Hysteresis in the magnetic behaviour of holmium

1990 J. Phys.: Condens. Matter 2 1363

(http://iopscience.iop.org/0953-8984/2/5/028)

J. Phys.: Condens. Matter 2 (1990) 1363–1366. Printed in the UK

LETTER TO THE EDITOR

Hysteresis in the magnetic behaviour of holmium

A M Venter†, P de V du Plessis‡, G A Eloff† and E Fawcett§

† Atomic Energy Corporation of South Africa Ltd., PO Box 582, Pretoria, South Africa
‡ Department of Physics, Rand Afrikaans University, Johannesburg, South Africa
§ Department of Physics and Scarborough College, University of Toronto, Toronto M5S 1A7, Canada

Received 26 September 1989

Abstract. Hysteresis in the intensity of elastic neutron scattering from the incommensurate Bragg peak in the spiral phase of holmium is studied as temperature is cycled from above the Néel temperature, $T_N = 132.2$ K, to various low temperatures, T_0. The nature of the hysteresis is different according to whether $T_0 < T_L$ or $T_0 > T_L$, where $T_L \simeq 100$ K is the lock-in transition temperature at which the magnetic structure is supercommensurate.

The HCP rare-earth metal holmium orders magnetically as a basal-plane spiral below its Néel temperature $T_N = 132.2$ K. Spins are aligned in ferromagnetic sheets within each basal-plane layer, but the spin orientation of adjacent sheets is rotated by a turn angle $\omega = \frac{1}{2}(q/c^*)\ 360°$ (Koehler *et al* 1966). The wave vector $\boldsymbol{q}$ along the c axis takes a value of 0.28 $\boldsymbol{c}^*$ at T_N and decreases continuously upon decrease in temperature to a lock-in value of $\boldsymbol{q}_0 = \boldsymbol{c}^*/6$, corresponding to $\omega = 30°$, at the Curie temperature $T_c \simeq 20$ K. Below T_c the magnetic moments tilt towards the c axis to form a ferromagnetic cone structure, with $\boldsymbol{q}$ retaining the lock-in value $\boldsymbol{q}_0$ down to 4 K.

Anomalies were observed in the velocity and/or attenuation for particular modes of propagation of ultrasonic waves in single-crystal Ho at temperatures where $\boldsymbol{q}$ passes through commensurate values, in particular $\boldsymbol{q} = \boldsymbol{c}^*/4$ ($\omega = 45°$) at about 96 K (Lee *et al* 1972, Simpson *et al* 1976, 1979), and $\boldsymbol{q} = 2\boldsymbol{c}^*/11$ ($\omega = 32.7°$) at about 24.5 K (Tachiki *et al* 1974). Vigren (1976) explained these anomalies as magnetoelastically driven resonances in what has now become known as the spin-slip structure.

The spin-slip structure in Ho was first observed in magnetic x-ray scattering studies using synchrotron radiation (Gibbs *et al* 1985). In the initial model, pairs of planes have their moments aligned along a particular easy magnetisation direction in the basal plane (one of the six b axes), thus leading to a region of commensurate spin structure formed by a spiral of doublets. Commensurate regions are separated by a spin slip in which only one plane has its moment along a particular easy direction. Regular occurrence of the spin slips leads to a higher-order commensurate (or supercommensurate) phase.

Cowley and Bates (1988) refined this picture of the basic spin-slip structure to explain the complex spectrum of elastic neutron scattering that they had observed in Ho between 18 K and 30 K. Thus, for each layer of the doublet, the moment direction deviates by up to 10° from the easy direction. Furthermore, a small c axis antiferromagnetic component probably exists. Bates *et al* (1988) showed, in a systematic study of the elastic tensor in the spiral phase, that the magnetic symmetries corresponding to the supercommensurate

0953-8984/90/051363 + 04 \$03.50

structures, which they found to occur at the temperatures 97.4 K, 40.5 K, 24.5 K and 19.8 K, give rise to anomalies in particular elements of the tensor.

