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On the magnetic phase diagram of erbium in a *c* axis magnetic field

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Abstract. The magnetic phase diagram of erbium with a magnetic field applied along the hexagonal *c* axis has been investigated using high-resolution neutron diffraction. We find that there are three distinct ordered phases occupying different regions of the field–temperature plane: a cone phase is found at high fields ($B \geqslant 2$ T) which has an incommensurate structure at temperatures above 40 K, but which becomes commensurate on cooling, first with a wavevector of $q_m = \frac{1}{4}c^*$ and then with $q_m = \frac{5}{21}c^*$; a nearly cycloidal structure in fields below 2 T, with a wavevector which passes through a series of commensurate values, with those structures having a net *c* axis moment being particularly favoured; and a phase at temperatures above 60 K which is mostly longitudinally modulated, but which also has a smaller basal plane component with a different wavevector. Our results are discussed with reference to a recent study of the zero-field structure of erbium by Cowley and Jensen, where it was proposed that the equivalence of the two magnetic sublattices in the chemical hexagonal unit cell is broken by an interaction with trigonal symmetry.

1. Introduction

There is renewed interest in the magnetic structure and excitations of the heavy rate earths, such as holmium and erbium, resulting from the discovery in the mid-eighties of long-period commensurable magnetic structures (Gibbs *et al* 1985, Cowley and Bates 1988). The existence of these phases is due to competition between the exchange interaction which favours in general a simple incommensurate ordering, and the crystal field interaction which favours commensurable structures. It has been further demonstrated more recently that the application of a magnetic field alters the balance between these two interactions, producing modified structures different from those found in zero field. For the case of holmium metal at low temperatures, the wavevector describing the magnetic order was found to form a devil's staircase when a field was applied along the *c* axis (Cowley *et al* 1991), whereas a field applied along the easy *b* axis produced a completely new type of magnetic order (Jensen and Mackintosh 1990, Jehan *et al* 1992).

The magnetic structure of erbium was first solved by Cable *et al* (1965) using neutron scattering techniques. They found that below the Néel temperature, T_N, of 87 K the magnetic moments were longitudinally modulated along the *c* axis with a

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wavevector which decreases from just above $q_m = 0.3c^*$ at T_N to around $q_m = \frac{2}{7}c^*$ at 54 K. At this temperature the basal plane moments order, with the same wavevector as the c axis moments, forming what they suggested to be a tilted helix. On further cooling the wavevector was observed to decrease continuously, until at $T_c = 18$ K a transition to a ferromagnetic cone structure occurs, where there is a net moment along the c axis and the moments in the basal plane rotate in successive planes about this axis. A more detailed study by Habenschuss *et al* (1974) did not substantially alter this picture. It was, however, challenged by Jensen (1976), who proposed that instead of forming a tilted helix between 54 K and T_c, the moments actually form a cycloidal structure in the a–c plane.

When long-range commensurable magnetic structures were discovered in erbium by Gibbs *et al* (1986), using high-resolution magnetic x-ray scattering, the cycloidal model provided a natural explanation of the observed commensurable wavevectors. It was proposed that in the cycloidal phase the magnetic structure is composed of blocks of three or four moments which are aligned predominantly parallel or anti-parallel to the positive c axis. (For example, the phase with $q_m = \frac{2}{7}c^*$ found at a temperature of approximately 50 K in zero field can be formed from one block of three spins along the c axis and one block of four spins along the $-c$ axis.) A comprehensive and detailed study, using neutron diffraction, of the commensurable structures found in erbium has recently been reported by Cowley and Jensen (1992). Their main result is that the observed scattering from the cycloidal phase can be explained only if the magnetic moments possess a small oscillatory component perpendicular to the a–c plane, and that in order for such a component to exist it is necessary for there to be a magnetic interaction which distinguishes between the two lattice sites in the chemical unit cell.

In this paper we present the results of our neutron diffraction measurements of the magnetic structures in erbium when a field is applied along the c axis. Similar neutron scattering measurements have been reported recently by Lin *et al* (1992). However, our measurements differ from theirs in that whereas they concentrated on deducing the phase diagram mainly by studying the principal magnetic satellites at many values of field and temperature, we have concentrated on measuring the higher-order satellites, particularly in the cycloidal phase. This has enabled us to develop structural models for several of the observed phases, and to compare these structures with those found in zero field.

