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1992 J. Phys.: Condens. Matter 4 9927

(http://iopscience.iop.org/0953-8984/4/49/020)

J. Phys.: Condens. Matter 4 (1992) 9927–9930. Printed in the UK

Lock-ins in a *b*-axis field in holmium

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Received 7 August 1992

Abstract. We report neutron scattering observations of the stabilization of the locked-in phase with spiral pitch of $\frac{1}{4}c^*$ in holmium by a magnetic field applied along the *b*-axis. In contrast to a theoretical suggestion that stabilization of this phase by a *c*-axis field was due to imperfect alignment causing a small component of field in the basal plane, it is shown that the effect of a field of 3 T along the *b*-axis is to stabilize the phase over a somewhat smaller temperature range than the stabilized range in an identical field along the *c*-axis. The centre of the locked-in phase moves from 96 K in zero field to 103 K in a *b*-axis field of 3 T, while in a similar *c*-axis field the phase remains centred at around 96 K. We also observe a locked-in phase at a wave-vector of $\frac{5}{18}$, in the temperature range from 125 K to the Néel transition.

There is now quite a large body of neutron scattering work (reviewed by Jensen and Mackintosh (1991, 1992)) on the behaviour of holmium in magnetic fields applied along the *c*-axis, supported by magnetization (Willis *et al* 1990) and thermal expansion and magnetostriction studies (Steinitz *et al* 1987). Magnetization measurements (Ali *et al* 1989) with the magnetic field along the *b*-axis have revealed a rich variety of phases. Our recent neutron diffraction work has concentrated on the study of the stabilization of phases in a *c*-axis field in which the wave-vector, τ, of the spiral phase is locked in at commensurate values $\frac{1}{4}$ (Noakes *et al* 1990, Tindall *et al* 1991, Steinitz *et al* 1992 and Tindall *et al* 1992b) and $\frac{1}{5}$ (Tindall *et al* 1992a). Theoretical work (Plumer 1991) has suggested that the observed stabilization of these phases might be due to slight misalignment of the field, allowing a small, symmetry-breaking component to be present in the basal plane. This prompted us to examine the same crystal of holmium as used in our previous studies, in the same field of 3 T, but now applied along the *b*-axis.

The sample was mounted on the N5 triple-axis spectrometer at the NRU reactor at the Chalk River Laboratories of AECL Research, in a manner which permitted us to measure reflections in the $(h0l)$ plane. The field was applied along the *b*-axis using a superconducting magnet (Tennant *et al* 1989) which allows the neutrons 350° access in the horizontal plane. The measurements were made at zero energy transfer with a neutron energy of 8.225 THz. An analyser crystal was placed in front of the detector to eliminate higher-order contamination. The thermal expansion coefficient of holmium is very large in the region of interest and so the lattice parameters vary strongly with temperature, which causes all the Bragg peaks to shift in position as

0953-8984/92/499927+04$07.50

the temperature is changed. To minimize the effect of this on the determination of the magnetic ordering wave-vector, τ, we measured the nuclear (100) peak and its magnetic satellite (10τ) at each temperature, fitted a Gaussian curve to find the position of each, and then took the difference to obtain τ. The fitting error in the position of each peak was less than ± 0.0001 reciprocal lattice units (RLU) and we estimate the absolute values of τ to be good to ± 0.001 RLU. Figure 1 shows some results from this procedure.

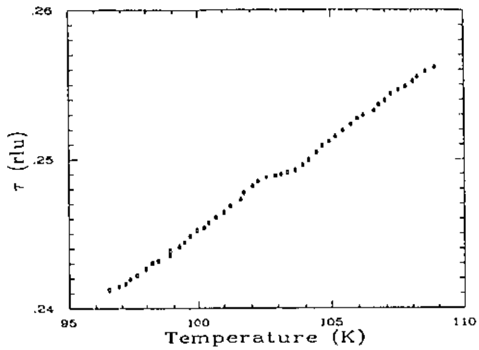

Figure 1. The variation of τ, obtained from the (10τ) satellite, with temperature in a b-axis magnetic field of 3 T in the vicinity of the lock-in at 103 K.

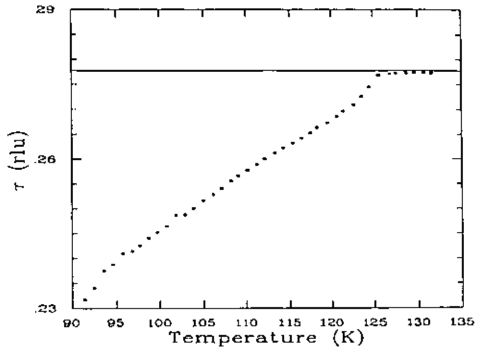

Figure 2. The variation of τ, obtained from the (10τ) satellite, with temperature in a b-axis magnetic field of 3 T from 90 K to the Néel temperature. A line drawn across shows $\tau = \frac{5}{18}$.

