Magnetic structures and excitations in rare-earth metals

Jens Jensen
Ørsted Laboratory, Niels Bohr Institute, Universitetsparken 5, 2100 Copenhagen, Denmark
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The recent development within the studies of magnetic structures and excitations in the rare-earth metals is reviewed. The fine resolution obtainable in x-ray synchrotron experiments led to the discovery of a number of commensurable magnetic structures in Ho and Er which helped renew interest in these long-period helically or cycloidally modulated structures. Shortly after, the helifan phases in Ho were established and effects due to trigonal couplings were detected in Er and Ho. The development of the technique of molecular-beam epitaxy has allowed the fabrication of superlattices and alloys of rare-earth metals. One of the important discoveries made possible by the use of this technique is the isolation of a pentacritical point in the magnetic phase diagram of the Ho-Er and Ho-Tm alloys. The investigation of the magnetic structures in the two light rare-earth metals Nd and Pr gives rise to a number of challenging problems, because of the complexity of the different structures and, in Pr, because of the low value of the Néel temperature (50 mK). The extra excitation branch observed a couple of years ago, in the paramagnetic phase of the singlet-ground-state system Pr at small wave vectors, still awaits a full explanation.

I. INTRODUCTION

The two dominant magnetic couplings in the rare-earth metals are the single-ion anisotropy terms due to the crystalline electric field acting on the magnetic $4f$-electrons, and the Ruderman-Kittel-Kasuya-Yoshida RKKY-exchange interaction by which the localized $4f$-moments on different ions are coupled indirectly through the polarization of the conduction electrons. To a first approximation, the indirect exchange leads effectively to a Heisenberg Hamiltonian for the coupling between the $4f$-moments, when assuming the conduction electrons to behave as free electrons. The conduction electrons at the Fermi surfaces of the rare-earth metals are predominantly d-electron-like, and orbital modifications of the exchange coupling introduce anisotropic two-ion interactions in the magnetic Hamiltonian. It is difficult to separate two-ion anisotropy effects from those due to the single-ion crystal-field terms. However, the indications are that the coupling between the different moments in the rare-earth metals involves a great variety of anisotropic two-ion interactions in addition to the isotropic Heisenberg interaction [1,2].

The heavy rare-earth metals are all hexagonal close-packed, and the magnetically ordered phases of the $4f$-moments found in these metals are either ferromagnetic or the moments are antiferromagnetically modulated along the c axis. In the latter structures, the individual hexagonal layers are uniformly magnetized in a direction which changes from one layer to the next. The basic arrangements of the moments in these systems were determined by neutron-diffraction experiments by Koehler and his colleagues during the 1960s [3].

The first inelastic neutron-scattering experiment was carried out on ferromagnetic terbium in 1966 [4]. During the next two decades, similar measurements of the spin-wave energies in the other heavy rare-earth metals were performed [5,1], and one main result is shown in Fig. 1. The exchange interaction, $\mathcal{J}(\mathbf{q})$, is proportional to the magnetic susceptibility of the conduction electrons, and should according to the simple RKKY-theory scale as $(g-1)^2$. The metallic properties of the heavy rare-earth metals are very similar, nevertheless, the results shown in Fig. 1 indicate a pronounced variation of the exchange coupling in the c direction with atomic number. The magnitude of the peak, which stabilizes the observed periodic magnetic structures, increases monotonically through the series. Here we shall be concerned

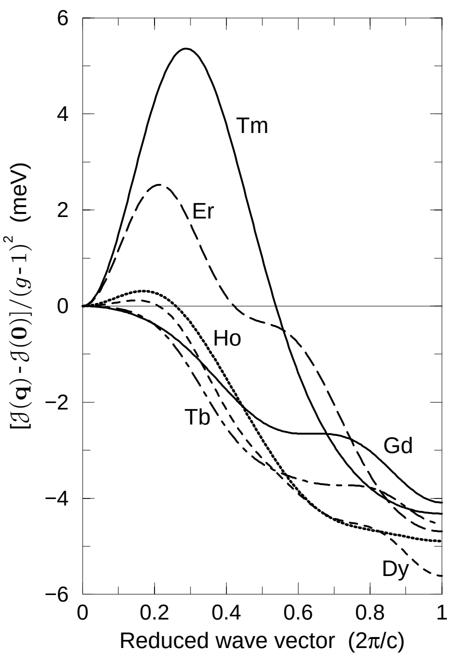

Fig. 1. The exchange coupling $[\mathcal{J}(\mathbf{q})-\mathcal{J}(\mathbf{0})]/(g-1)^2$, with $\mathbf{q}$ parallel to the c axis, determined experimentally in the magnetic heavy rare-earth metals (after Ref. [6]).

mostly with the three heavy rare earths, Ho, Er, and Tm, in which a rich variety of different antiferromagnetically ordered phases have been detected.

