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Magnetic structures and excitations in rare earth metals: old problems and new solutions

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Recent progress in understanding the magnetic structures and excitations in rare earth metals is summarized. The novel, long-range periodic structures which have been identified in Ho and Er, including spin-slips and helifans, are described and some aspects of their excitation spectra considered. It is shown how these new concepts account for a number of phenomena which have remained unexplained for decades. The potential contribution of neutron scattering to the further elucidation of rare earth magnetism is discussed.

1. Introduction

The magnetic structures and excitations of the rare earth metals are generally rather well understood [1]. However, a number of features have remained unexplored or unexplained for decades, and some of them have only recently been accounted for in terms of new insights and principles, and new structures and modes of excitation. It is the purpose of this brief review to take up some of these old problems, to explain how a few of them have been solved during the last few years, and to consider how the judicious application of neutron scattering could elucidate some of the areas which are at present obscure.

Neutron scattering has, of course, made incomparably the greatest experimental contribution to our understanding of rare earth magnetism. Neutron diffraction has allowed the detailed description of most of the magnetic structures, in conjunction with mean-field theory, which works well for the closely packed structures and long-range interactions which are characteristic of the rare earths, and allows an accurate description of even the most complex moment configurations. Furthermore, its time-dependent extension, the random phase approximation, provides a method of calculating the generalized susceptibility, which determines the excitation spectrum and the inelastic neutron scattering cross-section, and hence allows a critical comparison with experiment.

A number of topics of particular current interest in the magnetism of the rare earths are treated in other contributions to these Proceedings. We shall not therefore consider in detail the exciting developments which have occurred in studies of, for example, the multiple-Q magnetic structures of Nd [2], or the spin-wave and crystal-field excitations in the periodic and ferromagnetic structures of Tm [3]. We shall rather restrict our discussion to long-range, generally commensurable, periodic structures and their excitations, describing recent advances in this field and the prospects for further progress. In conclusion, we broaden the perspective somewhat by identifying a number of neutron scattering experiments which appear to be practicable, and which would significantly deepen our general understanding of rare earth magnetism.

2. Long-range periodic structures

By the early 1970s, the magnetic structures of most of the rare earths had been determined under widely varying conditions of temperature and, in some cases, magnetic field, primarily by Koehler [4] and his colleagues. However, a number of details remained unclear. For example, the effect of the competition between the exchange in Ho, which tends to produce an incommensurable helical structure characterized by a Q-vector which varies continuously with temperature, and the very strong hexagonal anisotropy, which pulls the moments towards the b-directions in the plane, was not understood. It was generally believed that Q would indeed vary continuously, but that the structure would be strongly distorted by the anisotropy. High resolution studies with both X-rays and neutrons by Gibbs et al. [5] revealed instead that Q does not increase uniformly with temperature, but rather a series of commensurable wave vectors is traversed, with apparently discontinuous jumps between them. In general, the direct or indirect coupling of the moments to the lattice periodicity, through the anisotropy field, will always favour a commensurable structure. By introducing "phase-slips" into an otherwise regular structure, it is possible to obtain a commensurable structure with a fundamental wave vector arbitrarily close to the non-commensurable value favoured by the exchange. A periodic distribution of the phase-slips may thus lead to a long-period structure which is commensurable, as preferred by the coupling to the lattice, without increasing the exchange energy significantly.

In order to adjust $\boldsymbol{Q}$ in Ho, spin-slips [5] are introduced into the bunched helical structure [4], at which one plane of a bunched doublet is omitted while the remaining member orients its moments along the adjacent easy axis, as illustrated in fig. 1. The hexagonal anisotropy in Ho declines rapidly with temperature, because the thermal expectation value $\langle O_6^6 \rangle$ decreases with the relative magnetization, roughly as σ^{21}, so that the bunching of the moments decreases and it becomes difficult to identify commensurable structures above about $30\,\mathrm{K} \sim 0.2\,T_N$. In the cycloidal structure of Er, also illustrated in fig. 1, the coupling to the lattice is however predominantly due to the axial anisotropy, which declines relatively slowly, roughly like σ^3. A lengthy sequence of commensurable structures is therefore observed [6], extending up to above $50\,\mathrm{K} \sim 0.6\,T_N$. These may also be described as "spin-slip" structures, characterized by the numbers of successive planes in each block of moments with components alternately parallel and antiparallel to the c-axis. The Er structure of fig. 1 has a period of 7 hexagonal layers, 4 with the moments predominantly parallel to the c-axis followed by 3 in the opposite direction, and it may be designated (43). This structure has a small net moment of about $0.6\mu_B$/atom along the c-axis, and it is therefore very effectively stabilized by a magnetic field in the c-direction [7]. The Ho one-spin-slip structure of fig. 1 also has a net moment, this time in the basal plane, but whereas this is the only structure observed in Ho with this property, the (4443) and (444443) structures, which are present in Er at lower temperatures, both possess c-axis moments. The intermediate structures of the type (443443) do not however have a spontaneous moment, nor does the terminal structure (44), which is stable just above the transition to the cone. The presence of a net moment has been observed as peaks in the low field AC susceptibility in the c-direction in Er [8] at the temperatures where the three ferrimagnetic structures are stable: the one-spin-slip structure in Ho has also been detected by magnetization measurements [9].