In the next section we outline the experimental methods used for our study. Our results are presented in section three. We then discuss in section four our results in the light of the models of the zero-field structure proposed by Cowley and Jensen (1992), and also other work on the magnetic structure of erbium in a field. Finally, we present a phase diagram which incorporates not only our own results, but takes into account all other work done to date, especially that of Lin *et al* (1992).

2. Experimental details

Our experiments were performed using the triple-axis spectrometer D10 at the Institute Laue–Langevin, Grenoble, France. It was configured with a copper (002) monochromator and a pyrolytic graphite (002) analyser crystal; second-order contamination of the beam was suppressed using a graphite filter. The spectrometer was set to detect elastically scattered neutrons with an energy of 14 meV, giving a wavevector transfer resolution in the scattering plane of approximately $0.005c^*$, and an energy resolution of 0.3 meV.

The single crystal of erbium (a cube of length 5 mm) used in our study was supplied by the University of Birmingham, and had been investigated using ultrasound (Eccleston and Palmer 1992). It was mounted with the $(H0L)$ plane in the scattering plane inside a horizontal field cryomagnet. We estimate that the field was applied along the c axis to within ±2°.

Unfortunately, the construction of the magnet severely restricted the accessible regions of reciprocal space. This meant that we were limited to performing scans of the component of the neutron wavevector transfer $Q_c = Lc^*$ along c^* from $L = 0.95$ to $L = 2.05$ and from $L = 2.8$ to $L = 3.4$ near $(10L)$. As neutron scattering is sensitive only to the magnetic moment perpendicular to the wavevector transfer, we were able to obtain accurate information about the components of the moments in the basal plane, and less accurate information on the behaviour of the component moments along the c axis.

Inside the cryomagnet the temperature of the sample could be controlled between 2 K and 100 K with a precision of 0.1 K, and a maximum field applied of up to 4 T (±0.1 T). The data were collected as the sample was cooled from above T_N in fields of 0, 1, 2 and 4 T. We note the results presented in the next section have not been corrected for demagnetizing fields.

3. The results

3.1. *The high-temperature phase*

At high temperatures, just below the zero-field ordering temperature of T_N (approximately 87 K), and in a field of 1 T a series of scans of the magnetic satellites around (002) and (103) revealed a surprising feature. From the zero-field measurements it was expected (Cable *et al* 1965) that the basal plane moments should not order until the temperature is reduced below 54 K. However, as can be seen from figure 1, a weak magnetic satellite was observed around (002) above this temperature, indicating ordering in the basal plane. The width of this peak, although greater than that of the nuclear peak at $L = 2.0$, was found to be independent of temperature, and cannot therefore be critical scattering. The intensity of this peak was found to increase slowly with decreasing temperature until there was a sharp increase at approximately 56 K, as shown in figure 2(a). The wavevector describing the ordering of the basal plane moments, as deduced from the separation of the magnetic peak from the nuclear peak at $L = 2.0$, above the temperature of the sharp increase in intensity, is shown in figure 2(b). It can be seen that it remains roughly constant with $q_m = 0.295(2)c^*$ over a temperature interval of more than 20 K, and that at about 56 K it jumps to a value of 0.286, which is very close to the commensurate value of $\frac{2}{7}c^*$. Over the same range of temperature the position of the magnetic peak in the scattering around (103), which arises partly from the component of the moment in the basal plane and partly from the c axis component, is significantly less than the wavevector of the scattering observed around (002). However, below 56 K the wavevectors of the scattering around (002) and (103) are essentially identical, and equal within error to the commensurate value of $\frac{2}{7}c^*$. Figure 3 shows the temperature dependence of the magnetic scattering intensity near (103), which increases steadily with decreasing temperature.