While some models give a continuous lock-in phase transition (McMillan 1976), the hysteretic behaviour seen in Ho shows that the succession of supercommensurate structures encountered as temperature decreases must give rise to a series of first-order transitions. Thus, for temperatures below 50 K, Gibbs *et al* (1985) observed thermal hysteresis, irreversibility and coexistence of phases with different wave vectors. Cowley and Bates (1988) reported hysteresis as large as 3 K at temperatures below 30 K, with relaxation times of at least two hours after the temperature was changed. Bates *et al* (1988) found hysteresis of about 1 K in the ultrasonic anomalies at 19.8 K and 24.5 K, and Simpson *et al* (1976) observed hysteresis in ultrasonic attenuation at 94 K.

We report measurements in holmium of the temperature dependence of the integrated intensity of neutron scattering from the (0 0 2-δ) Bragg position corresponding to the incommensurate wave vector of the helical magnet. A two-axis spectrometer was used, with the neutron beam of wavelength 1.08 Å diffracted from the reactor spectrum by a Ge(111) monochromator crystal. Intensity measurements were performed against a constant monitor count rate in order to negate the effect of variations in reactor power. Furthermore, the intensity of the (002) nuclear reflection was monitored during all temperature runs and observed to be free of hysteresis. (A typical result is shown later in figure 4). Thus the large hysteresis effects in the magnetic scattering to be reported below cannot be ascribed to instrumental sensitivity changes during the five-day period of a typical experimental run.

Three samples have been studied: Ho I of nominal purity 99.99% obtained from Metals Crystals, UK; and Ho IIa and Ho IIb of residual resistivity ratio, RRR, $\simeq$100, obtained from Materials Preparation Center, Ames, IA, USA. Ho I was a cylinder of diameter 6 mm and height 20 mm, while each Ho II sample was an approximate cube with edge 6 mm. Ho II samples were spark-cut from the same single-crystal ingot.

For the Ho II samples, we show in figures 1 and 2 hysteresis in neutron intensities over almost the whole range of the spiral phase for temperature cycles starting above T_N (typically from about 140 K, but data in the paramagnetic region are not shown) and extending to various low temperatures T_0 before subsequent heating. The temperature dependence of the hysteresis is qualitatively different when $T_0 > 100$ K from what it is when $T_0 < 100$ K. It is characteristic of temperatures $T_0 > 100$ K that the intensities during the heating part of the cycle are lower than those observed during cooling (see figures 1(C and D) and 4). When $T_0 < 100$ K on the other hand, intensities are larger for the heating curve than those observed during cooling (apart from an excursion to lower intensity values for about 10 K immediately above T_0). Furthermore, the magnitude of the hysteresis grows as the interval between T_0 and 100 K increases. In figure 1, curve B shows little hysteresis, but this may result from approximate cancellation of the effects manifested in curves A and C, whose T_0-values bracket that of curve (B).

The significance of the temperature $T \simeq 100$ K is believed to be its proximity to the temperature T_L at which the turn angle of the spiral is commensurate, i.e. $\omega = 45°$, and where an anomaly in the shear elastic constant C_{44} has been observed (see figure 2(*c*) of Bates *et al* 1988). Bates *et al* (1988) appear not to have investigated hysteresis effects, whereas our data indicate that the lock-in transition to the supercommensurate phase, for which $b = 2$ in the notation of Cowley and Bates (1988), is strongly hysteretic. This agrees with the ultrasonic observation of hysteresis at T_L by Simpson *et al* (1976).

We also believe that the small bump in the neighbourhood of 100 K, seen in all the

