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## Experimental verification of the significance of trigonal coupling for the $\frac{1}{4}$ lock-in in holmium

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### Abstract

In the region between 20 and 132 K, holmium is in a spiral anti-ferromagnetic phase, with the wave vector,  $\tau$ , of the spiral increasing from 0.16 to 0.28 reciprocal lattice units. Near simple values of  $\tau$  (such as  $\frac{1}{4}$ , which occurs near 100 K), a magnetic field can cause a “lock-in”, where the variation of  $\tau$  with temperature is arrested over a small temperature range. Recently, Jensen [Phys. Rev. B 54 (1996) 4021] predicted that the temperature width of this lock-in should be very strongly dependent on the magnetic field orientation in the plane containing the  $c$ - and  $b$ -axis. We have used the N5 triple-axis spectrometer at the NRU reactor, Chalk River, to measure the temperature width of the  $\tau = \frac{1}{4}$  lock-in in holmium as a function of the orientation of a 2.6 T magnetic field applied in the ( $c$ - $b$ ) plane. Our results are substantially in agreement with Jensen's prediction. © 1998 Elsevier Science B.V. All rights reserved.

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It has been known for many years [1] that the magnetic structure of holmium between 20 and 132 K is a spiral antiferromagnetic structure with a wave vector,  $\tau$ , which increases monotonically and apparently continuously, in the absence of a magnetic field, from 0.17 to 0.28. However, it has become increasingly clear that commensurate (or “lock-in”) effects are very important when magnetic fields are present. For example, we discovered [2]

that a  $c$ -axis magnetic field could produce a lock-in when  $\tau$  passed through  $\frac{1}{4}$  at 96 K. In this paper we concentrate on the behaviour of holmium near to its  $\tau = \frac{1}{4}$  lock-in in a 2.6 T magnetic field applied at various angles,  $\theta$  from the  $c$ -axis, lying in the plane containing the  $c$ - and  $b$ -axis.

Plumer [3] suggested that the observed stabilization of these phases in a  $c$ -axis field might be due to slight misalignment of the field. In order to test this, we [4] turned the field completely along the  $b$ -axis and found [4] similar behaviour. This appeared to lay to rest the hypothesis that a basal plane field component was necessary for lock-in behaviour. We

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shall see below that, in retrospect, it was a mistake to make such a large change to the field orientation, without covering intermediate angles!

Recently, Jensen [5] has presented a mean-field calculation of the stability of commensurate phases which includes not only the terms considered by Plumer [3], but also the trigonal coupling discovered by Simpson et al. [6,7]. This produced the remarkable prediction [5], shown in his fig. 7 (similar to the solid line in our Fig. 1), that the width of the lock-in region should be zero when the field is exactly along the  $c$ -axis ( $\theta = 0^\circ$ ) and rise *very* rapidly for small  $\theta$ ; giving, for example, a predicted lock-in width of nearly 3 K at  $\theta = 1^\circ$ . We thought this very unlikely to be the case, as we had consistently obtained a much smaller width in  $c$ -axis field and, of course, it is difficult to ensure that the field lies precisely in the desired crystallographic direction, so that misalignments of field and sample should have produced widely varying results for the lock-in width. Nevertheless, as measurements had only been made at field angles  $\theta = 0^\circ$  and  $90^\circ$ , we decided to make measurements at intermediate field angles.

The experiments were performed at Chalk River, using the N5 triple-axis neutron spectrometer. Elastically scattered neutrons with a wavelength of 0.237 nm were used. A single crystal of holmium ( $20 \times 10 \times 10 \text{ mm}^3$ ) was mounted in the M2 cryostat [8] that allows  $350^\circ$  access for the neutron beam. This clear access throughout most of the spectrometer plane is very important for this experiment, in which  $\theta$  varied over a wide range. The sample orientation was such that we observed ( $h0l$ ) reflections and, as described more fully in Ref. [9], we used the relative positions of (100) and (10 $\tau$ ) to determine an accurate value for  $\tau$ . In order to obtain accurate values for the boundaries of the locked-in region, we used a fine temperature step (0.2 K) and the Bragg peaks were scanned at 0.001 rlu intervals.

In order to compare our results with Jensen's predictions we, of course, need criteria for the boundaries of the locked-in region. In these experiments, as in all others that we have performed near lock-ins, when the locked-in region boundaries are crossed, there is a distinct rise in the intensity of the magnetic satellites. As we have noted previously

[10,11], the origin of this anomalous behaviour is still not understood, but it is very useful for marking the lock-ins. Also, we showed a long time ago [12] that taking the peaks in the temperature derivative of  $\tau$  would give the same values. We used these anomalous intensity peaks to determine the temperature of the lock-in boundaries. Our results are shown by the points in Fig. 1.

Jensen's published calculations [5] apply to a field of 3 T internal to the sample, but he has repeated these for our applied field value (assuming a spherical sample). These are plotted in Fig. 1. The agreement, while not perfect, is really astonishing. It seems clear that the trigonal coupling term is a key to understanding the complex behaviour of holmium. From the experimental point of view, while we have great regrets at not doing this experiment before (instead of going [4] directly from  $\theta = 0^\circ$  to  $90^\circ$ ), it is gratifying to note that our field alignment, which is now measured by a *small*  $c$ -axis lock-in width, is good to within a degree. It is apparent from Fig. 1 that additional work at a variety of field values is desirable to investigate the changeover, if any, from the Fan to the Helix phase.

We should like to express our deep gratitude to Jens Jensen, who provided the stimulus for these experiments and re-did his lengthy calculations for

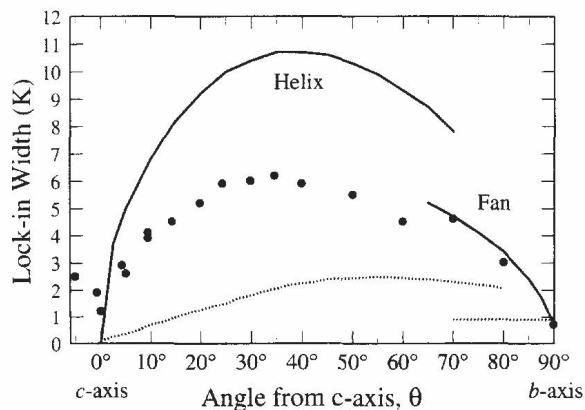


Fig. 1. The points (●) show our measurements of the width in temperature of the  $\frac{1}{4}$  lock-in versus the angle,  $\theta$ , of the applied 2.6 T magnetic field in the ( $c$ - $b$ ) plane. The lines show Jensen's model calculations with (solid line) and without (dotted line) the trigonal term.

us to make them applicable to our conditions. This work was supported by AECL, by NSERC and by CINS.

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