The concept of blocks of moments may also be usefully applied to the structures which arise when a helix is subjected to a field in its plane [10]. As the field is increased, the helix first distorts, giving rise to a moment along $\boldsymbol{H}$, and then undergoes a first-order transition to a fan structure, in which the moments oscillate about the field direction, as illustrated in fig. 2. A further increase in the field reduces the opening angle of the fan which, in the absence of magnetic anisotropy, goes continuously to zero, establishing a ferromagnetic phase at a second-order transition. Hexagonal anisotropy may modify this process by inducing a first-order transition or, if it is large enough, eliminate the fan phase entirely. Through mean-field calculations of the effect of a magnetic field on commensurable periodic structures, Jensen and Mackintosh [11] identified the intermediate phases which had been observed, by neutron diffraction [12] and other measurements, above about 40 K between the helix and the fan. If the helix is considered as blocks of moments with components alternately parallel and antiparallel to the field, written schematically as $(+-+-)$, and the fan structure is described as $(++++)$, the new structures, the helifans, corre-

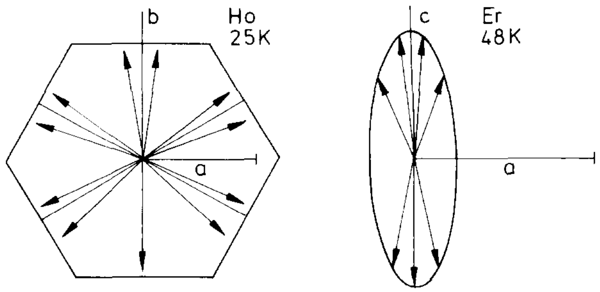

Fig. 1. Self-consistent mean-field calculations of commensurable periodic structures in Ho and Er. Each arrow represents the magnitude and direction of the ordered moment in a specific plane normal to the c-axis, relative to the size of the moment at absolute zero ($10\mu_B$ and $9\mu_B$, respectively), indicated by the length of the horizontal lines. The orientation of moments in adjacent planes is depicted by the positions of neighbouring arrows. The bunched pairs of moments in the 11-layer one-spin-slip structure of Ho are disposed unsymmetrically with respect to the easy axis in the vicinity of the spin slip. The Er structure is restricted to the a–c plane, and can be considered as comprising four planes of moments with a positive component along the c-axis, followed by three with a negative moment, with the designation (43).

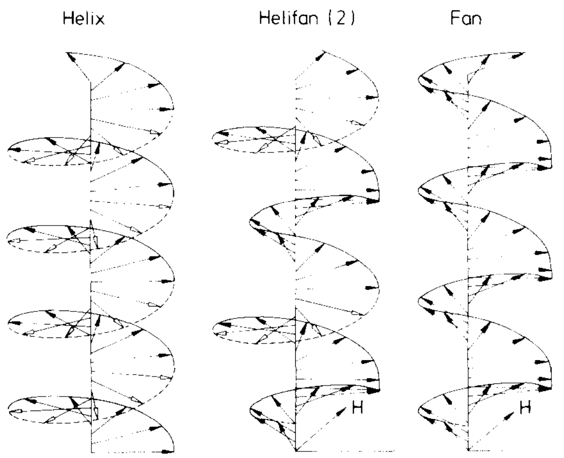

Fig. 2. The helix, helifan(2) and fan structures in Ho at 50 K. The moments lie in planes normal to the c-axis and their relative orientations are indicated by arrows. A magnetic field of 11 kOe is applied in a b-direction in the basal plane to the helifan and fan, and moments with components respectively parallel and antiparallel to this direction are represented by filled and open arrow-heads.

