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LOW TEMPERATURE SPECIFIC HEAT OF Th–Gd SPIN GLASS

J.G. Sereni* and T.E. Huber

Centro Atomico Bariloche (CNEA), 8400 Bariloche, Argentina

and

C.A. Luengo

Universidade Estadual de Campinas, Campinas, S.P., Brasil

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The specific heat measurements of Th–Gd alloys, with Gd concentration ranging from 0.15% to 10.8% and temperatures between 0.4 and 8 K, are reported. The characteristic spin glass specific heat enhancement and its maximum was found linearly proportional to Gd concentration. At low temperatures a T^2 dependence of the specific heat was observed and the entropy associated with the magnetic ordering was the 70% of the expected value for the spin $S = 7/2$.

WHEN a Gd ion is diluted in a metallic host it conserves its magnetic moment characterized, after Hund's rules, by $J = S = 7/2$ and $L = 0$. The exchange interaction between the $4f$ and conduction electrons induces oscillatory spin and charge densities [1] extended to many interatomic distances. Other impurities, placed at random, in the perturbation region of the original ion, will be polarized. All impurities interacting through this mechanism will give rise to an effective magnetic field with different modulus and direction in each atomic site. This, neither ferro nor antiferromagnetic order, is the so-called "Spin Glass" [2].

As the thermal disorder decreases with the temperature decrement, the ionic magnetic moment is frozen at a characteristic temperature (T_m) in an "at random" direction determined by the local magnetic field [3]. This phase transition leads to an anomaly in the specific heat and magnetic susceptibility, with a maximum at T_m, as was observed in a number of transition metals diluted in noble metals [4] or in rare earths diluted in metallic hosts [4–7].

The Th–Gd system is particularly appropriated for the study of this magnetic phase because of: (1) the above mentioned Gd ion properties; (2) the absence of long-range magnetic order at the underpercolation Gd concentration, as was determined by neutron diffraction [8] and electrical resistivity measurements [9]; (3) due to the absence of an orbital moment, no crystal field effects are expected; and (4) the study of Th–Gd alloys will complement the systematic study made on other matrices, i.e., Sc, Y, La, $LaAl_2$ and La_3In [4–7].

The samples were f.c.c. solid solutions of Gd 99.9% pure and Th 99.98% pure, melted in an arc furnace. All samples, wrapped in Ta foils, were annealed 10 hr at 900°C under vacuum. The specific heat measurements were performed in a semi-adiabatic ^3He calorimeter, using the heat pulse technique [10]. A previously calibrated Ge resistor was used as thermometer. Eight samples were studied in the range between 0.4 and 8 K, and the Gd concentration was: $n = 0.14, 0.23, 0.51, 1.66, 2.85, 5.24$ and 10.8 in at.%.

The spin glass specific heat (C_{sg}) was obtained by the subtraction of the matrix specific heat (C_{Th}) to the measured one (C_t), i.e., $C_{sg} = C_t - C_{Th}$. It was observed that 10% of La or Y diluted in Th does not change the Debye temperature of Th in more than 2% [11], which means a change of 3% in the lattice specific heat. This value is negligible with respect to the C_{sg} one at the reported Gd concentration. As Gd lies nearer than La and Y to Th (in mass as in volume), it is not expected to have larger corrections.

In Fig. 1 we have plotted C_{sg}/n vs T/n, showing that both, the maximum of $C_{sg}(C_m)$ and the temperature at which occurs (T_m), look proportional to n (see also Table 1). The mean values obtained are:

$$C_m/n = (50 \pm 8) \text{ mJ mole}^{-1} \text{ K}^{-1} \text{ Gd\%}^{-1}$$

and

$$T_m/n = (0.55 \pm 0.05) \text{ K Gd\%}^{-1}$$

(that means $T_m = 55$ K in atoms of Gd units).

The specific heat enhancement at $T \gg T_m$ was

* Present address: II Physikalisches Institut, Universität Köln, 77 Zülpicher Strasse, 5 Köln 41, W. Germany.

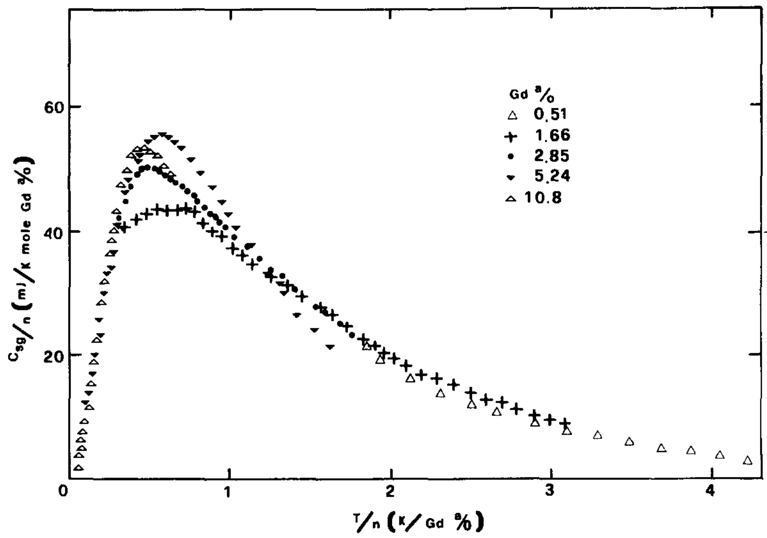

Fig. 1. Reduced magnetic contribution to the specific heat: C_{sg}/n vs reduced temperature: T/n.

