# PERSPECTIVES FOR RARE EARTH RESEARCH

During the last three decades, a remarkable transformation has occurred in our understanding of rare earth magnetism. This development has been described in the preceding chapters, and we conclude with a short epilogue, in which we attempt briefly to summarize the status of the field and the perspectives for future research in it. The close interplay between experiment and theory is particularly pronounced in rare earth research. It was measurements on pure materials and single crystals which stimulated the early development of the subject, but the construction of the standard model pointed the way to more refined and varied experiments, which in turn required more sophisticated explanations. At the moment, we appear to be in a period where the theory is in the ascendancy; it is able to account for the great majority of the observations, and also to suggest a wide variety of new investigations. Much of the following will therefore be concerned with the indication of promising directions for experimental study.

However, it is clear that the standard model is indeed a model, and transforming it into a fundamental theory will require a deeper quantitative understanding of the magnetic interactions. The key to such an understanding lies in the electronic structure. Band structure calculations are able to predict the ground-state properties of solids with impressive accuracy, and to the extent that comparisons with experimental results exist for the rare earth metals, they are highly successful. Nevertheless, this comparison between theory and experiment is incomplete for even the two most carefully studied examples, Gd and Pr. In both cases, the Fermi surface has been measured in considerable detail with the dHvA effect, but even though the general agreement with the calculations is satisfactory for the larger sheets, the small areas are still not fully accounted for. These discrepancies point to the necessity both of more accurate measurements and of first-principles self-consistent calculations, in which the spin-orbit coupling, the exchange splitting, and the external field are rigorously incorporated. A more complete description of the conduction electrons in Gd would allow further progress in the computation of the sf exchange, although it would not immediately solve the most intractable part of the problem, the screening of the exchange field of the 4f electrons, which reduces its effect by a large factor. The contribution of the spin-orbit coupling in the conduction-electron gas to the single-ion anisotropy could also be estimated and compared with the low-temperature experimental results, taking into account the readily computed dipolar contribution. A knowledge of both the experimental and theoretical masses on a variety of orbits would allow a stringent test of the theory of mass-enhancement by the spin waves. The rich structure in the low-frequency dHvA spectrum of Pr may reflect the hybridization with the 4f electrons, which presumably gives rise to the quasielastic neutron-diffraction central peak, and also apparently makes a contribution to the binding in the light rare earths. An immediate goal of a first-principles calculation of the indirect exchange in Pr would be an explanation of the large anisotropy which is revealed by the dispersion relations for the magnetic excitations. This would require an extension of the theory of the influence of the orbital angular momentum on the two-ion coupling to more realistic electronic structures than the free-electron model which is normally considered. If this could be accomplished, a first-principles account of the observed scaling of the exchange with the de Gennes factor in the elements could be envisaged.

An accurate knowledge of the electronic structure would also open the way to a calculation of the source of the other primary interaction, the crystal field, which is determined by the charge distribution. The non-spherical terms, which give rise to the higher-l components of the crystal field, are neglected in the averaging procedure adopted in the construction of the muffin-tin potential, but both they and the nonuniformity of the charge distribution in the interstitial regions can in principle be calculated self-consistently. Such a calculation for Y, for example, would cast some light on the origin of the crystal-field splittings observed in dilute alloys with magnetic rare earths. However, the main barrier to calculating the full crystal-field Hamiltonian again resides in the other part of the problem, the screening of the fields by the atomic core and the 4f electrons themselves. This is indeed a formidable difficulty, but presumably not one which is insurmountable.

A constant theme, running in parallel with the steady improvement of the standard model, has been the question of the nature of the 4fstates in Ce and its compounds. As has been apparent for some time, the standard model is not applicable to, for example,  $\alpha$ -Ce, as the 4felectrons are itinerant, make a substantial contribution to the binding, and must be described by the band model. We have said relatively little about mixed-valent Ce compounds, primarily because they lie outside the main scope of this book, but also because the subject is in a very rapid state of development, making any kind of meaningful summary both difficult and ephemeral. Nevertheless, there is no question that the study of the magnetic properties of these materials, and of the

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related actinide compounds, and of the possible connection between magnetism and superconductivity in heavy-fermion systems, is one of the most fruitful areas of research in solid state physics, and one in which great progress can be anticipated in the next few years.

