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Non-Planar Magnetic Structures and Trigonal Interactions in Erbium.

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Abstract. – The magnetic structures of erbium metal have been studied using a high-resolution elastic-scattering technique. The results show that the elliptical cycloidal structures between 55 K and 18 K are not planar. A comparison of the experimental intensities with the predictions of a mean-field model shows that these structures are distorted by two-ion interactions of trigonal symmetry, which distinguish between the two h.c.p. sublattices. The resulting structures are wobbling cycloids, in which there is a b-axis moment perpendicular to the cycloidal (a, c)-plane, oscillating with a period different from that of the basic structure. In the cone phase below 18 K, the trigonal interactions introduce bunching effects around alternating a directions in a threefold symmetric pattern.

The magnetic structures of erbium were first thoroughly studied by Cable *et al.* [1] who found three distinctly different magnetically ordered phases: i) between $T_N \simeq 84$ K and $T'_N \simeq 55$ K, a sinusoidal longitudinal phase with a modulation period of approximately 7 layers along the c-axis; ii) between T'_N and $T_C \simeq 18$ K, a modulated ordering of the basal-plane and longitudinal components, which was later shown [2] most likely to be a planar-cycloidal phase in which the moments lie in a plane containing the c-axis and one direction in the basal plane; iii) for temperatures below T_C, a cone structure with a ferromagnetic moment along the c-axis and an opening angle of about 28°, and an ordering wave vector of $\sim (5/21)\,\tau_c$, where $\tau_c = (0, 0, 1)\,2\pi/c$. This letter is primarily concerned with the structure of the cycloidal phase, in which both the basal-plane and longitudinal moments are modulated. We also discuss briefly the structure in the cone phase.

The neutron scattering measurements were made with a triple-axis crystal spectrometer at the HFIR reactor at Oak Ridge National Laboratory in a similar way to earlier experiments on holmium [3]. The incident neutron energy was fixed at 14.8 meV, a pyrolytic graphite filter was used to reduce higher-order contaminant neutrons, the horizontal collimations from reactor to counter were 20′, 20′, 20′, open, and pyrolite graphite was used for both monochromator and analyser crystals. Scans were performed by varying the wave vector transfer along the directions [00L] and [10L]. The isotopically enriched single crystal

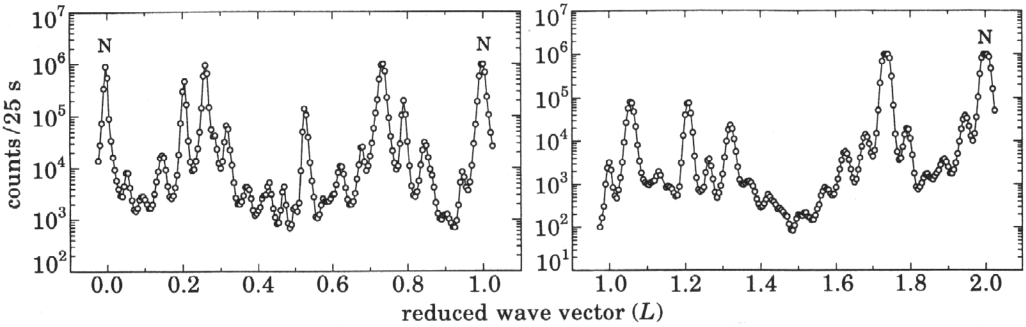

Fig. 1. – The neutron scattering observed from erbium at 29 K. The left part of the figure shows a scan along [10*L*] and the lower one along [00*L*]. The peaks marked N are from nuclear scattering and the remainder are magnetic in origin.