The two "normal" light rare earths, Pr and Nd, are double hexagonal close-packed (dhcp) with an *ABAC* stacking sequence of the hexagonal layers, which has two non-equivalent sites. The nearest surroundings of the *A* sites is close to be fcc like, whereas the local surroundings of the *B* and *C* sites has hcp symmetry. The difference in lattice structure implies that the RKKY coupling in Pr and Nd does not have its maximum in the *c* direction, but at a wave vector lying in the basal plane at about 0.13 $\mathbf{a}^*$. The $(g-1)^2$-scaling of the RKKY coupling means that it is weak in Pr and Nd in comparison to the crystal-field splittings. Due to the Kramers degeneracy of the ground state the moments in Nd order at about 20 K, whereas Pr is a singlet-ground-state system in which the RKKY coupling is smaller than the threshold value required for inducing a magnetic ordering. This system eventually orders at about 50 mK because of the hyperfine interaction to the nuclear spins.

II. LONG-PERIOD MAGNETIC STRUCTURES IN THE HEAVY RARE-EARTH ELEMENTS

Tb, Dy, Ho are easy-planar magnets in which the moments order in a helical structure at T_N. At lower temperatures, Tb and Dy become basal-plane ferromagnets, whereas the ordered moments in Ho change into a cone-structure at low temperatures, with a small ferromagnetic component along the *c*-axis superimposed on the helical arrangement in the basal plane. In Er and Tm the *c*-axis is the easy axis, which implies that they order in a longitudinally polarized structure where, just below T_N, the *c*-component of the moments is sinusoidally modulated. In Tm there is a lock-in transition to a seven-layered structure and, in the low temperature limit, the *c*-component saturates, being positive for four layers followed by three layers in which it is negative. The tendency towards a minimization of the variation of the magnitudes of the moments, which becomes more pronounced the lower the temperature, results in the case of Er in a phase in which one of the basal-plane components is nonzero, leading to an elliptic cycloidal structure. At the lowest temperatures, the cycloid is replaced by a *c*-axis cone structure.

The observation of long-period structures commensurable with the lattice, which was made in the high-resolution synchrotron x-ray experiments on Ho [7] and Er [8] by Gibbs and coworkers, has renewed interest in the magnetic ordering in these systems. In the low-temperature limit of Ho, the ordering wave vector locks in to $\frac{1}{6}\mathbf{c}^*$ [9], corresponding to a period of twelve hexagonal layers. The hexagonal anisotropy in Ho causes the helical component of the moments to bunch around the easy *b* axes. In the twelve-layered structure, the moments of the different layers bunch pairwise about successive easy directions making angles of about $\pm 5.8°$ with the nearest *b* axis. As the temperature is increased, the ordering wave vector becomes longer and commensurable structures with different periods are derived from the twelve-layered structure by replacing one or more of the pairs in a period with a single layer where the basal-plane moment is along the easy axis, forming the so-called "spin-slip" structures [7]. The formation of the spin-slip structures was confirmed by high-resolution neutron diffraction experiments performed on large crystals on a triple-axis spectrometer, allowing the observation of peaks of up to 4 orders of magnitude smaller than the main ones [10].

In the presence of an external field new magnetic structures appear in Ho as detected in resistivity measurements [11–13], in x-ray experiments [14], and in neutron diffraction experiments [9,15,16]. As the field applied perpendicular to the *c* axis of the helical structures is increased, the helix first distorts, giving rise to a moment along the field, and then undergoes a first order transition to a fan structure, in which the moments oscillate around the field direction. A further increase in the field reduces the opening angle of the fan which, in the absence of hexagonal anisotropy, goes continuously to zero. The hexagonal anisotropy causes the transition to the ferromagnet to be of first order or, if it large enough, eliminates the fan phase. A detail mean-field analysis of the transition between the (distorted) helix and the fan [17] has shown that long period combinations of the two structures, "the helifans", may appear as intermediate phases, in accordance with experimental findings in the case of Ho. Experiments on Dy do not show any indications of a helifan phase [18] consistent with mean-field analyzes which in this case predicts the helifan phases to be unstable. The magnetic structures in Er with a field applied in the basal plane have been studied by Jehan *et al.* [19] who followed the transformation of the cycloidal structure into different fan-like structures.

