_{ext}/c for several constant values of T/c indicate that the law of corresponding states is also obeyed for applied fields up to 4900 Oe.

The specific heat both in zero and applied fields of a dilute Ising system of spins coupled by RKKY-like interactions has been calculated by Klein[14] using the mean-random-field approximation, whereby all functions of molecular field (which may include an applied field) are replaced by their averages over the distribution of molecular fields $P(H)$. By assuming that the solute concentration is so low that the oscillatory nature of the RKKY interaction can be averaged, a form for $P(H)$ was found which is Lorentzian and symmetric about an average field $H=0$. In the presence of an applied field, $P(H)$ remains Lorentzian but is broadened and shifted by an amount H_{ext}. Using this form of $P(H)$, Klein showed that the specific-heat peaks will be broadened and shifted to higher temperatures by the field, in qualitative agreement with our results, but he also predicted that T_m will increase faster than linearly with increasing H_{ext}, in contradiction to the results shown in Fig. 2.

A more heuristic molecular-field theory, proposed by Liu,[15] assumes a form for $P(H)$ based on the arguments of Marshall.[16] Since the numerical values for the parameters for a given alloy system are not found from first principles in this theory, they must be determined by a computer fit to the zero-field specific-heat data. When this is done the field dependence of C_M can then be calculated. Such a calculation for a 1-at.% La:Gd alloy yielded specific-heat curves very much like those in Fig. 1, predicting that the shift in T_m increases linearly with H_{ext} but slower by a factor of 4 than that actually observed in $LaAl_2$:Gd. We point out that if the shift in T_m is proportional to H_{ext} then the concentration independence of the shift follows from the law of corresponding states.

A theory based on the assumption of inhomogeneities in the solute distribution was developed by Bennemann *et al.*[17] to explain the anomalous behavior of the susceptibilities of La:Gd and La_3In:Gd in small applied fields. While the theory quantitatively explains the observed susceptibility results, it also predicts that as long as $T_m < \theta_p$, where θ_p is the paramagnetic Curie temperature, the effect of small fields on C_M will be a rapid narrowing and suppression of the peak with increasing H_{ext}. Such behavior is not observed for $LaAl_2$:Gd. The T_m values we obtain are less than θ_p values obtained by Maple[1] for concentrations greater than about 3 at.%, but specific-heat and susceptibility measurements have not been made on the same samples.

CONCLUSION

The qualitative aspects of the results reported here can be understood in terms of theories based on the RKKY interaction and the existence of a continuous molecular-field distribution, without making allowances for the presence of magnetic clustering.

It may be reasonable to conclude that the effect of applied fields (at least up to several kOe) on the $LaAl_2$:Gd system is a modification of $P(H)$ which manifests itself in a modification, but not a destruction, of the magnetic ordering process. The transitions observed in small fields by other techniques are also observed here in fields up to 4900 Oe, making attractive the hypothesis that the nuclear relaxation technique becomes insensitive to ordered spins at high fields, but offering little insight as to why this is so.

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