of erbium was that used in earlier measurements of the spin waves [4], and it was mounted in a variable-temperature cryostat with the scattering plane perpendicular to (010). Measurements were taken at a variety of temperatures within the intermediate phase, and typical results at 29 K are shown in fig. 1. In this scan, in common with those taken at different temperatures, there are a large number of harmonics, showing that the structures are not simply sinusoidally modulated but are very strongly distorted. Furthermore, the results can generally be described as arising from long-period commensurable structures. In a few cases the crystal exhibited two commensurable structures at the same time. These long-period structures provide striking confirmation of the experiments and conclusions of Gibbs *et al.* [5], who suggested that, in the intermediate phase ii), the structures arise from regular arrangements of 3 or 4 layers of moments aligned with an alternating positive or negative component along the *c*-axis. The results at 29 K shown in fig. 1 have peaks for $L = n/19$ with, along [10*L*], maxima at $n = 5$ and 14. These peaks arise largely from the components of the moments along the *c*-axis and are consistent with the proposed 2(44443) structure, in which the numbers in the bracket give the size of the blocks aligned similarly with respect to the *c*-axis. A commensurable period must contain an even number of up/down blocks, and the factor 2 in front of the sequence (44443) means that it is repeated twice in one such period.

The peaks observed in the [00*L*] scan have exactly the same wave vectors, showing that the basal-plane component and longitudinal component have the same modulation and hence are in phase coherence. In general, this arises only in the cycloidal structure [2]. Similar results have been obtained for the 2(43), 2(443), 2(4443) and 2(444443) structures and are reported elsewhere [6].

In order to determine the structures in more detail, a number of corrections must be applied to the results. Firstly, the use of a large crystal means that extinction reduces the most intense peaks substantially. The results of Habenschuss *et al.* [7] were therefore used to obtain the relative intensities of the strong peaks and for relating these intensities to the scattering functions. Multiple scattering is always a problem in determining the weak intensities. For some peaks, the crystal was rotated about the scattering vector to investigate whether they arise from multiple scattering, and the results from different but equivalent regions in reciprocal space were compared, in order to obtain a reliable set of results. These were then corrected for the form factor and geometrical factors to obtain the intensities of the peaks. The uncertainty in these corrections can give rise to errors in the relative intensities by factors of the order 2-3, especially when they differ by several orders of magnitude.

Despite these problems, the data could be analysed to deduce the structure of the cycloidal phase in detail. It became immediately apparent that the planar cycloidal model, which does not reflect the different orientations of the two hexagonal sublattices, must be modified. If the effective lattice periodicity is $c/2$ along the c-axis, then for the 38-layered cycloidal structure, the peaks at $L = n/19$ with n even should have zero intensity in the scans along $[00L]$, in contrast to the observations. This discrepancy can only be satisfactorily explained if there are magnetic interactions which distinguish between the two different sublattices of the h.c.p. lattice. One lowest-order term which does so is

$$\mathscr{H}_3 = \sum_{ij} K_{31}^{21}(ij)[O_3^2(i)\,J_{yi} + O_3^{-2}(i)\,J_{xj}], \tag{1}$$

where the Stevens operators are given by $O_3^{\pm 2} = (1/2)(J_z O_2^{\pm 2} + O_2^{\pm 2} J_z)$, with $O_2^2 = J_x^2 - J_y^2$ and $O_2^{-2} = J_x J_y + J_y J_x$. The x-, y- and z-axis are taken along the orthogonal a, b and c directions of the crystal. This term changes sign from one sublattice to the next and furthermore produces a component of the magnetization perpendicular to the plane of a cycloidal structure, if this plane is parallel with the (x, z)-plane. This is seen most easily by noting that, in this case, eq. (1) and mean-field theory give a molecular field in the y-direction which varies from site to site with a periodicity determined by the wave vectors $2\boldsymbol{q}_c \pm \boldsymbol{q}_c + \tau_c$, where $\boldsymbol{q}_c$ is the wave vector of the cycloidal structure. In erbium, the a-axis is the easy planar axis, and these peaks occur for the 2(44443) phase for $L = n/19$ when $n = 24$ and $n = 34$, both of which are present in the experimental results shown in fig. 1, as well as other peaks with even n. We suggest therefore that these even-n peaks result from deviations from the planar cycloidal structure and that these deviations derive from the trigonal interaction between the moments given by eq. (1).