We have emphasized the efficacy of neutron scattering for the experimental study of rare earth magnetism, but macroscopic measurements provide an invaluable complement. The macroscopic magnetic properties are frequently very sensitive to impurities, and many of the parameters on which we rely for comparison with theory were measured on crystals substantially less pure than can be prepared today. The measured value of the conduction-electron polarization in Gd, for instance, increased significantly with crystal purity. The magnetic moment of the conduction-electron gas is an important quantity, which gives valuable information on the exchange interaction with the 4f electrons, but since it is typically an order of magnitude smaller than the ionic moment, its accurate determination is not straightforward, either in the paramagnetic or the ordered phases, and the values of Table 1.6 could in many cases be improved upon. The anisotropy parameters, describing the angular dependence of the free energy in a field, are also difficult to determine precisely, paradoxically because they are frequently so large in the rare earths. It may consequently be very difficult to pull the moments out of the easy direction, and the torque on a crystal in a field can be huge. Apart from the question of sample purity, many of the values in the literature therefore suffer from substantial uncertainties for technical reasons. The same may be said for the multifarious magnetoelastic parameters, which characterize the dependence of the free energy of the magnetic systems on the lattice strain. The temperature dependence of this free energy is reflected in the heat capacity, which therefore in principle contains useful information on the energetics of magnetic materials. However, the unscrambling of the nuclear, lattice, magnetic and electronic components may be a formidable task, and small amounts of impurity may make the results essentially useless. Nevertheless, it would be worthwhile to attempt to improve upon the accuracy of the available measurements, and the effect of an external magnetic field on the heat capacity could be pursued further, since the few studies which have so far been made have been very informative.

It was the revelation by neutron diffraction of the exotic magnetic structures of the rare earths which initiated the revolutionary progress of the 1960s, and since that time countless studies have been performed of the patterns of the ordered moments. It is therefore remarkable that so much remains to be done. The temperature dependences of the structures of the heavy rare earths have been determined in great detail, although there is still scope for further study of some phases, for instance in the intermediate temperature-range of Er. However, the effect of other external constraints, especially magnetic fields and pressure, have only been cursorily investigated. It is apparent, for instance, that a variety of helifan and analogous structures may be produced by applying a field to periodic structures, especially if the interactions are adjusted by taking advantage of the almost perfect mutual solubility of the elements. Apart from their intrinsic interest, measurements of the resistivity may provide a very sensitive method for determining the complex phase diagrams which frequently arise in a magnetic field. The same may be said of the lattice strain, and of the elastic constants (Bates et al. 1988). Since the magnetoelastic effects in the rare earths are so pronounced, external pressure or uniaxial stress can have a profound effect on the stability of different magnetic states, as the few examples which have been examined have demonstrated. The light rare earths remain a largely unexplored terrain. The more that is learned about the magnetic structures of Nd, the wider loom the areas which remain to be investigated. Our understanding of  $\gamma$ -Ce is still at a rudimentary stage, nor is the magnetic structure of Sm by any means completely resolved. Although its crystal structure is complicated, and neutron experiments require isotopically enriched samples to circumvent the large absorption in the natural state, there is no fundamental obstacle to attaining a more detailed description of the configurations of the moments under different conditions than we have at present. The form factor is particularly interesting and unusual, and the theoretical understanding of its variation with  $\kappa$  is still incompete. 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 mean-field theory, in conjunction with the standard model, has proved to be ideally suited for explaining the general features of the magnetic structures in terms of the interactions. Furthermore, detailed self-consistent calculations provide an accurate description of the arrangement of the moments determined by neutron diffraction and macroscopic measurements, under specified conditions of temperature and field, and also give a good account of the variation of the macroscopic anisotropy and magnetostriction parameters with the magnetization. Although the crystal-field parameters are adjusted to fit, for example, the low-temperature magnetic properties, they are not normally thereafter varied in the calculation. The indirect-exchange interaction

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must be taken as explicitly temperature dependent, but its variation is constrained to be consistent with the excitation spectrum. Only in the vicinity of the critical temperature does the mean-field theory fail seriously in the rare earths. Apart from this, the most severe challenge which has so far been presented to the theory is the explanation of the manifold structures of Nd in terms of a set of fundamental interactions. This challenge has not yet been fully met, but this is probably more due to an incomplete knowledge of the interactions than to any fundamental limitation of the method.