spond to intermediate patterns. For example, the helifan(2), illustrated in fig. 2, may be depicted as $(+++-)$. Both the neutron-diffraction experiments and the calculations show that the helifan(3/2) $(++-++-)$ is more stable than the helifan(2), but there are indications that the latter may be observed if the field is increased rapidly, since it is readily formed from the helix. Other stable and metastable helifans could be observed under suitable conditions. Helifans, or analogous structures, may also occur in other rare earth systems where periodic ordering is observed. An example is provided by the modulated structures with wave vectors in the basal plane observed in Nd. These may be described as $(+-+-+-+-)$, indicating blocks of moments with a component parallel or antiparallel to a magnetic field applied in the plane. A periodic reversal of $(-)$ blocks will then generate subharmonics of the basic $\boldsymbol{Q}$-vector. Thus the sequence $(+++-+-+-)$ generates $\boldsymbol{Q}/4$, and $(+++-+++-)$ gives $\boldsymbol{Q}/2$, both of which have been observed by neutron diffraction in a magnetic field [2].

It might be questioned whether the long-period structures considered above, some of which involve up to 50–100 layers in one commensurable period, have any counterpart in real physical systems. Although some irregular phase-slips may indeed occur, there are experimental indications that the correlation lengths in the rare earth metals are extremely long, due to the very long range of the RKKY-exchange interaction. This long-range correlation is for example reflected in the detailed neutron-diffraction measurements of Cowley and Bates [13] on Ho, where peaks corresponding to harmonics of the order 25 times the fundamental wave-vector are resolved.

3. Excitations of long-period systems

The magnetic excitations of the ferromagnetic heavy rare earths have been studied in great detail [1], but much less information is available on periodic structures. A periodic modulation of the moments generally has drastic effects on the spin-wave spectrum, especially by introducing energy gaps distributed through the Brillouin zone. The only exception is the regular helix (and cone), which is equivalent to a ferromagnet in a coordinate system rotating with the helix. Nevertheless, in Ho above ~50 K, where the distortion of the helix due to the hexagonal anisotropy is negligible, an RPA-calculation [14] predicts that the gaps in the spin-wave spectrum should persist, due to the large absolute value of B_6^6. However, thermal broadening at high temperatures will smear out these gaps, and they have not yet been observed. With the exception of the lowest branch, the gaps in the excitation spectrum are not greatly dependent on whether the period of the magnetic structure is commensurable with the lattice or not, if finite resolution effects are included. In the incommensurable case, the free energy is independent of an overall shift in phase of the magnetic structure relative to the lattice, so that the corresponding susceptibility component is infinite. However, this does not always lead to a well-defined phason mode linearly approaching zero energy, as in the unperturbed helix. The use of Goldstone theorem requires that the generator of the phase shift commutes with the Hamiltonian, which is generally not the case if the system shows any tendency towards commensurability. Instead the phason mode becomes diffusive, with diverging intensity but non-zero width close to the ordering wave vector. This may occur even in the zero temperature limit, as in the case of the pressure-induced longitudinally polarized antiferromagnetic phase of Pr [15]. In the commensurable case, the susceptibility components stay finite, and the excitation spectrum is separated from the ground state by a non-zero energy gap. An example was observed at the origin in the bunched helix by Larsen et al. [16], who thereby explained why the low temperature electrical resistivity of Ho gives evidence for such a gap [17], even though it was believed that it should not occur. The detailed behaviour of the gap also indicates why it is the cone and not the tilted helix which is the stable structure at low temperatures. The dipole interaction is large enough to reduce the coupling of the c-axis moments at the wave vector $\boldsymbol{Q}$ below that at zero wave vector. The long-wavelength modes with $\boldsymbol{q} \perp c$-axis therefore lie at lower energies than the spin wave at $\boldsymbol{Q}$. The former then go soft at the second-order transition to the cone, where the total axial anisotropy vanishes.

The bunching of the moments doubles the period of the helix in the rotating coordinate system, and thereby also gives rise to an energy gap in the centre of the zone. This gap was too small to be observed in the bunched helix or zero-spin-slip structure [16], but an analogous effect was observed in the one-spin-slip structure by Patterson et al. [18]. In this case, the 11-layer structure causes an eleven-fold reduction in the Brillouin zone, but only the first order gap at $5/11 \times 2\pi c$ is calculated to be readily observable. This gap, on the other hand is amplified by about a factor two, as compared to that in the structure without spin slips.