The symmetry of the hexagonal close-packed lattice of the heavy rare-earth metals allows the presence of magnetic two-ion interactions with only three-fold symmetry around the *c* axis [1]. These so-called trigonal terms manifest themselves only under certain conditions, for instance they cancel out in the ferromagnetic case. To first order their contribution to the free energy is

$$\Delta F \propto \sum_p (-1)^p J_\parallel J_\perp^3 \cos(3\phi_p). \tag{1}$$

where $J_\parallel$ and $J_\perp$ are the components of the magnetic moments parallel and perpendicular to the *c* axis, respectively, and ϕ_p is the angle which the basal-plane moments in the *p*th layer make with the *a* axis. Its change of sign from one layer to the next implies that the double-zone representation in the *c* direction ceases to be valid if this term is important. The first example of a trigonal coupling found in the heavy rare earths was the strong coupling between acoustic magnons and optical phonons propagating along the c direction in the ferromagnetic phase of Tb [2,20].

The high-resolution synchrotron x-ray studies of the intermediate cycloidal phase of Er by Gibbs *et al.* [8] indicated the presence of a number of long-period commensurable structures, which they explained to be regular arrangements of 3 or 4 layers of moments with an alternating positive or negative component along the c axis. The 7 layered or (43)-structure observed close to 50 K, thus comprises 4 hexagonal planes of moments with a positive c-component followed by 3 planes of moments with a negative c-component. As the temperature is lowered more and more triplets are replaced by quartets, until the system just above T_C only consists of quartets, which is the (44)-structure with $\mathbf{q} = \frac{1}{4}\mathbf{c}^*$. Cowley and Jensen [21] were able to determine the intensities of most of the harmonics in the commensurable structures of Er. The neutron-diffraction results were compared with the intensities of the corresponding structures predicted by a mean-field model. This comparison confirmed that the basic feature of the commensurable structures is the one proposed by Gibbs *et al.* Unexpectedly, the neutron experiments also showed the presence of scattering peaks along the c axis at $\pm(2n+1)q + mc^*$ for odd integer values of m. These indicate that the magnetic structures depend on the two orientations of the hexagonal layers in the hcp lattice, implying that the structures are distorted by trigonal couplings. 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. This means that, for instance, the (43)-structure should rather be denoted as the 2(43)-structure as it only repeats itself after each 14 layers. In the case of Ho, the trigonal couplings have a weak, second order, effect on the helical structure. The application of a field in the c direction enhances the structural distortions due to the possible trigonal interactions and by analyzing various experiments, Simpsons *et al.* [22] were able to establish that these couplings are also of significance in Ho.

The tendency for the period of the magnetic ordered structures in the rare earths to lock into values which are commensurable with the lattice, is determined by a complex interplay between the exchange coupling and the magnetic anisotropy terms. The commensurable spin-slip structures detected in Ho at low temperatures by Gibbs *et al.* [7] are explained by the large basal-plane six-fold anisotropy. This hexagonal-anisotropy energy and thus the bunching effect rapidly declines with the magnetization M, approximately like M^{21}, and the commensurable effects deriving from this anisotropy term should decrease when applying a field along the c axis. Nevertheless, neutron diffraction measurements indicate a strong increase with field of the lock-in of the 8 layered spin-slip structure at about 100 K [23] and of the 10 layered structure at about 40 K [24]. At 100 K the hexagonal anisotropy energy is so small that it only gives rise to a bunching of the order of a tenth of a degree and the spin-slip model of Gibbs *et al.* no longer applies. In comparison, the trigonal anisotropy energy decreases with a much lower rate with the magnetization (like M^7) and increases linearly with a c-axis field, as long as the c-axis moment is small, see Eq. (1). A mean-field analysis of the stability of the different commensurable structures in Ho has been carried out [25]. The model calculations show that the trigonal coupling is capable of explaining both observations. It was predicted that the width in temperature of the lock-in of the 8 layered structure should increase strongly, if the magnetic field is applied in a direction making a non-zero angle θ with the c axis, being proportional to $\sqrt{\theta}$ at small angles. As shown in Fig. 2 this prediction has been confirmed by Tindall *et al.* [26]. The experimental lock-in is not as strong as predicted in the middle of the interval (it is weaker by a factor of 0.6 in the helix). However, the over-all behavior is very similar to that predicted when including the trigonal coupling determined from the low-temperature experiments [22], and contrast strongly with the behavior expected when this coupling is neglected. Incidentally, the lock-in effect calculated in the case where the trigonal coupling is neglected, is due to the Zeeman energy.

