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1989 J. Phys.: Condens. Matter 1 4381

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On magnetic first-order transitions in erbium

H U Åström and G Benediktsson

Department of Solid State Physics, Royal Institute of Technology, S-100 44 Stockholm, Sweden

Received 1 December 1988, in final form 27 January 1989

Abstract. The magnetic transitions in a single crystal of erbium have been investigated with a high-resolution scanning microcalorimeter. The transition between the ferro- and antiferromagnetic states takes place at 19.2 K (T_c) and is of first order with a latent heat of $18.5 \pm 1.0 \text{ J mol}^{-1}$. In the antiferromagnetic state between T_c and 51 K four small energy peaks were observed which are ascribed to commensurate transitions. These transitions exhibit hysteresis effects and are of first order. At 53.4 K a relatively localised energy peak of $1.3 \pm 0.3 \text{ J mol}^{-1}$ was found which probably arises from the ordering of the basal-plane moments. Also this transition was found to have a temperature hysteresis and is of first-order type. The antiferro-paramagnetic transition occurs at 85.6 K in this sample and its second-order nature is confirmed.

We report on a high-resolution calorimetric study of the magnetic transitions in erbium. Erbium has a complex magnetic structure [1] with a high-temperature region below the Néel temperature T_N (85 K) where the *c*-axis moments order in a sinusoidally modulated structure, an intermediate region below about 50 K with an additional component of the magnetic moment ordered in the basal plane in a helix, and a region below T_c (19 K) with a conical ferromagnetic ordering. For temperatures below about 22 K a 'squared-up' alternating cone structure was suggested [2] for the moments in the intermediate region. The turn angle between successive spins in the basal plane decreases monotonically from 50° at 52 K to 44° at T_c . At certain temperatures the turn angle attains values that make the magnetic structure commensurate with the lattice in the basal plane. These commensurate states are separated by regions where the magnetic structure is incommensurate, but from about 25 K to T_c the magnetic structure remains commensurate with the lattice [2]. The transitions to the commensurate structures have been described in terms of a spin-slip model, the wavevector locking to fixed values over a limited temperature interval [3].

The sample used for the calorimetric measurements was cut from a single crystal of erbium with a purity of 99.9 at. % or better kindly supplied by Dr D Fort of the Department of Metallurgy and Materials of the University of Birmingham. It had the shape of a half-disc with a diameter of about 6 mm and with a mass of 98.3 mg. The energy was measured with a sensitive high-resolution scanning microcalorimeter which was used in our previous study of the magnetic transitions in dysprosium [4]. The measurements were performed in warming and cooling runs by changing the temperature of the measuring cell linearly with time around the transitions. The rate of change of temperature is not critical within rather wide limits but a rate of about 1 mK s^{-1} was chosen

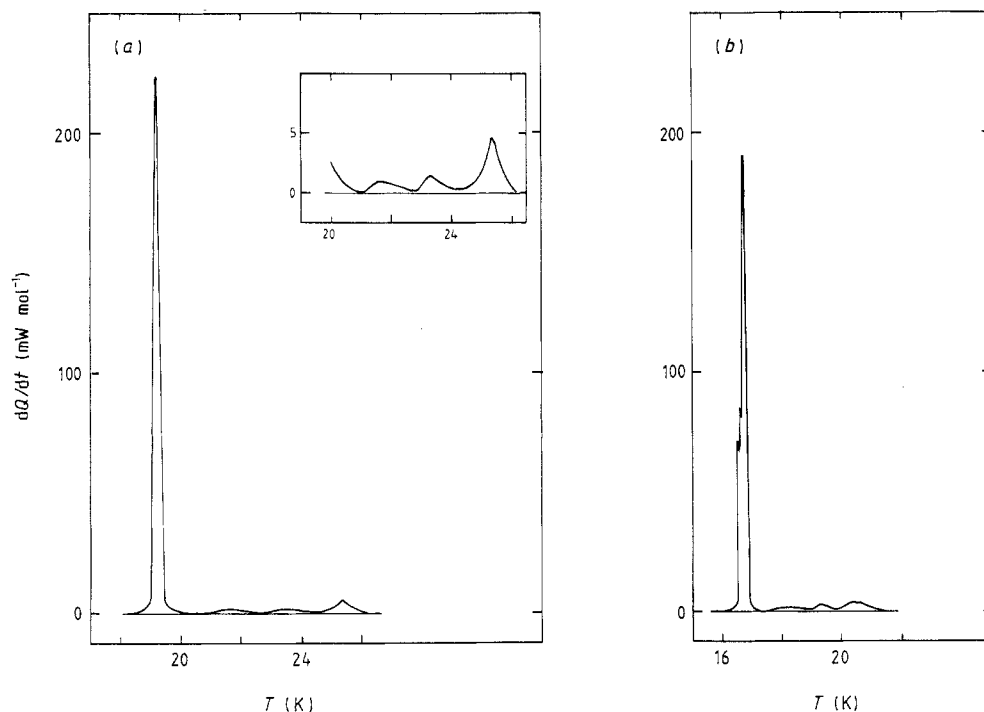


Figure 1. The change in energy content of erbium at the ferromagnetic transition as measured with increasing temperature (*a*) and decreasing temperature (*b*). The large peak below 20 K caused by the ferromagnetic transition corresponds to an energy of $18.5 \pm 1.0 \text{ J mol}^{-1}$. The inset shows a blow-up of the energy scale between 20 and 26 K. The energy of the peak at 25 K is $1.2 \pm 0.3 \text{ J mol}^{-1}$. The zero line is drawn as a guide to the eye.