Table 1.

n (Gd%)	T_m/n (K Gd%$^{-1}$)	$C_{sg}\,T^2/n^3$ (mJ K mole^{-1} Gd%$^{-3}$)	γ_{sg}/n (mJ K^{-2} mol^{-1} Gd%$^{-1}$)
0.14	–	65	2.5
0.23	–	73	2.0
0.51	–	68	3.0
1.66	0.65	75	2.6
2.85	0.50	–	2.8
5.24	0.65	–	–
10.8	0.45	–	–

evaluated as a contribution to the electronic specific heat as:

$$\gamma_{sg}/n = (2.5 \pm 0.5)\ \text{mJ mole}^{-1}\ \text{K}^{-2}\ \text{Gd\%}^{-1}$$

this enhancement can be attributed to a change in the density of states and to an increment of the electron–phonon coupling. The γ_{sg}/n and the T_m/n values are significantly similar to the previous values obtained from other Gd alloys [4–7].

At $T > T_m$, the expected T^{-2} dependence of C_{sg} has been found, with:

$$C_{sg}T^2/n^3 = 70\ \text{mJ K mole}^{-1}\ \text{Gd\%}^{-3}.$$

A further accurate analysis of the low temperature behaviour ($T < T_m$) was made in Fig. 2, which shows a T^2 dependence of C_{sg}.

The T^2 term coefficient can be evaluated as:

$$C_{sg}n/T^2 = 1.2\ \text{J Gd\% mole}^{-1}\ \text{K}^{-3}.$$

From early theoretical descriptions of spin glass systems, a linear increment of C_{sg} with T has been predicted [12]. But in the recent Thouless *et al.* "solution" [13] of the Sherrington and Kirkpatrick "solvable model of a spin glass" [14], a T^2 dependence of C_{sg} at $T < T_m$ was found. Using the formula [13]

$$C_{sg} = 1.54\,N\,(T/\tilde{J})^2$$

we have calculated $\tilde{J}$, obtaining $\tilde{J} = 2.72$ meV $\simeq 32$ K. It must be noted that the ratio $T_m/\tilde{J} = 1.7$ has practically the same value of the α parameter estimated by Thouless ($\alpha = 1.66$).

We have calculated the entropy associated with the anomaly, assuming a perfect scaling of C_{sg} with n in the Souletie [15] sense, as it is justified by the low and high temperature behaviour of C_{sg} and the C_m and T_m proportionality with n. The entropy value was obtained by numerical integration from Fig. 2 and it is $S/R = 0.7 \ln 8$ (in the case of Gd it is $2J + 1 = 8$). A similar result was obtained for (La–Gd)Al_2 [16].

In conclusion, the Th–Gd specific heat exhibits an agreement with the pictures that predict an impurity concentration scaling of C_{sg}, C_m, and T_m. The low

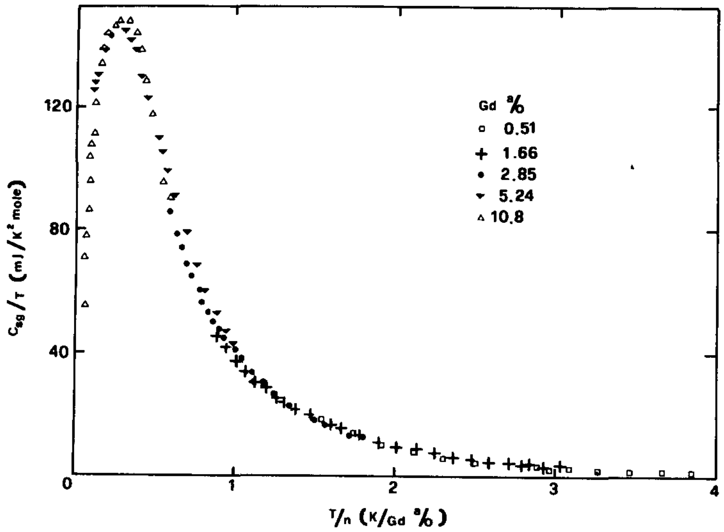

Fig. 2. C_{sg}/T vs T/n, revealing the T^2 behaviour of the magnetic specific heat in the low temperature region.

temperature T^2 behaviour only agrees with the Thouless *et al.* "solution".

It must be pointed that the difference between the expected and the measured value of entropy was just quoted by Anderson [3] as a characteristic of a glass, but was not discussed in any theory.

The effective magnetic field (H_{eff}) induced by the localized magnetic moments, can be estimated from the T_m/n value by the expression:

$$H_{eff}/n = k_B/\mu_B \; T_m/n$$

where k_B and μ_B have the classical values. One finds:

$$H_{eff}/n = 8 \text{ KOe Gd\%}^{-1}.$$

In order to check this value, we have applied an external magnetic field to the 1.66 Gd% sample, finding a shift in temperature of the C_m peak to $\sim 2T_m$ when the applied field was 9 KOe. To clarify this point further, more detailed experiments on the specific heat of Th–Gd under magnetic field and susceptibility measurements are in progress.

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