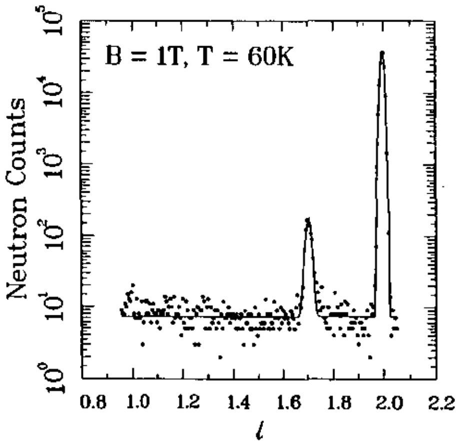

Figure 1. The neutron scattering observed with the neutron wavevector transfer along $(00L)$ at 60 K in an applied field of 1 T. The peak at $L = 2$ is from nuclear scattering and that at $L \approx 1.7$ from magnetic scattering. The solid line is a fit to two Gaussians, including a flat background.

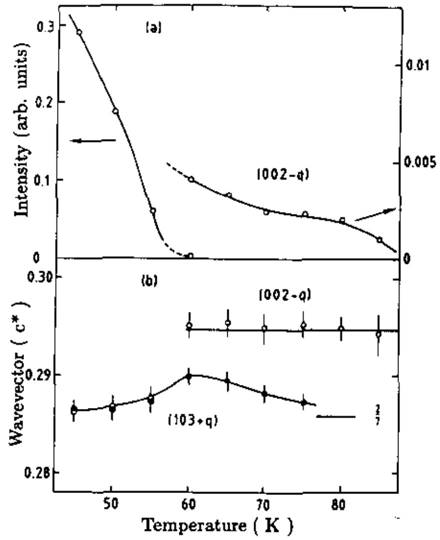

Figure 2. (a) The intensity of the magnetic satellite observed near (002); (b) the wavevector q_m of the magnetic satellites observed around (002) (open circles) and around (103) (closed circles). Note: the scattering at (103) has components from both the basal plane and longitudinal magnetic moments, whereas the scattering near (002) arises from the basal plane moments only.

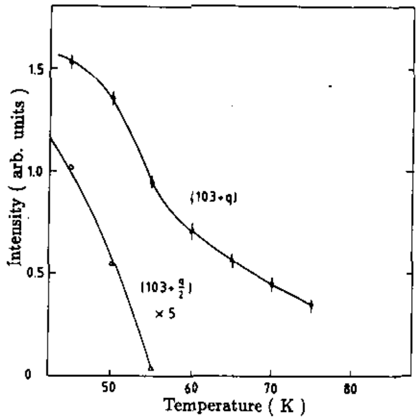

Figure 3. The intensities of the magnetic satellites observed in scans along $(10L)$ near $L = 3$. Full circles: the principal satellite at $L = 3 + q_m/c^*$; open triangles: the subsidiary satellite at $L = 3 + q_m/2c^*$.

These results suggest that at high temperatures the ordered phase of erbium in a field does not have the moments entirely longitudinally modulated along *c*, but rather that a small component of the moment, with a different modulation, is ordered in the basal plane. This latter component probably arises from a helical ordering in the basal plane.

3.2. The cycloidal phase

For fields less than 2 T, and for temperatures less than approximately 56 K, the magnetic structure of erbium is found to become cycloidal. One of the most noticeable effects of applying a field is that the temperature intervals over which the wavevector of the cycloidal phase is commensurate are enhanced at the expense of those in which it is incommensurate. In fact we found no evidence for incommensurate structures in fields of 1 T or larger, while the commensurate structures which have a ferromagnetic moment parallel to the c axis, namely, $q_m = \frac{2}{7}$, $\frac{4}{15}$ and $\frac{6}{23}$ (in units of c^*) are stable over a wider range of temperatures in a field than in zero field. In addition, and unlike in the previous study (Lin *et al* 1992), there is evidence for phase coexistence near the boundaries separating two commensurate phases. This is illustrated in figure 4, where all of the peaks can be indexed either with $L = n/11$ or with $L = m/7$ (n and m integers). This result suggests that at this value of field and temperature, the crystal is in an admixture of phases with underlying wavevectors of $q_m = \frac{2}{7}c^*$ and $q_m = \frac{3}{11}c^*$. If, however, an attempt is made to ascribe a particular wavevector to the structure solely from the position of the most intense magnetic peak near $L = 1.7$, then it would be incorrectly inferred that the wavevector is incommensurate.