to be most appropriate for the present measurements. The energy change of the sample was measured in a differential mode using an Au-0.3 at. % Fe-chromel thermocouple glued to the sample and an Au dummy of equal thermal mass. After amplification the signal from the thermocouple was recorded as the time rate of energy change dQ/dt against time and temperature. The sensitivity of the calorimeter was of the order of $0.1 \mu\text{W}$ per mm deflection on the recorder. After correction of the curves for the background, which varies linearly due to a slight difference between the thermal capacity of the dummy and that of the sample, the energy was obtained by integrating dQ/dt over time. Each run was repeated several times. The uncertainty in the calculated energies was of the order of a few per cent but for the smallest energies it could be up to 20%. The temperature was measured to within $\pm 0.2 \text{ K}$ by means of an AuFe-chromel thermocouple glued to the sample.

Curves characteristic of the measurements are plotted in figures 1 and 2 as dQ/dt against temperature where dQ/dt has been corrected for the background. At the ferromagnetic transition a narrow energy absorption peak was observed at 19.2 K during warming (figure 1). During cooling the transition took place at 16.7 K. The latent heat measured was $18.5 \pm 1.0 \text{ J mol}^{-1}$. Increasing the temperature above T_c , three minor endothermic peaks occurred between 21 and 26 K (see the inset to figure 1(*a*)), the largest with an energy of $1.2 \pm 0.3 \text{ J mol}^{-1}$ and with its maximum at 25.3 K, and the smaller peaks of about 0.5 J mol^{-1} at 21.6 K and 23.3 K. Instead of the two smaller peaks

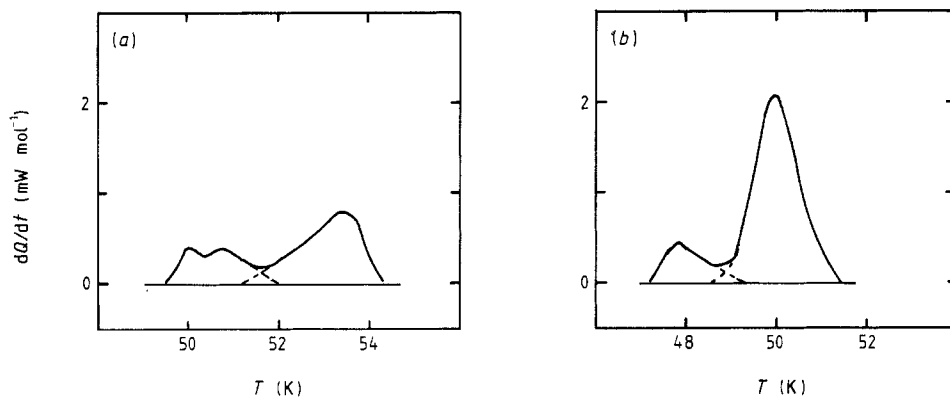


Figure 2. The change in energy content of erbium near the basal-plane ordering temperature for (a) increasing temperature and (b) decreasing temperature. The energy of the larger peaks is $1.3 \pm 0.3 \text{ J mol}^{-1}$. The zero line is drawn as a guide to the eye.

sometimes only one occurred at 21.6 K, of about the same energy. When cooling, similar exothermic peaks were obtained but were shifted to lower temperatures, the main peak then being at 20.5 K (figure 1(b)). We interpret the largest of the peaks as due to the commensurate transition observed at approximately 24 K in [3] with a locking of the wavevector to $\frac{1}{4}$ (in units of c^*). The nature of the two minor peaks is uncertain but they may also be due to commensurate transitions and/or to a transition caused by the squaring-up of the c -axis moments. The energy changes both for these transitions and for the ferromagnetic transition are of magnetoelastic origin and are due to distortions in the lattice when the moments change their orientation.

On further warming a broad energy absorption extending from 49 to 54 K was observed (figure 2(a)). It consists of two smaller peaks, one with its maximum at 53.4 K of $1.3 \pm 0.3 \text{ J mol}^{-1}$ and another at 50.8 K of $0.7 \pm 0.3 \text{ J mol}^{-1}$. When measured for decreasing temperature the peaks occurred at 50 and 48 K, respectively (figure 2(b)). (The cooling rate was somewhat higher than the warming rate which means that the areas under the curves for warming and cooling are not directly comparable.) The peak at 53 K is probably connected with the basal-plane ordering while the peak at 51 K could be due to a commensurate transition, presumably with locking of the modulation wavevector to $\frac{2}{3}$ [3]. Also, a commensurate transition at 33 K was resolved in [3]. We have, however, not been able to detect any significant energy effect around this temperature although susceptibility studies on the same crystal [5] revealed a slight change at 34 K. Possibly the energy change is in this case below the resolution of the calorimetric measurements.