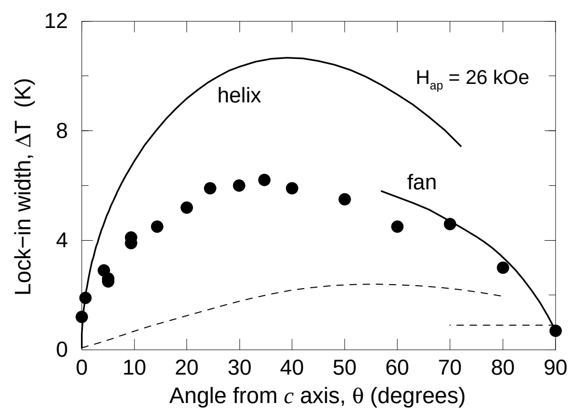

Fig. 2. The experimental results (solid circles) of Tindall *et al.* [26] show the lock-in temperature width of the 8 layered structure versus the angle θ of the applied field of 26 kOe in the c–b plane of Ho. The lines show the theoretical results obtained from the mean-field model calculations [25] with (solid lines) and without (dotted lines) the trigonal coupling.

The period of some of the commensurable structures observed in Ho and Er are remarkably long, as for instance the 2(444443) structure in Er with a repeat length of 46 hexagonal layers, and it is clear from the experiments that some disordering and hysteresis effects may occur noticeably at low temperatures. The neutron-diffraction experiment of Cowley *et al.* [27] shows that Ho may contain several domains with different spin-slip structures below 40 K. The regularity in the spacing of the spin-slip layers in the low-temperature spin-slip structures in Ho may be distorted. The x-ray diffraction measurements of Helgesen *et al.* [28] indicate that this is the case. They have observed a reduction of the longitudinal correlation length between 40 and 20 K by a factor of three, a reduction which is partly removed, when the spin-slip layers disappear at the lock-in transition to $\mathbf{q} = \frac{1}{6}\mathbf{c}^*$ at about 20 K. In the alloy $Ho_{0.9}Er_{0.1}$ the $\frac{7}{36}\mathbf{c}^*$ structure is stable at the lowest temperatures, and Rønnow [29] has observed that the widths of the neutron diffraction peaks in this phase are much larger for the higher harmonics than for the first one. The $\frac{7}{36}\mathbf{c}^*$ spin-slip structure consists of alternating two and three pairs of layers between the spin-slip planes, (2212221), and Rønnow has found that a structure in which the succession of the two sequences 221 and 2221 is completely random, predicts a diffraction spectrum close to the observed one.

III. MAGNETIC EXCITATIONS IN THE HEAVY RARE-EARTH METALS

The magnetic excitations in the heavy rare earths are in most cases well-defined spin waves at low temperatures [1]. Although Tm belongs to the heavy elements of the rare-earth series the RKKY-exchange interaction, being proportional to $(g-1)^2$, is weak compared to the crystal-field anisotropy energies. This implies that crystal-field excitations are important in Tm at elevated temperatures, both below and above T_N. In the low-temperature spin-wave regime the weakness of the exchange coupling implies that the bandwidth of the spin-wave spectrum, of the order of 2 meV, is small compared to the energy gap of approx. 8.5 meV [6,30]. One consequence of this is that the possible incommensurable broadening of the excitations occurring in the quasi-periodic regime (above 32 K), discussed for example by Lovesey [31], is of no importance in Tm. Using the infinite-continued-fraction formulation for the response function [1] the expansion rapidly converges in the case of Tm, because $\mathcal{J}(\mathbf{q})$ is small, and the response function becomes largely independent of whether the ordering is commensurable or not.

In Ref. [6] a complete RPA-theory for the magnetic excitations in Tm is developed. This theory accounts for the periodic magnetic ordering of the moments and for the interaction of the magnetic excitations with the phonons. Since the magnetic periodicity is seven times that of the lattice along the c axis, the spin waves are split into seven closely spaced energy bands. Because of the weak dispersion it was not possible to resolve experimentally the seven branches, instead, the calculated scattering function as a function of wave vector was compared directly with the experimental inelastic neutron-cross-section. A similar investigation of Er in its linearly polarized phase has been conducted by Nicklow and Wakabayashi [32]. The exchange coupling is relatively larger in Er than in Tm in comparison with the crystal-field splittings. However, the modifications of the spin waves in Er, due to the population of the different crystal-field levels, are important and dominate the possible effects caused by the quasi-periodic modulation of the magnitude of the moments.