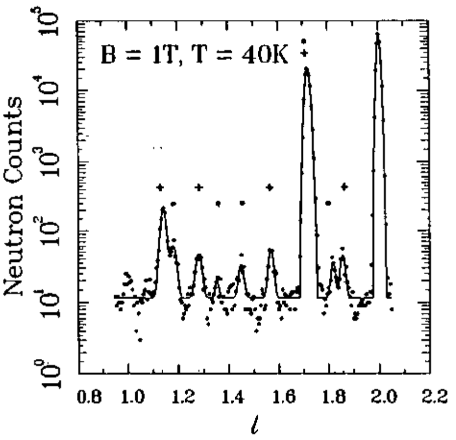

Figure 4. The neutron scattering observed from the cycloidal phase at 40 K in a field of 1 T for a scan of the wavevector transfer along $(00L)$. The solid line is a fit to a sum of Gaussians, including a flat background. Note: all of the weak magnetic satellites can be indexed as arising from either the $q_m = \frac{2}{7}c^*$ (crosses) or $q_m = \frac{3}{11}c^*$ (dots) phases.

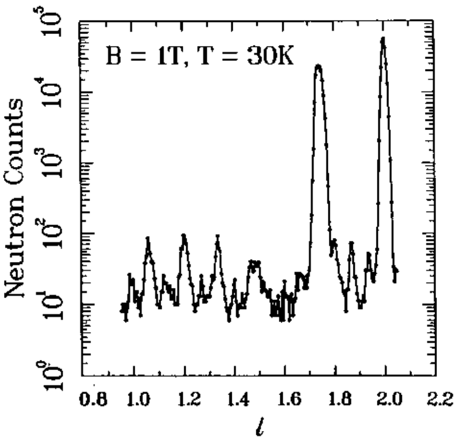

Figure 5. The scattering observed from the cycloidal phase of erbium at 30 K in an applied field of 1 T, in a scan of the wavevector transfer along $[00L]$. The higher-order harmonics can all be indexed with $L = n/15$: the stronger peaks have n even, and the weaker ones n odd.

Phase coexistence arises only close to the phase boundaries, and for most values of field and temperature a single dominant phase is found. This is illustrated in figure 5 where we show the scattering observed at a temperature of 30 K and in a field of 1 T. It is evident that the most intense magnetic satellites occur when $L = n/15$ with n an even integer, but that there are in addition weaker peaks with n an odd integer. The former can be identified as being characteristic of a commensurate cycloid with $q_m = \frac{4}{15}c^*$, whereas the latter arise from an oscillatory component of the moment perpendicular to the plane of the cycloid, as described in detail by Cowley and Jensen (1992). Following the same techniques described by these authors for deducing the structures found in zero field, we have fitted the data shown in figure 5 to obtain the projection of the basal plane moments in this fifteen-layer structure. The results of this exercise are shown in figure 6, where they are compared with the results from the zero-field phase with the same wavevector. In making this comparison between the two experiments we have to allow for the fact that there is a certain degree of uncertainty in the normalization procedures, because the two sets of experiments were performed on different crystals using different instruments. This fact prevents us from drawing any firm quantitative conclusions. However, it is clear that the application of the field does indeed reduce the basal plane components in the plane of the cycloid (labelled S_x in figure 6) relative to their value in zero field. Also shown in figure 6 are the components of the moments in the basal plane, S_y perpendicular to the cycloidal plane. The weakness of the peaks which arise from the out-of-plane component (the $L = n/11$ peaks with n odd in figure 5) necessarily makes accurate modeling of this component difficult. In spite of this, it is evident that this component has the same qualitative features as found in zero field.

