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INTERPRETATION OF SPECIFIC HEAT AND SPONTANEOUS MAGNETIZATION ANOMALIES AT THE RE-ENTRANT SUPERCONDUCTING–FERROMAGNETIC TRANSITION IN $(Ho_{0.6}Er_{0.4})Rh_4B_4$

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Analysis of neutron diffraction data on the compound $(Ho_{0.6}Er_{0.4})Rh_4B_4$ indicates that the Curie temperature is depressed by about 0.2 K, due to the occurrence of superconductivity, in agreement with theoretical predictions. The temperature dependence of the specific heat in the vicinity of the first order re-entrant superconducting–ferromagnetic transition was computed by means of a simple model from the temperature dependence of the spontaneous magnetization of the Ho ions and was found to be in good agreement with the experimental data.

Specific heat and neutron diffraction measurements have proven to be very useful in determining the basic nature of the re-entrant transition in ternary and pseudoternary ferromagnetic superconductors [1]. In particular, Mook et al. [2] have studied the re-entrant transition in $(Ho_{0.6}Er_{0.4})$ Rh_4B_4 via neutron diffraction measurements while MacKay et al. [3] have measured the specific heat of a different sample at this composition. We have analyzed their data which, in conjunction with a simple model, elucidate several important aspects of re-entrant superconductive behavior.

The low-temperature phase diagram of the $(Ho_xEr_{1-x})Rh_4B_4$ system has been studied in detail by Johnston et al. [4]. The superconducting transition temperature (T_s) of the $(Ho_{0.6}Er_{0.4})Rh_4B_4$ sample used in the neutron study was $T_{s2} = 7.1$ K, as determined from ac magnetic susceptibility (χ_{ac}) measurements, whereas the re-entrant transition determined from χ_{ac} occurred at $T_{s2} = (3.50 \pm 0.05)$ K. Using the relationship between the magnetic neutron intensity (I) and the spontaneous magnetization (M), $I \sim M^2$, the M/M_0 vs. T/T_c data taken upon warming for the (101) peak shown in fig. 1 were derived. Here $M_0 = M(T = 0)$, while T_c is the "effective" Curie temperature, as discussed below.

Ott et al. [5] have shown that $HoRh_4B_4$ accurately follows mean field theory (MFT) in the temperature dependence of M, and Mook et al. [2] and MacKay et al. [3] have argued that the ordering which takes place in $(Ho_{0.6}Er_{0.4})Rh_4B_4$ at 3.67 K is due solely to the Ho ions. We have therefore fitted MFT to the M/M_0 vs. T/T_c data in fig. 1; for temperatures below T_{s2} excellent agreement was found for a MFT fit (solid curve

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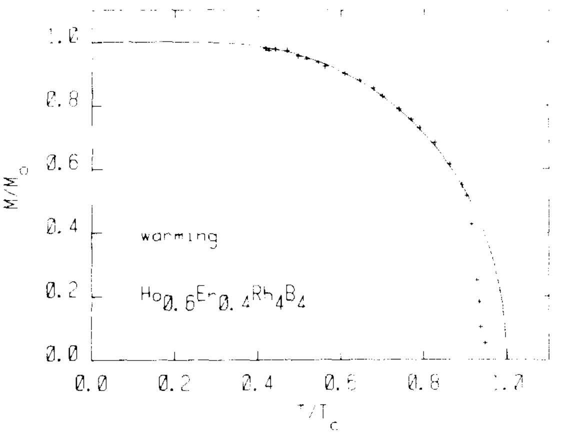

Fig. 1. Temperature dependence of the magnetization $M(T)$ derived from the (101) peak intensity. The solid curve is calculated from MFT.

in fig. 1) with $M_0 = 163\ (\mathrm{counts})^{1/2}$ and $T_c = 3.875$ K according to

$$\frac{M(T)}{M_0} = \tanh\left(\frac{T_c}{T}\frac{M(T)}{M_0}\right). \tag{1}$$

The observed Curie temperature as determined from neutron diffraction ($T_c = 3.67$ K) is seen to be considerably depressed from the "effective" T_c extrapolated from the MFT curve shown ($T_c = 3.875$ K). Deviations from the MFT curve begin at the temperature at which superconductivity begins to set in, $T = 3.45$ K, which is the 10% point of the χ_{ac} transition. This analysis clearly shows that the actual T_c is less than would have occurred in the absence of superconductivity, in agreement with theoretical predictions [6]. The effective T_c of 3.875 K is the same as that derived from the extrapolation to $x = 0.6$ of the magnetic ordering temperatures in $(Ho_xEr_{1-x})Rh_4B_4$ from above $x_{cr} = 0.89$, as shown by the dashed line of fig. 2 in ref. 4.