No measurements have been reported on the spin waves in the above-mentioned long-range structures created by a magnetic field, even though the effect of such a field on the dispersion relations was analysed about 30 years ago by Cooper and Elliott [19], and further discussed by Nagamiya [20]. In practice, such experiments are likely to prove rather difficult, and it is worthwhile briefly to consider why. The fan structure may to a good approximation be written:

$$
\begin{aligned}
\langle J_a(\boldsymbol{R}) \rangle / |\langle \boldsymbol{J} \rangle| &= \sigma_1 \cos(\boldsymbol{Q} \cdot \boldsymbol{R} + \varphi) , \\
\langle J_b(\boldsymbol{R}) \rangle / |\langle \boldsymbol{J} \rangle| &= \sigma_0 - \sigma_2 \cos 2(\boldsymbol{Q} \cdot \boldsymbol{R} + \varphi) ,
\end{aligned}
\qquad (1)
$$

where $\langle J_a(\boldsymbol{R})\rangle$ and $\langle J_b(\boldsymbol{R})\rangle$ are respectively the components of the moment perpendicular and parallel to the field, assumed to be along the b-axis, and $|\langle \boldsymbol{J}\rangle|$ is the average magnitude of the ordered moments. The dynamical susceptibility $\bar{\bar{\chi}}(\boldsymbol{q}, \omega)$ may be calculated in the RPA in a coordinate system in which the z-axis is locally along the direction of the oscillating moment, and the x-axis is perpendicular to the plane of the fan. The components in the coordinate system defined by the lattice, with the field in the b-direction, are then

$$\begin{aligned} \chi_{aa}(\boldsymbol{q}, \omega) &= \sigma_0^2\chi_{yy}(\boldsymbol{q}, \omega) + \tfrac{1}{2}\sigma_2^2\{\chi_{yy}(\boldsymbol{q}+2\boldsymbol{Q}, \omega) \\ &\quad + \chi_{yy}(\boldsymbol{q}-2\boldsymbol{Q}, \omega)\}\,, \\ \chi_{bb}(\boldsymbol{q}, \omega) &= \tfrac{1}{2}\sigma_1^2\{\chi_{yy}(\boldsymbol{q}+\boldsymbol{Q}, \omega) + \chi_{yy}(\boldsymbol{q}-\boldsymbol{Q}, \omega)\}\,, \\ \chi_{cc}(\boldsymbol{q}, \omega) &= \chi_{xx}(\boldsymbol{q}, \omega)\,. \end{aligned} \tag{2}$$

For the parameters which we have used earlier for Ho [14], at 50 K and in a field of 13 kOe, σ_0^2, σ_1^2, and σ_2^2 are approximately 0.92, 0.5, and 0.08 respectively, so that the $2\boldsymbol{Q}$-components in $\chi_{aa}(\boldsymbol{q}, \omega)$ are negligible.

The positions of the main peaks in the imaginary parts of $\chi_{aa}(\boldsymbol{q}, \omega)$ and $\chi_{cc}(\boldsymbol{q}, \omega)$, corresponding to the energies of the excitations detected in a neutron scattering experiment, are plotted in fig. 3. Within a resolution of about 0.3 meV, there is only one branch at high values of $\boldsymbol{q}$, and the dispersion relation is very similar to the one calculated for the helical phase at the same temperature [14]. The gaps due to the hexagonal anisotropy are at the most a few tenths of an millielectron volt, and are neglected in the figure. At low $\boldsymbol{q}$, on the other hand, there are two branches, deriving from the mixing of the $\boldsymbol{q}$ and the $\boldsymbol{q} \pm \boldsymbol{Q}$ spin waves. At the origin, the upper mode has a very low intensity, which vanishes in the low temperature limit, whereas the lower evolves smoothly into the uniform mode of the ferromagnetic structure as the fan angle approaches zero at the critical field. At the wave vector $\boldsymbol{Q}$, the symmetric cosine or phason mode corresponds to an oscillation of the origin of the fan along the c-axis [20], which requires no energy in the absence of hexagonal anisotropy, or if the ordering is incommensurable. At the high temperature of the calculation, the energy due to a possible commensurable ordering is very small, but in any case the mode becomes diffusive near $\boldsymbol{Q}$. The higher energy, antisymmetric sine or amplitude mode at $\boldsymbol{Q}$ corresponds to an oscillation of the fan angle. The energy goes to zero with the fan angle, so that both modes at $\boldsymbol{Q}$ have zero energy at the critical field, but thereafter rise rapidly with field. The eccentricity of the spin wave modes, as determined by the effective anisotropy, becomes increasingly pronounced with decreasing energy, so that the fluctuations in the limit of zero energy at $\boldsymbol{Q}$ are completely confined to the plane. This means that the low energy branch dominates $\chi_{aa}(\boldsymbol{q}, \omega)$ at low $\boldsymbol{q}$, whereas the upper branch is most pronounced in $\chi_{cc}(\boldsymbol{q}, \omega)$, except very close to $\boldsymbol{q} = \boldsymbol{0}$. From eq. (2), and the form of the cross-section, it is apparent that neutron scattering experiments should be performed with a large component of κ along the b-direction, in order to reduce the disturbing influence of $\chi_{bb}(\boldsymbol{q}, \omega)$, whose peaks are displaced by $\pm\boldsymbol{Q}$ relative to the other components. This condition is however very difficult to reconcile with the requirement that the field is also along b, while $\boldsymbol{q}$ is along c.