The random-phase approximation, which is a time-dependent extension of the mean-field method, provides a similarly powerful theoretical technique for treating the excitations. It is also quite general, but the results usually have to be obtained by numerical means. The linear spin-wave theory is an attractive alternative when the low-temperature moment is close to its saturation value, since it may be treated analytically, may readily be made self-consistent, and allows the identification of the combinations of parameters which determine the essential features of the excitation spectrum. The corrections to the theory may be computed systematically as an expansion in 1/J, and considerable progress has been made in the calculation of such higher-order terms. Experimental investigations of the finite lifetime effects which appear in the third order of 1/J, due to the magnon-magnon interactions, have so far been limited. The theory of this effect is well established for the Heisenberg ferromagnet, but it remains to be combined with the 1/J-expansion in the anisotropic case.

The RPA theory has the merit of providing the leading-order results for the excitation spectrum in the systematic 1/Z-expansion. This expansion is particularly well-suited for the rare earth metals, as it takes advantage of the large value of the effective coordination number Z, due to the close packing and the long range of the indirect exchange. Except in the immediate vicinity of a second-order phase transition, where any perturbation theory will fail, it seems capable of giving a rather satisfactory account of the many-body correlations even in the first order of 1/Z, if there is a gap in the excitation spectrum, as demonstrated by the example of Pr. The usefulness of the theory in this order is much improved by the fully self-consistent formulation presented in Section 7.2. However, a substantial effort is still required to calculate the 1/Z-terms in systems with more complicated single-ion level schemes than those considered there. Furthermore, in spin-wave systems, or system with gapless excitations,  $(1/Z)^2$ -corrections are important for the linewidths. We can however conclude that the 1/Z-expansion indicates quite generally that the  $(1/Z)^0$ -theory, i.e. the RPA, should be a good first-approximation in the low-temperature limit.

The study of the excitations by inelastic neutron scattering has provided diverse and detailed information on the magnetic interactions. A glance at the dispersion relations of, for instance, Gd, Tb, and Pr immediately reveals the relative importance of the exchange and crystal fields in these elements. Careful measurements as a function of temperature and external fields, combined with a systematic analysis, yield an abundance of knowledge about these and other interactions. Further information is contained in the neutron-scattering intensities, though these have so far been relatively little utilized. However, in some cases, relative intensities have allowed a discrimination between different models, and if the difficult experimental problems can be overcome, absolute intensity measurements could provide a valuable supplement to the energies and lifetimes. The philosophy adopted in analysing measurements of excitation spectra has generally been to use the simplest theory and set of interaction-parameters which can provide a satisfactory fit to the experimental results available at any particular time. The anisotropic exchange may, for example, be subsumed in effective isotropic exchange and crystal-field parameters, so that it is necessary to treat such quantities with caution when comparing with values deduced from different kinds of measurements, or from a fundamental theory.

The excitations to which the greatest efforts have been devoted are the spin waves in the heavy rare earths. Gd, with negligible anisotropy, is the simplest example and the dispersion relations have been carefully measured over a range of temperatures, in an isotopically pure sample. The lifetimes have also been studied, but not as a function of wave-vector at low temperatures, which would allow an examination of the scattering by the conduction electrons, without the interference by the magnonphonon interaction which partially disturbed the experiments on Tb. Gd would also be the prime candidate for a detailed comparison with a realistic theory, using the calculated band structure and measured Fermi surface rather than the free electron model. 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. Despite the attention which has been devoted to elucidating the excitation spectrum of Tb, a number of questions remain. The relative importance of single- and two-ion contributions to the macroscopic anisotropy and spin-wave energies could be further clarified by more precise measurements in a field, taking full account of the influence of the dipolar coupling at long wavelengths. The origin of the hard-axis axial anisotropy, and the discrepancy between the macroscopic and microscopic hexagonal anisotropy, could thereby be further investigated. The study of the magnon-phonon interaction, which has earlier provided some fruitful surprises, could profitably be

extended to other branches, and the magnon energies should be measured in a single-domain crystal, in order to establish whether the modification of the hexagonal symmetry by the magnetic ordering is reflected in the dispersion relations. It would be interesting to extend the measurements of Nicklow *et al.* (1971b) on ferromagnetic Dy, and subject it to an equally rigorous investigation, since the anisotropy parameters are rather different from those in Tb, but its large hexagonal anisotropy restricts the range of measurements in a field. Studies of ferromagnetic Ho are even more constrained.