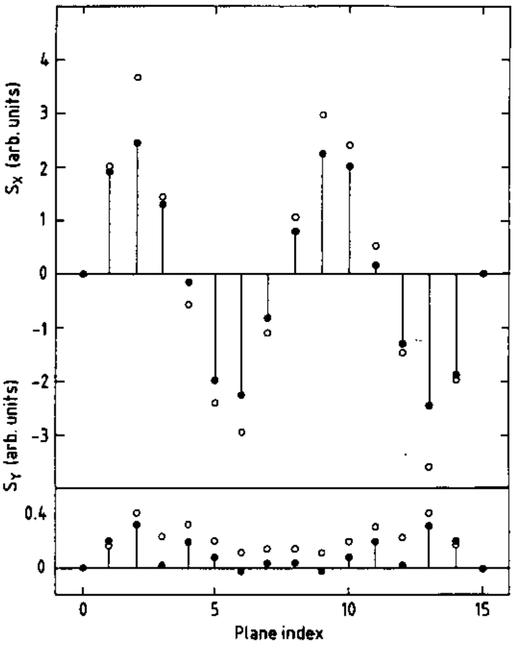

Figure 6. A comparison of the basal plane moments of the $q_m = \frac{4}{15}c^*$ structure in a field of 1 T (filled circles) with the zero-field structure having the same wavevector (open circles). The zero-field data was taken from Cowley and Jensen (1992). Note: the components S_x in the plane of the cycloid have a different periodicity to the components of the magnetic moment, S_y, perpendicular to the a–c plane.

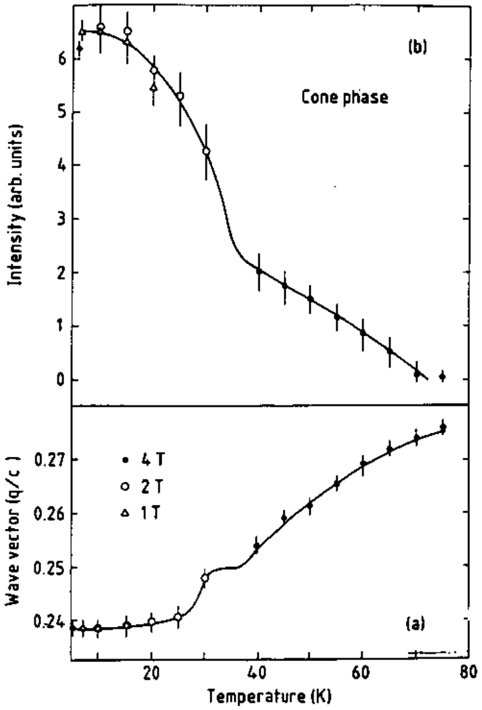

Figure 7. (a) The wavevector dependence of the basal plane moments in the cone phase for various applied fields. (b) The temperature dependence of the intensities of the principal magnetic satellite at $L = 2 - q_m/c^*$. The lines are guides to the eye, but in (a) we have indicated the region where a lock-in to $q_m = \frac{1}{4}c^*$ was observed by Lin *et al* (1992).

3.3. *The cone phase*

In zero field, below $T_c = 18$ K the magnetic moments in erbium form a cone structure, with a net ferromagnetic moment along the c axis and with the components of the moments in the basal plane forming a helix. We have studied the stability of this structure at low temperatures by applying fields up to 4 T along the c axis. (Here we remind the reader that, as discussed in section 2, we were prevented by the construction of the magnet from obtaining detailed information about the c axis moments, and so what follows in the rest of this section pertains to the basal plane components only.)

Our results for the cone phase are summarized in figures 7(a) and 7(b), where we show the temperature and field dependence of the wavevector and intensity respectively. For temperatures below 15 K, the structure of the cone phase was found to be essentially independent of field; even in a field of 4 T the wavevector remained at $\frac{5}{21}c^*$, its zero-field value at these temperatures. At temperatures above about 37 K with the maximum field of 4 T applied it is found that the wavevector is incommensurate. Between this incommensurate phase and the cone phase, another commensurate phase with $q_m = \frac{1}{4}c^*$ has been discovered by Lin *et al* (1992). Our data in this regime are sparse but nonetheless consistent with their findings.