The modulated (43)-structure in Tm is changed into a ferromagnetic structure when applying a c-axis field of more than 28 kOe. The model developed in Ref. [6] predicts this field to be about 42 kOe. Measurements [33] have later shown that there is a large change of the c-axis lattice parameter of about 0.7% at the transition, and the extra magnetoelastic energy gained in the ferromagnetic phase is able to explain the discrepancy. The spin-wave energies in the ferromagnetic phase of Tm have been measured [34]. The comparison with the excitations seen in the zero-field phase shows that both the crystal-field energy gap and the exchange coupling in the c direction is rather strongly modified at the transition. These changes must be the consequences of the disappearance of the superzone energy gaps and of the large shift in the c-axis lattice parameter.

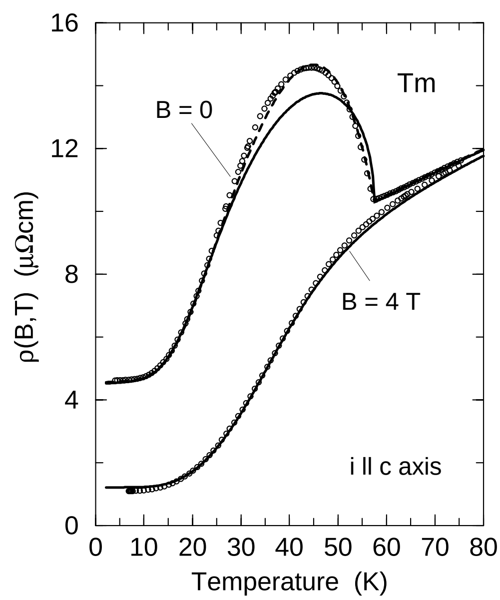

Fig. 3. The c-axis resistivity in Tm at zero field and in a field of 4 T applied along the c axis. The solid lines show the calculated results, and the dashed line indicates the quadratic rather than linear dependence on magnetization of $\rho(B,T)$ just below T_N (after Ref. [38]).

The transport properties and among them the electrical resistivity of the rare-earth metals were studied in many details during the 1960s, see for instance the review by Legvold [35]. The RKKY-coupling leads to a scattering of the electrons against the magnetic excitations. In addition, the effective number of conducting electrons is affected by the magnetic ordering. Particularly in the antiferromagnets, where the periodicity of the magnetic system introduces new zone boundaries with energy gaps in the conduction bands at the intersections. This so-called "superzone" effect was proposed by Mackintosh [36] and the theory worked out in detail by Elliott and Wedgwood [37]. The results shown in Fig. 3 are from a recent study of the electrical resistivity in Tm [38]. They give a clear illustration of the superzone effect by comparing the c-axis resistivity obtained in the modulated, zero-field, phase with the one obtained in the ferromagnet. The superzone effect vanishes in the latter case and the c-axis resistivity in the low temperature limit is increased by a factor of 3.7 by the superzones in the zero-field phase corresponding to a reduction of the Fermi-surface area perpendicular to the c-axis by 73%. The theoretical curves in Fig. 3 show that the resistance in Tm behaves in a way which is consistent with the RPA model developed by McEwen *et al.* [6]. A similar work on Er is in progress. According to model calculations [1] the cone phase in Er below 20 K is suppressed, when applying a hydrostatic pressure of about 2.5 kbar. Neutron-diffraction measurements at 11.5 and 14 kbar [39] show that the cycloid is indeed stable at 4 K at these values of the pressure. The resistivity of Er has been measured as a function of hydrostatic pressure [40]. The change of the magnetic phase from the cone to the cycloidal structure is observed to occur between 1 and 3 kbar. It is found that the superzone effects are a factor of 2–3 stronger in the cycloidal phase than in the cone. The combination of the calculated scattering rate with the Elliott-Wedgwood theory for the superzone effects accounts for most of the observations made in Er; except that the c-axis resistivity in the intermediate phase at ambient pressure is somewhat smaller than predicted.