When the moments vary with position in a periodic structure, the excitations become less amenable to study and the available information on them is relatively sparse. 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 **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. Apart from Pr, to which we return shortly, the only observations of inelastic neutron scattering in the light rare earths are preliminary studies of crystal-field excitations in the longitudinal-wave structure of Nd by McEwen and Stirling (1982). It may be an advantage to apply to these complex systems a magnetic field large enough to induce ferromagnetic ordering, in order to decouple the complications of periodic ordering from the problems of the interplay of crystal-field and exchange interactions in the excitation spectrum.

A number of reasonably complete studies of the spin waves in the c-direction have been made in incommensurable periodic phases of the heavy rare earths, notably in the helical structure of  $Tb_{90}Ho_{10}$  and the conical phase of Er. The former is a good example of how the mutual solubility of the rare earths can be utilized in modifying the magnetic properties in a convenient manner, in this case by extending the temperature range over which the helix is stable. The effect of varying the temperature has only been cursorily explored, however, and lifetimes and field effects have not yet been investigated. Experimental spectra for the longitudinal-wave structure of Er bear little resemblance to the predictions of the theory, and both will presumably have to be improved before they can be expected to converge. A fairly good understanding has been attained of the excitations in the commensurable spin struc-

tures of Tm and Ho, and the first effects of spin slips in introducing new energy gaps within the zone have been observed in the latter. However, no experiments have yet been carried out on the spin waves in the helifan and fan structures which may be induced by a magnetic field, nor has the soft mode which should accompany a second-order transition from the latter to the ferromagnet been observed.

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. The  $|3_{\rm S}>$ level has not yet been detected directly, however, and a further study of the lifetimes of the long-wavelength acoustic modes as a function of magnetic field would give detailed information about their interaction with the conduction electrons. The excitations on the cubic sites are much less precisely described, and even though they are of lesser significance, a full characterization of their energies and the underlying interactions is essential for the explanation of the temperature dependence of the magnetic properties. In particular, a measurement of the missing branch of excitations polarized in the *c*-direction, its possible splitting from the branches polarized in the plane, and of the field dependence of the energies, would help to determine the remaining magnetoelastic and crystal-field parameters, especially  $B_2^0(c)$  which is usually assumed to be zero, but is likely to be as large as the corresponding parameter on the hexagonal sites.

The magnetically ordered state in Pr provides a new set of challenges. The mechanism of ordering by different perturbations requires further investigation. The neutron-diffraction studies of the hyperfinecoupling-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 influence of magnetic impurities, such as Nd and Er, could be clarified by further inelastic-scattering experiments in a field. The energy of the  $|+1\rangle$  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. Many informative results have already been obtained from

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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.

Despite the impressive range of information which has been attained by accurate measurements of the magnetic structures and excitations under various external conditions, the investigation of the two-ion interactions which supplement the dominant isotropic RKKY exchange is by no means complete. Low-symmetry two-ion couplings are clearly important in Pr and Nd. In addition to the anisotropic contribution  $\mathcal{K}(ij)$  in (2.1.41), which accounts for the stability of the longitudinal ordering of the moments and the splitting of the doublet-excitations in Pr, a further term must be included to explain the *c*-axis moments on the cubic sites in Nd, which are induced by the ordered basal-plane moments on the hexagonal sites. In the heavy rare earths Tb and Er, there is indirect but weighty evidence that anisotropic interactions are essential for explaining the excitation spectra. The strong optical-phonon – acoustic-magnon interaction in the *c*-direction of Tb is of fundamental significance, since it reveals that the spin-orbit coupling of the conduction electrons results in a spatially varying deviation between the direction in which their spins are polarized and that of the ferromagnetic ionic moments. The spin-orbit coupling may also be important for explaining the possible occurrence of interactions, such as the example given in (2.1.39), which reflect the reduced three-fold symmetry of the *c*-axis in the hcp lattice. Indications that this kind of coupling is present in Ho have appeared in the possible detection by Cowley and Bates (1988) of a modulated *c*-axis moment in the commensurable helical structures, and the unusual stability of the  $Q = \pi/2c$ -structure around 96 K (Noakes *et al.* 1990). The c-axis moments should alternate in sign between each pair of planes in the spin-slip structures of Fig. 2.5, being zero on the spin-slip planes, so that  $Q_c = 2\pi/c - 3Q = \pi/c$  in the twelve-layered zero-spin-slip structure.