If we assume that the Ho^{3+} ground state doublet [5] is split by the internal field H, then the magnetic contribution to the specific heat C is related to the M/M_0 vs. T/T_c data according to the following equation [7]:

$$\frac{C(T)}{R} = -0.6\frac{\mathrm{d}}{\mathrm{d}T}\left[T\frac{M(T)}{M_0}\operatorname{arctanh}\left(\frac{M(T)}{M_0}\right)\right], \tag{2}$$

where R is the molar gas constant. Using the observed values of $M(T)/M_0$ vs. T, $C(T)/R$ was derived from eq. (2) and is shown as the solid curve in fig. 2. The actual magnetic heat capacity obtained by subtracting the electronic, lattice, and the Er electronic, Ho nuclear and Ho electronic Schottky anomaly contributions from the measured heat capacity data for a different sample of the same composition [3], is shown by the data points. The solid curve lies below the points at temperatures below 2 K, due to a Schottky anomaly arising from the two low-lying doublets of the Er^{3+} ions [8]. However, the curve and the points are in excellent agreement above 2 K and, in fact, even the spike in the heat capacity is very well reproduced. This implies that the spike-shaped feature in the heat capacity of $(Ho_{0.6}Er_{0.4})Rh_4B_4$ does not arise solely from the re-entrant superconducting to normal state transition as formerly presumed [3]. Instead, it appears to be associated primarily with the rapid onset of the spontaneous magnetization. A calculation of the entropy S under the theoretically

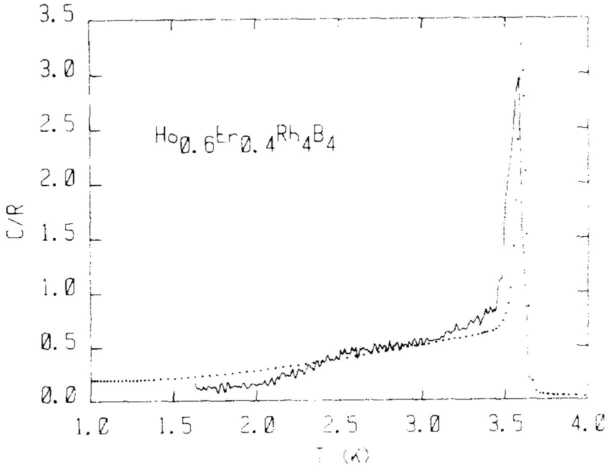

Fig. 2. Magnetic heat capacity C divided by the molar gas constant R vs. temperature T. Points: measured data; solid curve: calculated from the $M(T)$ data.

derived solid curve between 0 and 3.71 K yields $S/R = 0.396$, 95% of (0.6) ln (2), the value expected from magnetic ordering of the Ho ions with a doublet ground state.

The rapid increase in M apparently reflects a first-order transition from the ferromagnetic to the superconducting state at T_{s2} [2]; the jump in M occurs over a finite temperature interval ~0.2 K, possibly due to sample inhomogeneities. The spike-shaped feature in C at T_{s2} is presumably due to a latent heat of transformation that is associated with the first-order superconducting–ferromagnetic transition. This is consistent with the thermal hysteresis in various physical properties near T_{s2} [1] and in accord with various theories [1]. The analysis shows that the major contribution to the latent heat of transformation is magnetic in origin. We estimate the maximum contribution to the latent heat of transformation due to the superconducting–normal transition alone to be $\gamma T_{s2} \sim 90$ mJ/(mole · K) which is nearly an order of magnitude smaller than that attributable to the spike-shaped feature in C.

In conclusion, we have demonstrated that the Curie temperature of $(Ho_{0.6}Er_{0.4})Rh_4B_4$ is suppressed by the occurrence of superconductivity. We have also shown that the spike-shaped feature in the heat capacity of this compound arises primarily from the rapid change in the magnetization near T_c. It will be interesting to extend this analysis to other ternary and pseudoternary re-entrant superconductors [7].

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