With regard to the intensity of the scattering, it can be seen in figure 7(b) that on cooling the sample in the high-field incommensurate phase the intensity increases smoothly. However, as the wavevector becomes commensurate, first with $q_m = \frac{1}{4}c^*$ and then with $q_m = \frac{5}{21}c^*$, there is a more rapid increase in the magnetic scattering. In the incommensurate phase no peaks other than the main satellite at $L = 2 - q_m$ is observed, indicating that the basal plane moments are ordered in a simple helix. Higher harmonics are observed, however, at all values of field in the commensurate phases. When $q_m = \frac{1}{4}c^*$ an additional peak is found at $L = 1.5$, which is 0.26% of the intensity of the main satellite at $L = 1.75$: no scattering for this structure is observed at $L = 1.25$ or 1. In figure 8 we show a structure which is compatible with these observations.

In figure 9 we show the scattering from the cone phase ($q_m = \frac{5}{21}c^*$) at a temperature of 5 K in a field of 4 T. Our attempts to develop a structural model to account for this scattering have only been partially successful, and overall the detailed structure of this phase remains elusive. We know that the weak magnetic peaks which are observed in addition to the main satellite at $L = \frac{37}{21}$ cannot be due to a distortion of the structure arising from the sixfold basal plane anisotropy, as this would produce fifth and seventh harmonics at $L = \frac{25}{21}$ and $\frac{35}{21}$ which are not seen. The peaks found at $L = \frac{31}{21}$ and $L = \frac{41}{21}$ probably arise from the bi-quadratic trigonal coupling invoked by Cowley and Jensen (1992) to explain their zero-field data. The origin of the other peaks is unknown.

4. Discussion and conclusions

We shall start this section by briefly considering some of the experimental problems inherent in the type of experiment described in this paper, and which ultimately limit the information that can be extracted from the observed scattering. In order to solve accurately the complex magnetic structures of erbium it is necessary to measure the position and intensity of the primary satellite and of all the higher-order Bragg

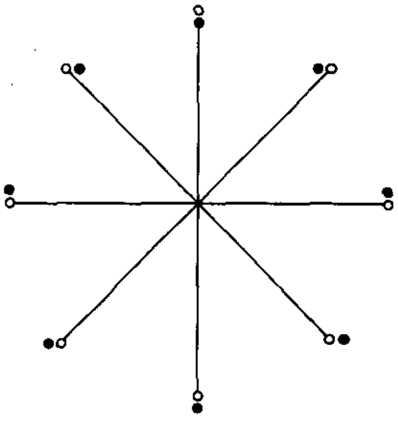

Figure 8. A projection of the moments into the basal plane for the $q_m = \frac{1}{4}c^*$ cone phase. The open circles are positioned for an undistorted structure with this wavevector, whereas the filled circles represent the projection of a structure consistent with our data.

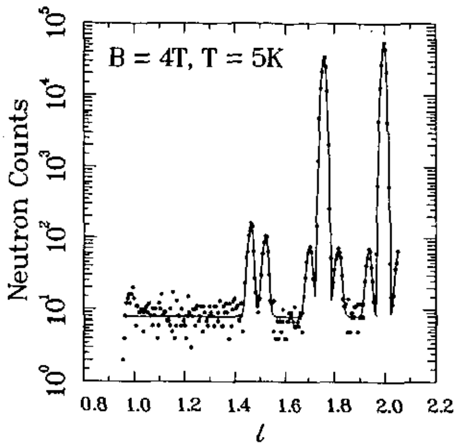

Figure 9. The scattering from the $q_m = \frac{5}{21}c^*$ cone phase observed along $(00L)$ at 5 K in a field of 4 T. Note: the weak magnetic peaks (intensity less than 10^3 counts) arise from distortions in the underlying helical structure and are discussed in the text. The solid line is a fit to a sum of Gaussians plus a flat background.

reflections. The intensities, in particular, pose a serious problem, as the more subtle and interesting structural distortions found in erbium produce weak Bragg peaks, and it is found that the intensity must be measured over at least four decades. The well-known procedures which would normally be used for checking the accuracy of the measured intensities, such as performing Renniger scans to check for multiple scattering processes, could not, however, be employed in our study. The reason for this is that our access of reciprocal space was severely limited by the design of the magnet, as described in section 3. Some of our conclusions are, therefore, necessarily tentative.

