The Specific Heat of $\text{Er}_x\text{Y}_{1-x}\text{Rh}_{1.1}\text{Sn}_{3.6}$ for $x=0.4, 0.8$ and 1

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Specific heat measurements between 50 and 800 mK of the three magnetic superconductors $\text{Er}_x\text{Y}_{1-x}\text{Rh}_{1.1}\text{Sn}_{3.6}$ with $x = 0.4, 0.8$ and 1 , respectively, are presented. For these compounds broad anomalies with maxima at 245, 360 and 435 mK, respectively, were observed. No sharp peaks were detected. This means that magnetic short-range order gives the main if not the full contribution to the specific heat. A low-lying doublet is responsible for the magnetic behaviour below 1 K. The results are compared with some theories.

1. INTRODUCTION

The magnetic superconductor with the approximate composition $\text{ErRh}_{1.1}\text{Sn}_{3.6}$ (ERS) can be relatively easily produced in single-crystal form. Electrical resistance measurements show reentrant behaviour [1], the specific heat has been determined between 0.4 and 1.3 K [1] and magnetic measurements are reported [2]. Hodeau et al determined the detailed structure, which is quite complicated [3]. The magnetic Er atoms occupy two different lattice sites. Moreover, one of the two sites is occupied for a large part by Sn atoms, thus causing a structural disorder. Therefore, a simple magnetic behaviour can hardly be expected. It was remarked [4] that low-temperature neutron diffraction measurements revealed no long-range magnetic order over a 200 Å distance range.

We have performed specific heat measurements at $T < 1$ K in order to obtain more information about a possible magnetic ordering. This was also done for materials of composition $\text{Er}_x\text{Y}_{1-x}\text{Rh}_{1.1}\text{Sn}_{3.6}$ (EYRS) with $x = 0.4$ and 0.8 , respectively.

2. METHOD

The heat capacity measurements were performed by means of a thermal relaxation method. Details of the measurement techniques, the data acquisition and the data analysis will be reported elsewhere. The mass of the samples was 70 - 200 mg.

Fig. 1. The specific heat of $\text{Er}_x\text{Y}_{1-x}\text{Rh}_{1.1}\text{Sn}_{3.6}$ (EYRS) for $x = 0.4, 0.8$ and 1 , respectively.

3. RESULTS

3.1.1. Measurements in zero field

Fig. 1 shows the results of the measurements in zero field. At $T > 150$ mK, we see Schottky-type anomalies with maxima at $T_{\text{max}} = 245, 360$ and 435 mK, respectively, for the three samples. At temperatures below about 150 mK, another maximum develops, due to the magnetic moments of the ^{167}Er nuclei. The data for $T < 70$ mK give a general indication, but they are not yet very reliable. In Fig. 2, the specific heat of the three samples is shown after the subtraction of the nuclear specific heat, as derived from the NMR data of Sano et al [5]. The full curves in the Figs. 1 and 2 are drawn as a guide to the eye.

3.1.2. Conclusions and comparison with theory

1. It is observed that $T_{\text{max}}(x)/T_{\text{max}}(1) > 1$ for $x = 0.4$ and 0.8 . Since no theory for magnetic phase transitions of magnetically dilute systems leads to this result, we conclude that the maxima should not be associated with phase transitions.

2. Just as Lambert et al [1], we have detected no sharp peaks which are indicative of a phase transition. This observation, combined with point 1 leads to the conclusion that at least a very large part of the entropy is removed by short-range order.

3. The change of entropy ΔS , associated with the anomaly in the specific heat, was determined for the three samples. We found: $\Delta S = 4.5 \pm 1$ J/mol.K for $x = 0.4$, 4.95 ± 0.5 J/mol.K for $x = 0.8$

Fig. 2. The specific heat of the three samples of EYRS after subtraction of the specific heat of the ^{167}Er nuclei.

Fig. 3. Best-fit comparison of the three curves of fig. 2 with the theory of Oguchi and Obokata.

and 5.1 ± 0.5 J/mol.K for $x = 1$, respectively. Since $R\ln 2 = 5.76$ J/mol.K, we conclude that the magnetic behaviour is dominated by a low-lying doublet.

4. In Fig. 3, the higher temperature parts of the three curves of Fig. 2 are compared with the theory of Oguchi and Obokata [6] for magnetically diluted systems in which the molecular-field energy is much smaller than the thermal energy. In this way, values for the exchange constant J and the effective number of nearest neighbours z were obtained for the Ising as well as for the Heisenberg model. In table 1 these values are summarized.

Table 1

x	$J/k(\text{Is})$	$J/k(\text{Heis})$	$z(\text{Is})$	$z(\text{Heis})$
0.4	0.54 K	0.24 K	4.2	8.7
0.8	0.79 K	0.32 K	2.3	6.1
1	0.62 K	0.21 K	3.2	10.7

The averaged values for J/k and z were used to calculate the magnetic ordering temperature T_m by means of relations from ref. 6. In table 2, these theoretical values are given. They are compared with experimental values (exp) of T that were defined as the temperature at which there is a sharp rise in the zero field, d.c. magnetization [2].

Table 2

x	$T_m(\text{Is})$	$T_m(\text{Heis})$	$T_m(\text{exp})$
0.4	-	-	-
0.8	0.514 K	0.610 K	0.375 K
1	0.678 K	0.813 K	0.550 K

For this "phase transition temperature", the Ising model gives better results.

5. If we conclude tentatively that the Ising model gives a somewhat better description of the behaviour of the specific heat of EYRS than the Heisenberg model, z is rather small. Therefore, we also compared the specific heat of independent pairs and tetrads, calculated with the Ising, XY and Heisenberg models, respectively, with the specific heat of ERS ($x = 1$). The two best fitting

Fig. 4. Comparison of the specific heat of ERS with the theory of Oguchi and Obokata (—), the specific heat of independent pairs in the XY model (----) and that of independent tetrads in the Ising model and $J > 0$ (— · — ·).

curves, as shown in Fig. 4, are the dashed line, representing the specific heat of pairs in the XY model, and the dash-dotted line, being the specific heat of tetrads in the Ising model, both with $J > 0$. This sign of J was chosen because all experiments suggest ferromagnetic interactions. The Oguchi-Obokata curve for high temperatures has also been drawn.

We conclude that we are dealing with a rather "loose" magnetic structure with a low z -number. This is in agreement with the large amount of short-range order that has been observed.

3.2. Measurements in an external field

Measurements were performed for $x = 0.8$ and 1 in a field of 50 mT. The results were analogous to those of Lambert et al [1].

4. RELAXATION EFFECTS

As mentioned above, the specific heat was determined by the measurement of thermal relaxation times. However, d.c. magnetization measurements in zero field show relaxation times around $T_m(\text{exp})$ of up to a few hundred seconds [2], see Table 2. We believe that the latter effects are mainly due to the shifting of domain walls of some kind, which probably gives a negligible contribution to the heat capacity. Almost the total change of entropy is associated with the magnetic behaviour within the domains (para process) which has relatively short relaxation times.

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