IV. FILMS AND SUPERLATTICES OF RARE-EARTH METALS

The technique of molecular beam epitaxy (MBE) was utilized for producing the first superlattices of rare-earth materials, Dy/Y [41] and Gd/Y [42], in the middle of the 1980s. Since then, a large number of different rare-earth superlattices has been studied using x-ray and neutron-scattering techniques. Recent reviews are given by Majkzak [43] and by Cowley [44]. One of the central concerns in these systems is whether the magnetic ordering of one of the constituents is coherent over many bilayers or not. Some rare-earth superlattices have coherent magnetic structures even when the magnetic layers are separated by non-magnetic layers of more than 40 atomic planes, as for instance Dy/Y, Ho/Y, and Ho/Lu. In the Pr/Ho or Nd/Y superlattices, or in the Ho/Sc case the magnetic coherence length is of the same size as the thickness of the magnetic layers. In the latter systems the lattice mismatch is important, and the same may be the case in films of a rare-earth metals, where the mismatch may occur between the rare-earth and the substrate on which the film is grown, see for instance the discussion of the Er thin films in Ref. [45,1]. The magnetoelastic effects are important for the transition to the cone phase in Er, or for the transition to the ferromagnet in Dy, but, in general, the magnetoelastic interactions are weak in comparison with the other magnetic couplings, and the films behave in most respect like the corresponding bulk materials.

The MBE-technique may be used for producing well-defined films of alloys, which has been utilized in an investigation of the Ho_xEr_{1-x} [46] and Ho_xTm_{1-x} [47] alloy systems. Ho is an easy planar system whereas Tm and Er are both easy c-axis systems. This implies a *pentacritical* point in the both phase diagrams where the different anisotropy energies compensate each other. At this point, the plane in which the ordered moments are lying may rotate freely and five distinctly different phases are coexisting. The five phases are the paramagnetic, the longitudinally polarized, the cycloidal, the tilted cycloidal/helical, and the helical phase.

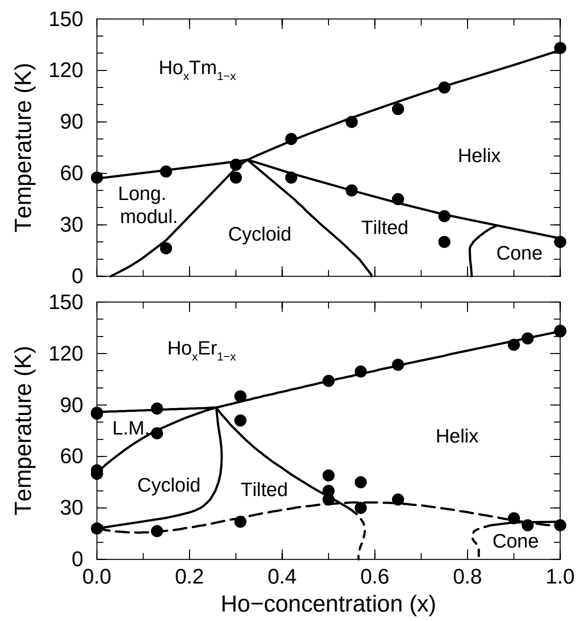

Fig. 4. The magnetic phase diagrams of Ho_xTm_{1-x} and Ho_xEr_{1-x} thin-film alloys. The circles show the transitions determined using neutron diffraction and the solid lines are mean-field calculations. The dotted lines in the Ho_xEr_{1-x} case indicate that these transitions are predicted to be of first order.

The experimental results obtained from neutron diffraction measurements are shown in Fig. 4. In the case of the Ho-Er alloy system the results are supplemented with results obtained from bulk measurements at $x = 0.5$, 0.9 and 1.0 [29]. The topology of the phase diagrams was established from a Landau expansion by Cowley *et al.* [46]. Here, the theoretical lines are determined using the general mean-field models of the pure systems, Ho, Er, and Tm, known from previous investigations. The results for the different alloys are obtained using the virtual crystal approximation, and the RKKY coupling, scaled with the appropriated $(g-1)$ factors, is considered to be determined by an averaged conduction-electron gas. This procedure leaves no free parameters for determining the phase lines for x different from 0 or 1. In the case of Er, the model is not developed so to predict accurately the first-order transition to the cone [1], and the dashed line indicating this transition in the Ho_xEr_{1-x} phase diagram is not the result of the calculations but a line through the points. The measurements shown in Fig. 4 demonstrate for the first time the occurrence of the tilted helix, most clearly in the case of Ho_xTm_{1-x}, which was advanced in 1972 by Sherrington [48] in relation to the phase diagram of pure Ho. The transition between the cycloid and the tilted cycloid, at which the plane of the cycloidal structures starts to tilt away from the c axis, is not isolated in the present experiments. A detection of this transition by neutron diffraction would require a preparation of the sample so to create a preferred orientation of the a–c cycloidal planes.