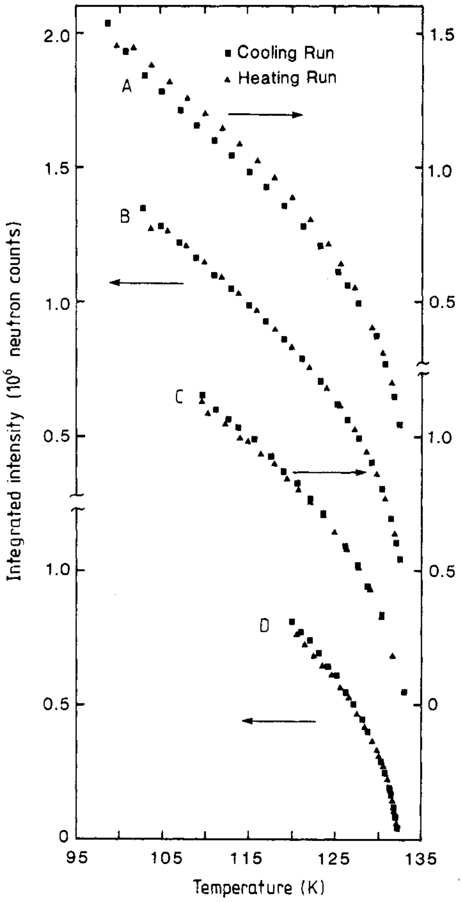

Figure 1. Integrated intensity of the magnetic (002-δ) Bragg reflection in holmium when temperature is cycled from above the Néel temperature, $T_N = 132.2$ K, to a low temperature T_0 and back: A, $T_0 = 98.4$ K, sample Ho IIb; B, $T_0 = 102.7$ K, sample Ho IIb; C, $T_0 = 109.5$ K, sample Ho IIa; D, $T_0 = 120.0$ K, sample Ho IIb.

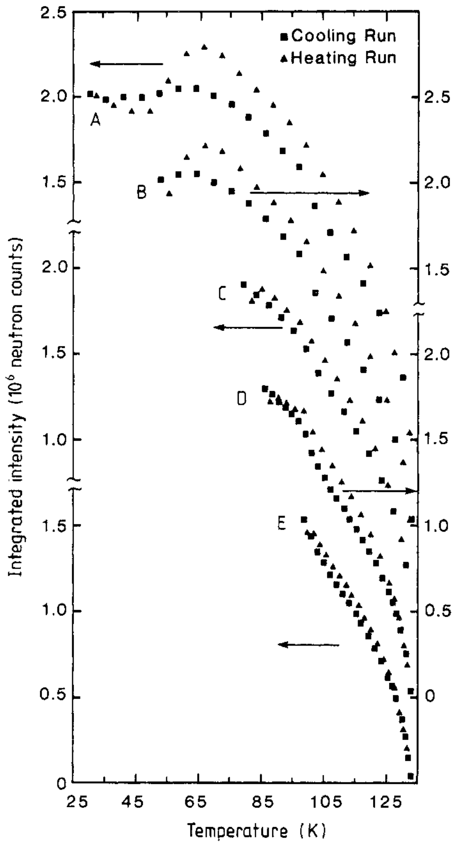

Figure 2. As figure 1, but for: A, $T_0 = 303$ K, sample Ho IIb; B, $T_0 = 52.5$ K, sample Ho IIb; C, $T_0 = 79.1$ K, sample Ho IIa; D, $T_0 = 88.0$ K, sample Ho IIb; E, $T_0 = 98.4$ K, sample Ho IIb.

decreasing temperature runs of figure 2, is associated with the lock-in transition at T_L. We note, however, that the nature of the hysteresis is sample dependent, since the less pure sample Ho I, as illustrated in figure 3 of Brits and du Plessis (1988), shows for $T_0 < 100$ K the opposite sign of hysteresis to that shown by the Ho II samples in figure 2, while for $T_0 > 100$ K, Ho I shows essentially no hysteresis. The two samples Ho IIa and Ho IIb, cut from the same ingot, appear to be identical in their magnetic scattering.

Further investigation of the hysteresis as it relates to the lock-in transition temperature at $T_L \simeq 100$ K is shown in figure 3. Successive temperature cycles of diminishing magnitude centred on $\simeq 100$ K show a decrease in the size of the hysteresis from about 5 K (measured at 100 K) to about 2 K.