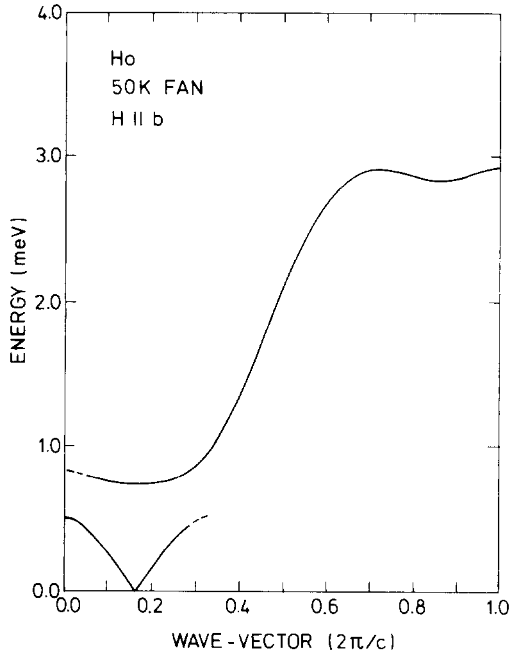

Fig. 3. Calculated dispersion relations for excitations propagating in the c-direction in the fan phase of Ho at 50 K, in a magnetic field of 13 kOe in the b-direction. The positions of the main peaks in the imaginary parts of $\chi_{aa}(\boldsymbol{q}, \omega)$ and $\chi_{cc}(\boldsymbol{q}, \omega)$ are plotted. There is only one branch at high values of $\boldsymbol{q}$, but at low $\boldsymbol{q}$ there are two, deriving from the mixing of the $\boldsymbol{q}$ and the $\boldsymbol{q} \pm \boldsymbol{Q}$ spin waves. At low $\boldsymbol{q}$, the low energy branch dominates $\chi_{aa}(\boldsymbol{q}, \omega)$, whereas the upper branch is most pronounced in $\chi_{cc}(\boldsymbol{q}, \omega)$, except very close to $\boldsymbol{q} = \boldsymbol{0}$.

The excitation spectra of the helifans will mirror the greater complexity of the structures, manifested in the increased number of neutron diffraction reflections [9]. However, we might expect that the behaviour at large $\boldsymbol{q}$ would still resemble that in the helical phase quite closely, and there is also a phason mode of zero energy in the absence of hexagonal anisotropy, which will be reflected in a low energy, partly diffusive branch near $\boldsymbol{Q}$ in the helifan phases of Ho.

4. Conclusion

The developments of the last few years have demonstrated that it is still profitable to carry out even more precise neutron diffraction studies of the magnetic structures of the rare earths. Ho and Er have proved to be particularly fertile subjects and much further work remains to be done, especially on alloys and in a magnetic field, where a variety of helifan and analogous structures may await discovery. Since the magnetoelastic effects in the rare earths are so pronounced, external pressure or uniaxial stress can also have a profound effect on the stability of different magnetic states, as the few examples which have been examined have demonstrated.