We summarize our results by presenting in figure 10 the phase diagram of erbium in a c axis field. In order to synthesize this diagram we have taken into account the neutron scattering results of Lin *et al* (1992), the magnetization measurements of Flippen (1964), Rhyne *et al* (1968) and Gama and Foglio (1988), the magnetostriction measurements of Rhyne and Legvold (1965) and the ultrasonic measurements of Eccleston and Palmer (1992). There are essentially three different types of ordered phase. Our phase diagram is in broad agreement with the one recently published by Lin *et al* (1992), although it does differ in some details concerning the position of the phase boundaries.

At high fields the magnetic moments form a c axis cone, in which the components of the moments in the basal plane at high temperatures are ordered in a simple incommensurable helix. When the temperature is reduced below approximately 40 K the helix distorts and the ordering wavevector locks into a value of $\frac{1}{4}c^*$, followed by an ultimate transition to $\frac{5}{21}c^*$. The reason why the structure locks into these particular wavevectors is not completely understood. The sixfold basal plane anisotropy has a negligible influence, in contradiction to the spin-slip model of Gibbs *et al* (1986), because if it was significant, then fifth and seventh harmonics of the

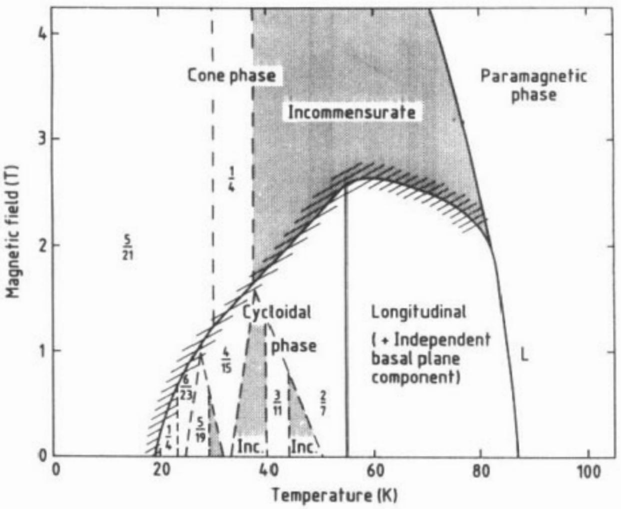

Figure 10. The magnetic phase diagram of erbium with a magnetic field applied along the *c* axis. The boundaries between the main phases are shown by solid lines, and the boundaries between different commensurate phases or commensurate and incommensurate phases are represented by dashed lines. The diagonally hatched boundary separating the cone phase from the other phases indicates a region of phase coexistence. Key: Inc. = incommensurate phase.

ordering wavevector would be observed, as are found in holmium.

For the cycloidal phase the applied field has the simple effect of stabilizing those commensurable structures which have a net *c* axis moment ($q_m = \frac{2}{7}$, $\frac{4}{21}$ and $\frac{6}{23}$, in units of c^*). The exact boundaries delimiting the cycloidal phase from the cone phase, and especially the different cycloidal phases from each other, are uncertain as we have shown that in these regions there is phase coexistence. Whether this is an intrinsic property of the magnetism in erbium, or whether it is extrinsic, possibly arising from either pinning at defects or non-uniform demagnetizing fields, will have to be left as an open question until a more detailed study is performed. In the cycloidal region, the scattering is characterized by the presence of many higher-order satellites. By including these satellites in our analysis we have shown that the cycloidal structures formed in a field are similar to those found in zero field, but with a reduced magnitude of the basal plane magnetic moment. In addition distortions of the cycloidal phase out of its plane are significant.

At high temperature, for fields less than 2 T, we find that there is a longitudinally modulated structure, with a sharp boundary between it and the paramagnetic phase (the line labelled L in figure 10). In addition we find that above approximately 56 K the components of the moments in the basal plane order with a slightly different wavevector to the longitudinal components. There is some evidence for the same decoupling of the moments in zero field (Cowley 1992), albeit existing over a much narrower temperature range. This effect was not reported by Lin *et al* (1992). The observation of two ordering wavevectors in erbium challenges the assumption that the exchange in erbium is purely Heisenberg in nature, as the two wavevectors imply that the longitudinal and basal plane parts of the exchange have maxima at different wavevectors and that the ordering of the moments occurs at different temperatures.

More detailed experiments focusing on this region of the phase diagram are planned to clarify these outstanding problems.

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