Finally, in figure 4 we find that a significant amount of hysteresis occurs when T_0 is only about 4 K below T_N. The data within about 2 K below T_N are nevertheless not hysteretic. Results for successive heating and cooling runs through T_N (not shown)

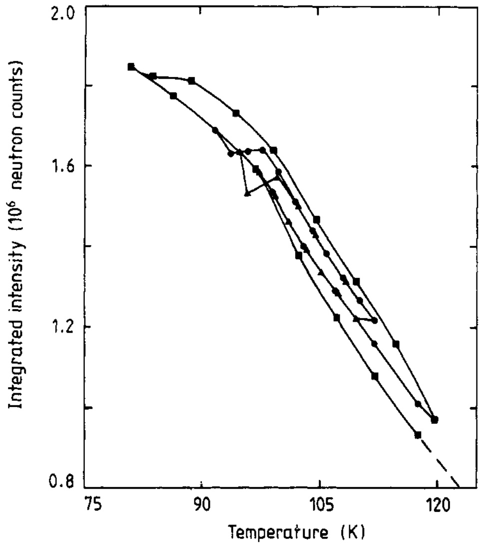

Figure 3. Integrated intensity of the magnetic (002-δ) Bragg reflection of sample Ho IIb depicting hysteresis loops for successive temperature cycles of diminishing magnitude centred on 100 K: ■, above $T_N \rightarrow$ 80.7 K $\rightarrow$ 119.6 K; ●, 119.6 K $\rightarrow$ 91.4 K $\rightarrow$ 110.0 K; ▲, 110.0 K $\rightarrow$ 94.5 K $\rightarrow$ 105.9 K. Smaller temperature cycles (not shown) do not decrease the width of the hysteresis loop which takes a limiting value of about 2 K.

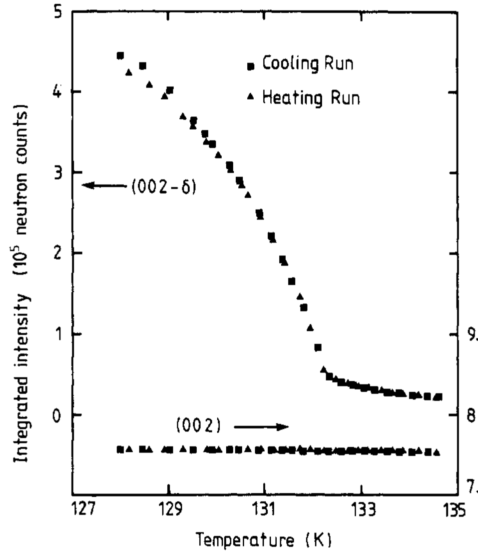

Figure 4. Integrated intensity of the magnetic (002-δ) Bragg reflection in sample Ho IIb when temperature is cycled from above the Néel temperature, T_N = 132.2 K, to T_0 = 128.0 K and back. The intensity of the (002) nuclear reflection is also shown. For clarity only every second data point is shown for the nuclear intensities as well as for satellite intensities for $T > T_N$.

indicate no temperature hysteresis in the position of T_N within our temperature resolution of better than ±0.02 K.

Support by the Foundation for Research Development, by the Atomic Energy Corporation of South Africa Ltd and by the Rand Afrikaans University is gratefully acknowledged. The visit by one of the authors (EF) to the Rand Afrikaans University was funded by the Foundation of Research Development and the Rand Afrikaans University. The work done by AMV and GAE was in partial fulfilment of the requirements of a PhD study for Rand Afrikaans University.

References

Bates S, Patterson C, McIntyre G J, Palmer S B, Mayer A, Cowley R A and Melville R 1988 *J. Phys. C: Solid State Phys.* **21** 4125
Brits G H F and du Plessis P de V 1988 *J. Phys. F: Met. Phys.* **18** 2659
Cowley R A and Bates S 1988 *J. Phys. C: Solid State Phys.* **21** 4113
Gibbs D, Moncton D E, D'Amico K L, Bohr J and Grier B H 1985 *Phys. Rev. Lett.* **55** 234
Koehler W C, Cable J W, Wilkinson M K and Wollan E O 1966 *Phys. Rev.* **151** 414
Lee M C, Treder R A and Levy M 1972 *Phys. Lett.* **38A** 499
McMillan W L 1976 *Phys. Rev.* B **14** 1496
Simpson A M, Jericho M H and Jain M C 1976 *Can. J. Phys.* **54** 1172
Simpson A M, Jericho M H and Roger W A 1979 *Can. J. Phys.* **57** 385
Tachiki M, Lee M C, Treder R A and Levy M 1974 *Solid State Commun.* **15** 1071
Vigren D T 1976 *Solid State Commun.* **18** 1599