Further evidence for these conclusions has been obtained from a mean-field analysis of a detailed microscopic model of the interactions in erbium. The mean-field model, which is a further development of that used earlier [2], utilizes measurements of the magnetization and low-temperature spin-waves to determine the anisotropy and the exchange parameters for erbium. The trigonal interactions in eq. (1) were then introduced between nearest, next-nearest, and third-nearest planes and the magnitudes adjusted through a comparison between the calculated and experimental peak intensities at even n in the $[00L]$ scans. The details of these calculations are given elsewhere [6]. In all of the calculated 2(4...3) structures, the ordered moments may be characterised by the expansions

$$\begin{cases} \langle J_{xp} \rangle = \sum\limits_{s=1,3,\ldots} (-1)^{(s-1)/2} A_x(s) \sin[s q_c p c/2], \\ \langle J_{yp} \rangle = \sum\limits_{s=1,3,\ldots} (-1)^{(s-1)/2} A_y(s) \sin[s(q_c + \tau_c) p c/2], \\ \langle J_{zp} \rangle = \sum\limits_{s=1,3,\ldots} (-1)^{(s-1)/2} A_z(s) \cos[s q_c p c/2], \end{cases} \tag{2}$$

where all the amplitudes $A_\alpha(s)$ are positive. p is the numbering of the layers in the period, and the sums over s include terms up to half the number of layers in one commensurable period.

The scattering functions $I(00L)$ and $I(10L)$ for the 2(44443) structure, as determined by the model and as derived from the corrected experimental intensities, are compared in fig. 2. The corresponding amplitudes $A_\alpha(s)$ obtained from the scattering functions in fig. 2 are given in table I. By using the phases predicted by the mean-field model, as given in eq. (2), the

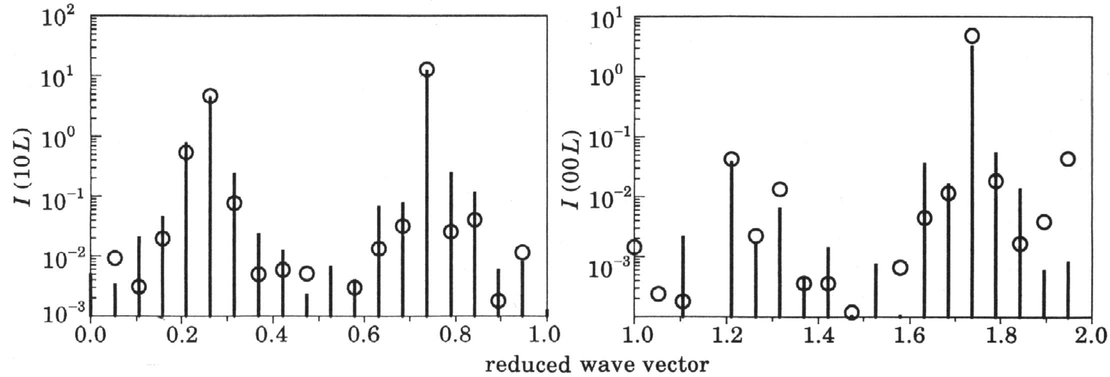

Fig. 2. – The correlation functions $I(00L)$ and $I(10L)$ of the commensurable magnetic 2(44443) structure in erbium at 29 K, in the intermediate cycloidal phase. The lines are derived from the calculations of the structures, and the circles are the experimental results determined from the neutron scattering intensities in fig. 1 combined with the results of Habenschuss *et al.* [7], as explained in the text. The logarithmic scales in the two plots differ by one decade.

TABLE I. – *The amplitudes* $A_\alpha(s)$ *for the* 2(44443) *structure at* 29 K. *The first column in each of the three cases gives the values derived from the mean-field model and the second column gives the experimental values, corresponding, respectively, to the solid lines and the circles in fig.* 2.

s	$A_x(s)$		$A_y(s)$		$A_z(s)$	
1	3.65	4.41	0.09	0.09	8.36	8.21
3	0.40	0.41	0.48	0.27	2.07	1.02
5	0.16	0.23	0.26	0.22	1.15	0.65
7	0.24	0.08	0.01	0.00	0.86	0.54
9	0.39	0.13	0.04	0.04	0.57	0.26
11	0.10	0.03	0.05	0.13	0.33	0.10
13	0.08	0.04	0.02	0.05	0.26	0.20
15	0.06	0.42	0.00	0.03	0.23	0.10
17	0.06	0.00	0.02	0.02	0.20	0.15
19	0.00	—	0.00	—	0.08	—

magnetic structure may be determined from the scattering functions, and fig. 3 shows the 2(44443) structure derived from the experiments, compared with the mean-field model. The experimental and theoretical structures are very similar. The main difference is that the configuration of moments obtained from the experimental intensities is somewhat more open than that deduced from the mean-field model.