Even though the structures of the heavy rare earths are rather well understood, a number of the lighter metals remain largely unexplored. Our understanding of γ-Ce is still at a rudimentary stage, nor is the magnetic structure of Sm by any means completely resolved. The form factor is particularly interesting and unusual, and our understanding of its variation with κ is still incomplete. A dhcp phase can also be stabilized in Sm; a comparison of its magnetic properties with those of the more common allotrope would further elucidate the relation between the crystal structure and the magnetic interactions. The magnetic structures of films and superlattices constitute a field which has only existed for a few years, and is in the process of rapid expansion. There appear to be unlimited possibilities for fabricating new systems, and for discovering new forms of ordering.

The study of the excitations by inelastic neutron scattering has provided diverse and detailed information on the magnetic interactions. The greatest efforts have been devoted to the spin waves in the heavy rare earths, particularly Tb. The dispersion relations of Gd, which has negligible anisotropy, have also been carefully measured over a range of temperatures, but the lifetimes have not been studied as a function of wave vector at low temperatures, which would allow a determination of the scattering by the conduction electrons. The isotropy of the exchange could be examined, to within the limitations of the experimental resolution, by applying a magnetic field, and a study of dipolar effects at long wavelengths might also be possible.

When the moments vary with position in a periodic structure, the excitations become more difficult to study. Eu corresponds to Gd in the role of an isotropic model system, but with a simple helical rather than a ferromagnetic structure. It is unfortunate that its intractable neutron properties (even the more favourable isotope absorbs inconveniently strongly) have so far precluded any measurements of the spin waves. It would be particularly interesting to investigate the mode of wave vector $\boldsymbol{Q}$, whose energy is determined by the small anisotropy, and its dependence on magnetic field. It is energetically favourable for the plane of the helix to rotate so that it is normal to the field direction, and this would be expected to occur via a soft mode transition analogous to that observed in Ho at low temperature, with a similarly decisive influence of the dipolar interactions.

A number of reasonably complete studies of the spin waves in the c-direction have been made in periodic structures in the heavy rare earths, notably in the helical phases of Tb, Dy, and Ho, and the cone structure of Er. A fairly good understanding has also been attained of the excitations in the commensurable spin structures of Tm and Ho. The effect of varying the temperature has only been cursorily explored, however, and lifetimes and field effects have not yet been investigated. No measurements have been made of the spin waves in the (44) or other cycloidal structures of Er, for example, nor have experiments yet been carried out on the spin waves in helifan or fan structures.

Such is the richness of the excitation spectrum of Pr that a number of experiments of fundamental importance remain to be performed, despite the considerable efforts which have already been devoted to this unique element. The magnetic excitons on the hexagonal sites and their relationship to the process of magnetic ordering are well understood, and the anisotropic exchange, magnetoelastic, and crystal field interactions have been measured with unprecedented accuracy, but the excitations on the cubic sites are much less precisely described. The magnetically ordered state provides a new set of challenges. The mechanism of ordering by different perturbations requires further investigation. The neutron diffraction studies of the hyperfine coupling-induced collective state should be taken to lower temperatures, and the electronic and nuclear components on the different sites disentangled. The excitations of this state would also naturally be of interest. The energy of the lowest excited crystal-field state on the hexagonal sites of Pr can be reduced by a magnetic field in the c-direction, which should affect the quasielastic central peak and precursor satellite. The precise nature of these unusual scattering phenomena is still a mystery, which further measurements under different constraints, especially of external pressure, could help to unravel. The application of a uniaxial pressure in the a-direction creates in effect a new magnetically ordered element, and one with very interesting properties [15]. Many informative results have already been obtained from this phase, but the full characterization of the excitations as a function of the strain, temperature, and field is clearly a major enterprise. Of immediate interest would be a renewed effort to observe the amplitude mode, which should be visible at low temperatures.

As may be deduced from this brief survey, which is by no means exhaustive, there are still many problems in rare earth magnetism which neutrons are ideally suited to solve. It is extremely fortunate that three of the elements which display quite different aspects of the multifarious magnetic properties of the rare earths, Tb, Pr, an Ho, have only a single stable isotope with moderate neutron absorption. Experience has taught us that each rare earth metal is a magnetic individual, and an equally careful study of the remaining members of the series would therefore be expected to reveal further examples of unique magnetic behaviour. Such studies require, in many cases, samples composed of single isotopes chosen for their favourable neutron properties. A good start has been made in preparing such samples, but further progress in understanding the magnetism of the rare earths depends on making separated isotopes more readily available to the neutron scattering community.

Acknowledgement

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