V. STRUCTURES AND EXCITATIONS IN THE LIGHT RARE EARTHS, Pr AND Nd

The magnetic structures in Nd have been studied extensively for more than 30 years [49–58]. The hexagonal B and C sites and the cubic A sites in the dhcp structure have different magnetic properties. The Néel temperature is 19.9 K, and below this temperature the moments on the hexagonal sites are ordered in a longitudinally polarized single-**q** structure with $\mathbf{q} \simeq 0.14\ \mathbf{a}^*$ [50]. The moments on the B and the C sublattice are antiparallel, and a small moment in predominantly the c direction is induced on the cubic sites. The transition is weakly discontinuous [51], and only about 0.8 K below T_N there is a new first-order transition to a structure, in which two of the three different single-**q** domains combines into a double-**q** structure [52]. At 8.3 K there is a transition to a triple-**q** phase, in which a single-**q** ordering of the moments on the cubic sites at a different $\mathbf{q} \simeq 0.18\ \mathbf{a}^*$, is superimposed on the double-**q** structure of the hexagonal moments. Finally, the low temperature phase, below 6.3 K, is a complex quadruple-**q** structure [53] with two ordering wave vectors associated with each of the two sublattices. Thermal expansion measurements [54] indicate one more transition at 5.8 K, and it is suggested that the **q**-vectors of the cubic moments may start to tilt out of the basal plane below this temperature, but the anomaly may also be the result of a lock-in transition to a commensurable structure.

The behavior of the transition between the single- and double-**q** structure is consistent with a mean-field analysis [55]. In the double-**q** phase, the moments corresponding to each of the two **q**-vectors rotate progressively towards each other as the magnetization increases. A numerical estimate [1], which predicts the transition to occur at about 0.9 K below T_N, indicates that the angle between the moments and their nearest b axis should be about 12° at maximum (8 K), corresponding to an angle of 96° between the two directions of the moments. The rotation of the moments is estimated to be accompanied by a rotation of the **q**-vectors by an angle that is smaller by a factor of about three, in accordance with the observations. It is worth noting that the reason for the rotation of the moments, and for the stability of the double-**q** structure in comparison with the single-**q** structure, is the change in entropy [1,55]. The small value of the magnetization implies that the sixth-order hexagonal anisotropy is of no importance close to the transition, which is in contradiction with the proposal made in Ref. [56].

Lebech *et al.* [57] have examined whether the different multiple-**q** structures in Nd may be commensurable with the lattice. Their neutron-diffraction experiments show clear lock-in effects in the low-temperature phases below 8.3 K. In the high temperature phases they suggest the presence of a series of two-dimensional higher-order commensurable structures, where the magnetic unit cell is rotated a certain angle around the c axis relatively to the crystallographic unit cell, so that commensurable structures are formed although each of the two basal-plane components of **q** appears to be incommensurable. The high-resolution measurements of the ordering wave vector made by magnetic x-ray diffraction [58] indicate a smooth variation of **q** between 11.5 K and T_N. The absence of lock-in effects in this experiment may be the result of the close spacing of the large number of higher-order commensurable structures. It does not necessarily indicate that the lock-in energies of the epitaxial structures proposed by Lebech *et al.* are negligible.