The agreement between the observed diffraction intensities and the scattering functions derived from the magnetic structures predicted by the microscopic model is satisfactory. We therefore conclude that, for the structures of the intermediate phase in erbium, the commensurable basis is the one proposed by Gibbs *et al.* [5]. Furthermore, we find that to a first approximation the hodograph of the moments is an elliptically polarized cycloid, whose plane lies in an (a, c)-plane of the crystal. The experiments, however, show clearly that the magnetic structures depend on the two different orientations of the hexagonal layers in the h.c.p. lattice. This can only be satisfactorily explained by trigonal couplings in the magnetic Hamiltonian, which have a magnitude which is a substantial fraction of the isotropic ex-

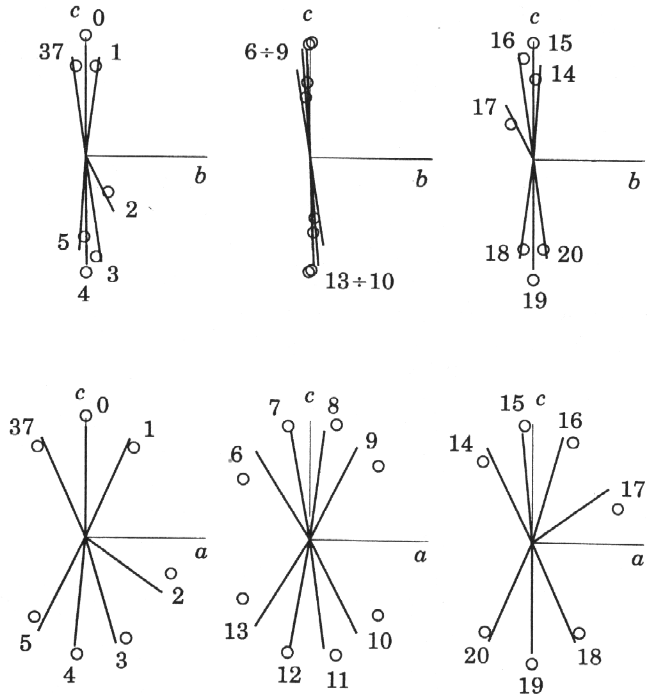

Fig. 3. – The structure of the 2(44443) phase. The lines are the calculated angular moments projected, respectively, onto the (b, c)- and the perpendicular (a, c)-plane in the upper and lower part of the figure. The circles are the corresponding results deduced from the experimental intensities shown in fig. 2 or the amplitudes given in table I. The experimental peak at (001) is neglected. The (a, b, c)-axes shown are of length $J = 7.5$, corresponding to the saturated 4f-moment, but the b-components have been multiplied by a factor of two. The moments are labelled by $p = 0 \div 37$ in accordance with the phase convention of eq. (2). The moments in the remaining part of the 38-layered commensurable period are related to those shown as follows: for the moments in the p-th and the $(p+19)$-th layer, the b-components are the same whereas the a- and c-components have their signs changed.

change interaction. The result is a wobbling cycloid, in which there is an oscillating moment in the b-direction whose period differs from that of the basic cycloidal structure.

In the cone phase, the trigonal couplings introduce an anisotropy term proportional to $(-1)^p \cos 3\phi_p$, where ϕ_p is the angle between the x-axis and the basal-plane moments in the p-th layer. This term leads to a bunching, analogous to the effect of the single-ion hexagonal anisotropy, but with trigonal symmetry, and thus to higher harmonics at $(3 \pm 1)\, q_c + \tau_c$. This bunching effect accounts for the recent neutron diffraction results of Lin *et al.* [8] and our similar results [6] in the cone phase, and explains the tendency to form a commensurable structure in this phase, despite the negligible influence of the hexagonal anisotropy.

Our results show that the two-ion interactions with trigonal symmetry may be a substantial fraction of the isotropic exchange interaction in erbium. They probably arise from the spin-orbit coupling of the conduction electrons and may be particularly large in erbium, because the orbital angular momentum of the ions is large.

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