The aforementioned mutual solubility of the rare earth elements gives unlimited possibilities for fabricating systems with adjustable magnetic properties. Dilute alloys of magnetic ions in non-magnetic hosts such as Y have proved particularly interesting, largely because the crystal fields may be studied in the absence of the exchange interaction and its possible anisotropy. The crystal-field levels have primarily been determined by measurements of the magnetic moments, but further neutronscattering studies, which are possible with modern experimental techniques even in very dilute systems, would be even more enlightening, especially in a magnetic field. These systems can also be effectively investigated by the straightforward means of measuring the electrical resistivity as a function of temperature and field, and comparing with the pure solvent element. As the concentration is increased, magnetic ordering is observed at correspondingly higher temperatures. Because of the long range of the indirect exchange interaction, all systems of rare earth Kramers-ions in Y which have so far been studied order in the *c*direction at sufficiently low temperatures, although there may be some disorder in the plane (Caudron *et al.* 1990). It would be interesting to examine by neutron diffraction even more dilute systems, to clarify the mechanism by which the moments are aligned. As the concentration of heavy rare earths in Y is further increased, the alloys change rather rapidly from crystal-field to exchange-dominated systems. This process, and especially the transition from crystal-field to spin-wave excitations (Wakabayashi and Nicklow 1974), deserves detailed examination.

The potentiality for adjusting the interactions to fabricate concentrated alloys with novel magnetic properties is restricted only by the imagination. The Ho-Er system is a good example, in which the richness of the phase diagrams of the two constituents, and the competing crystal-field anisotropies, give rise to a great variety of structures, especially in a magnetic field. The excitations of such binary alloys have only been investigated to a limited extent. The most systematic studies have so far been those on Tb alloys, which are well described by the average crystal or virtual crystal approximations. It would be interesting to examine a system in which lifetime effects are sufficiently pronounced to allow a comparison with the predictions of the coherent potential approximation; Pr alloys may be good candidates in this context. It would also be informative to investigate the behaviour of light rare earths dissolved in the heavy elements, and vice-versa. Preliminary studies have been made of Er in Pr, and Pr in Tb would be a natural choice for a complementary system. We have only peripherally discussed compounds of rare earths with other elements, but they may possess novel and interesting properties. Of particular significance are compounds with magnetic transition metals, such as  $ErFe_2$  (Clausen *et al.* 1982) and HoCo<sub>2</sub> (Castets et al. 1982), which display a striking interplay between itinerant and localized magnetism. As mentioned earlier, Ce compounds constitute a field of magnetism in themselves. Their properties vary from highly localized magnetism, often with very anisotropic interactions, through mixed-valent systems with heavily quenched moments, to non-magnetic heavy-fermion superconductors. They are thus ideally suited for investigations of the limits and breakdown of the standard model.

It is a vain ambition for any authors to aspire to write the last word on any subject; there is always more to say. We have rather attempted to summarize the present state of knowledge and understanding of rare earth magnetism, and indicate some directions for future research. It is

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sometimes argued that, since the standard model with suitably chosen interaction parameters accounts so well for the overwhelming majority of the magnetic properties, the field is essentially closed and no problems of fundamental interest remain, apart from the question of determining the parameters from first principles, and of possible failures of the model. This statement is however no more or less true or relevant than the assertion that the Schrödinger equation, or perhaps the Dirac equation, solves all of the basic problems of chemistry. The interesting and fundamental physics is to be found in the manner in which the interactions express themselves under different circumstances. The same model applied to Pr and Tb gives very different, if equally striking, behaviour. A sufficiently careful examination of any rare earth system will inevitably produce surprises, puzzles, and ultimately deeper understanding. In the last few years, this process has been clearly exemplified by the structures and excitations of Ho, which were formerly believed to be essentially understood, and not particularly interesting. We are therefore confident that, when the time comes to present another review of this subject, it will contain physical principles and phenomena which are scarcely hinted at in this book.

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