At the Néel transition the calorimetric measurements showed only a smooth s-like variation of the background in the recordings due to the marked specific heat change of the sample at this transition. No latent heat or thermal hysteresis was observed. The temperature of the transition was found to be 85.6 K.

For the ferromagnetic transition the present results for the transition temperature and hysteresis are in accord with previous observations [2, 3]. No direct measurements of the latent heat of this transition have been reported to date. Hence it might be of value to compare the result for erbium with the measured latent heats for the ferromagnetic transition of other rare-earth metals with closely related magnetic structures. For dysprosium, which undergoes a strong orthorhombic deformation at T_c , the

latent heat is between 14 and 35 J mol⁻¹ depending on the purity of the crystals [4]. For holmium, which like erbium does not have an orthorhombic deformation of the lattice at T_c , the latent heat is about 7 J mol⁻¹ [6]. Erbium, however, exhibits a more marked change of the elastic strain at T_c [7] than does holmium [8], which might explain why the latent heat is larger in erbium. Using the local strain formalism, Rosen and co-workers [9] calculated the jump in the magnetoelastic energy at T_c in erbium and obtained a value of 17 J mol⁻¹, while the same calculations for dysprosium gave an energy change of 26 J mol⁻¹ [10]. The latent heat at T_c can also be estimated using the Clausius–Clapeyron equation. With a relative volume change of $(-0.8 \pm 0.1) \times 10^{-3}$ at T_c [7] and a pressure dependence of T_c of $(-0.8 \pm 0.2) \times 10^{-8}$ K Pa⁻¹ [11] a latent heat of 30 ± 10 J mol⁻¹ is obtained. The measured latent heat at T_c for erbium is consistent with these results.

The commensurate lock-in transitions in erbium have been suggested [3] to be of first order. The relatively localised nature of the energy changes and the thermal hysteresis of the transitions support this interpretation. In very pure dysprosium, energies of commensurate transitions in the spiral regime have been measured [4]. They were found to be 0.2 ± 0.1 J mol⁻¹ which is of the same magnitude as for erbium. Differences in magnetoelastic properties or purity might explain the somewhat different energies connected with these transitions in erbium and dysprosium.

The temperature for the ordering of the basal-plane moments was in earlier investigations reported as 52 K [2, 3] and was found to increase with increasing purity from 48 to 55 K [12]. Our value of 53.4 is consistent with these results. The transition is in the present investigation found to have a thermal hysteresis of about 3 K. A hysteresis of about the same size is noticeable in the temperature dependence of the wavevector in the neutron diffraction study in [2] although the satellite intensities in the same investigation did not display any significant hysteresis. As to the transition order, synchrotron x-ray data [3] gave a discontinuous change of the wavevector at 52 K indicating a first-order transition, while the neutron results in [2] gave a more continuous change of the wavevector at the transition. From a strong anomaly in the elastic constants of erbium at the basal-plane ordering temperature, a first-order transition was suggested [9] while dilatometric measurements [13] showed a continuous change at this transition. The relatively localised energy change for this transition and the thermal hysteresis observed in the present investigation indicate that the transition cannot be of second order. The energy change is, however, somewhat too broad for an interpretation in terms of an ideal first-order transition to be completely justifiable. The transition may instead be characterised as a weak or disturbed first-order transition. This non-ideal behaviour might explain the diverging results for the transition order in the previous investigations.

The present results clearly show that the antiferromagnetic transition is of second order in accord with earlier experimental findings [2, 13] and theoretical predictions [14]. The transition temperature has earlier been reported to be 84.4 K [2] and 85 K [13]. It has, however, been shown [12] that T_N increases with increasing purity from 80 K in moderately pure erbium to 88 K in electrotransport-purified material. The present value of 85.6 K may then be taken as an indication that the purity of our sample is somewhat higher than that of the samples used in [2] and [13].

In conclusion, the ferromagnetic transition in erbium takes place at 19.2 K with a hysteresis of 2.5 K. It is of first order and the latent heat is 18.5 ± 1.0 J mol⁻¹ in good agreement with calculations. When increasing the temperature from T_c small endothermic peaks due to commensurate transitions were observed at 21.6, 23.3, 25.2 and 50.8 K. They are all of first order. The basal-plane ordering transition occurs at 53.4 K

on warming with an endothermic peak of $1.3 \pm 0.3 \text{ J mol}^{-1}$. The transition is connected with a thermal hysteresis of several degrees. The energy peak is more extended than is normal for an ideal first-order transition. Therefore the results may instead be explained as due to a weak or disturbed first-order transition. The second-order nature of the Néel transition, which takes place at 85.6 K, is confirmed in accord with earlier findings.

Acknowledgment

The work was supported by a grant from the Swedish Natural Research Council.

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