It can be readily seen in figure 1 that the temperature of the centre of the locked-in region at $\tau = \frac{1}{4}$ is raised from 96 K to 103 K by the application of a 3 T field along the b-axis. This indicates a certain increase in the stabilization energy, relative to the case with the field along the c-axis, where the transition remains centred on 96 K. On the other hand, it is apparent that the width of the locked-in region is about 1 K, significantly less than the width of 1.8 K we found for the corresponding region around 96 K in a 3 T field applied along the c-axis (Tindall *et al* 1991). This is in contrast to the conclusion which we would draw from the theoretical suggestion of Plumer (1991), that the stabilization of the locked-in phase by a c-axis field might only occur because of a small symmetry-breaking component of field in the basal plane, due to a slight misalignment of the sample with the field. It follows from Plumer's suggestion that the temperature width of the stabilized region should be greater if the entire field is oriented in the basal plane, along b, rather than just a small component, as in our c-axis measurements. This is just the opposite of what we observe. These b-axis measurements are consistent with those reported recently by Venter *et al* (1992), although Venter *et al* were unable to observe any lock-in in a c-axis field, indicating a significant difference between our respective results for the c-axis field which remains to be explained.

We have also observed a lock-in at $\tau = \frac{5}{18}$ in a b-axis field near the Néel temperature (132 K), as shown in figure 2. This helps to clarify the nature of the splitting of this transition, which we had discovered some time ago by dilatometry with a field applied along the a-axis (Steinitz *et al* 1987). The value of $\frac{5}{18}$ (i.e. 0.2777) is closer to our value of $\tau = 0.2773 \pm 0.001$ than the $\frac{8}{29}$ (i.e. 0.2759) suggested by Tarvin and Eckert (1979), and seems a more satisfying fraction in a hexagonal material. (Note that, as explained earlier, our fitting error for τ is about ± 0.0001 RLU, while its absolute error is estimated to be ± 0.001 RLU.) This would appear to be the first observation of a transition directly from the paramagnetic phase to a commensurate structure in the rare earths and will presumably have considerable efect on the understanding of the nature of the transition and its critical exponents.

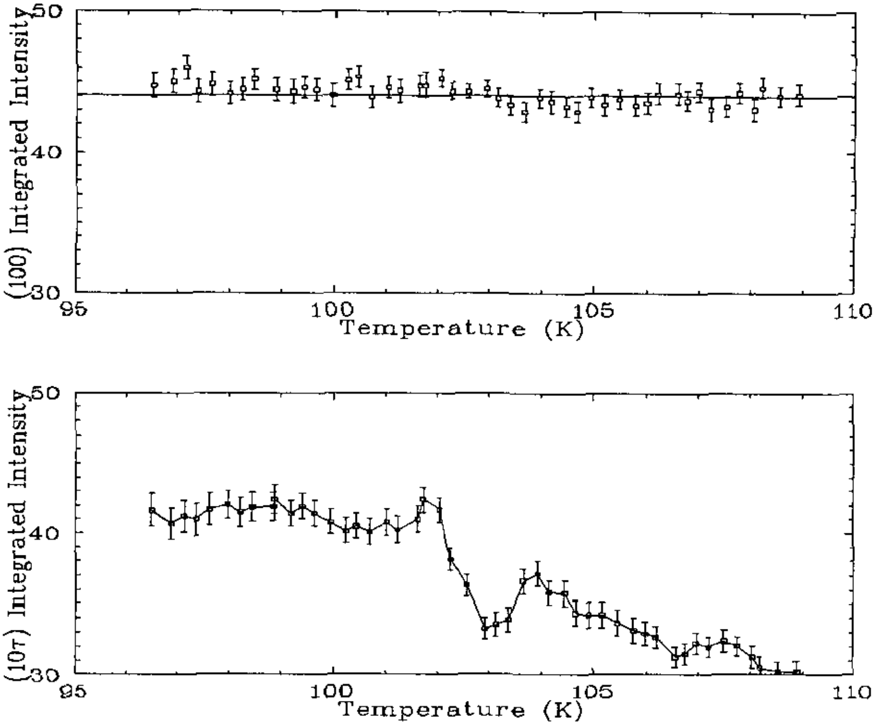

Figure 3. The lower figure shows the temperature variation of the integrated intensity (arbitrary units) of the (10τ) satellite in the vicinity of the 103 K lock-in. The upper figure shows the flat variation of the (100) nuclear peak through the same temperature region, for comparison.

Figure 3 shows the intensity of the (10τ) peak as it enters and leaves the locked-in region, in comparison with the constant intensity of the (100) nuclear peak. It is intriguing that the (10τ) satellite intensity again shows the same type of anomalous temperature variation that we have found in the vicinity of the lock-ins at 42 K and 96 K in a *c*-axis field (see, for example, figure 2 of Tindall *et al* (1991)). The explanation of these intensity anomalies remains a mystery.

So far, we have not observed a similar anomaly in the intensity of the (10τ) peak in the vicinity of 125 K as the wave-vector locks in at $\frac{5}{18}$. However, these measurements have, up to now, only been done with relatively coarse temperature resolution, and so we are proceeding with finer measurements in order to be able to make a clear statement about the existence of such an anomaly, or its absence.

In summary, we have discovered a further richness in the variety of phases shown by holmium in a magnetic field. Clearly, the work of mapping and identifying these phases has only just begun.

Acknowledgments

We are grateful to M J Pechan and C Stassis for the loan of the sample, to M L Plumer for helpful discussions, and to R LaPierre for assistance with data analysis and the figures. This work was supported in part by grants from the Natural Sciences and Engineering Research Council of Canada and the Canadian Institute for Neutron Scattering. We gratefully acknowledge the generous provision of facilities and skilled assistance by AECL Research, Chalk River.

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