Pr have the same dhcp lattice structure as Nd, and the magnetic two-ion interactions seem to be very similar in the two metals. The ground state of both the cubic and the hexagonal ions in Pr is a singlet, and the maximum value of the exchange coupling is about 92% of that required for inducing a magnetic ordered state on the hexagonal sites. The coupling is close to the threshold value and small modifications, like a uniaxial pressure of about 0.7 kbar or the introduction of a small percentage of Nd ions, may change the situation [1]. Without external disturbances, magnetic ordering is produced via the hyperfine interaction between the $4f$-electrons and the nuclear spins, and heat capacity measurements [59] indicate the Néel temperature to be 42 mK. As in Nd, the moments on the hexagonal ions in Pr order in a longitudinally polarized structure with $\mathbf{q} = 0.13\ \mathbf{a}^*$, possibly accompanied by an induced moment along the c axis on the cubic ions. It is not yet known, whether the structure just below T_N is a single- or triple-**q** structure, or whether, like in Nd, there is a transition between a single- and double-**q** structure at a lower temperature. The ordered phase has recently been studied by neutron diffraction [60,61] well below T_N. The value of the first harmonic of the moments is determined to be 0.54 μ_B at 10 mK, when assuming an equal population of domains, and as shown in Fig. 5 it was found that the antiferromagnetic phase is quenched, when applying a field of about 4.2 kOe along an a direction. These numbers may be compared with the results derived from the MF/RPA-model presented in Ref. [1], which are shown by the solid line in Fig. 5. The calculated moment is 0.63 μ_B and the critical field is 5.4 kOe at 10 mK. Both values are larger than the experimental ones, although the exchange coupling is adjusted so that the energy gap in the excitation spectrum at the ordering wave vector has the right value, in which case the model correctly predicts $T_N = 43$ mK (using $A = 4.394$ meV). The discrepancies indicate that the single-site fluctuations [1] may be important in the determination of the field dependence of the ordered moment.

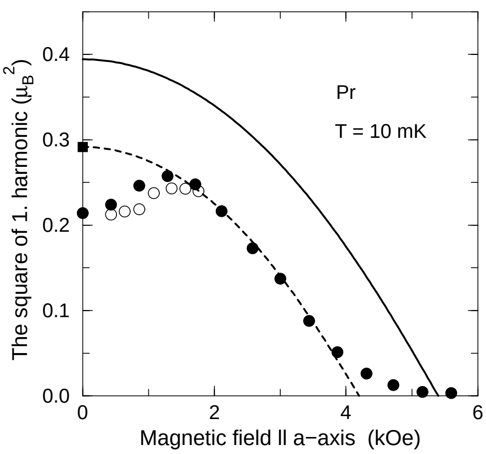

Fig. 5. The square of the first harmonic of the modulated magnetic moments on the hexagonal sites in Pr as a function of a field applied along an a axis. The circles are the experimental results from Ref. [60,61]. The filled (open) circles are the results obtained for decreasing (increasing) values of the field. The square is the result obtained under zero-field conditions. The experimental results indicate that domains are formed in the field experiment below 2 kOe. The solid line is the variation predicted by the MF/RPA model for Pr [1], and the dashed line indicates the experimental variation.

Most of the magnetic properties of Pr are well understood in terms of the MF/RPA-model, if including the leading-order corrections due to the single-site fluctuations in the description of, for instance, the linewidth of the magnetic excitations [1,62]. However, there are two phenomena in Pr which the "standard model" cannot easily explain. One is the broad quasielastic or central peak which appears at about 13 K as a ring in reciprocal space around Γ, which is perpendicular to the c axis and has a radius $|\mathbf{q}| \simeq 0.1\ a^*$, see the discussion in Ref. [1]. The other phenomenon is the two extra excitations observed a couple of years ago, hybridizing at small wave vectors with the normal magnetic excitations which propagate on the hexagonal and on the cubic sites in the paramagnetic phase [63]. At 4.2 K the linewidth of the extra excitations shows a sharp drop when applying a field of about 15 kOe along the a axis. The quasielastic peak disappears [61] at this field (at 1.8 K) suggesting a connection between the two phenomena. The coincidence might be accidental, though it is proposed that both phenomena may involve a weak hybridization of the $4f$ electrons with the conduction electrons. Another proposal made recently [64] is that the quasielastic peak is due to transverse fluctuations locked to the critical longitudinal fluctuations in a kind of a spin-slip configuration. However, this theory involves a number of postulates in the analysis of the dynamics, and it is inconsistent with the dominance of the quasielastic peak above 4 K. In addition, the excitation spectrum indicates that the transverse fluctuations are centered around $|\mathbf{q}| \simeq 0.07\ a^*$, not $0.1\ a^*$, and should be much weaker than the longitudinal ones in the critical regime. The energy difference between, for instance, the dhcp and the Sm-structure is small, and stacking faults in the $ABAC$-sequence of hexagonal layers along the c axis may occur relatively frequently. In case two layers, B and C, with local hcp symmetry happen to be neighbors, the effective $\mathcal{J}(\mathbf{q})$ within the two layers is probably increased. An increase by 23% would create a double-layered magnetic system with an estimated MF transition temperature of 14 K, a zero-temperature moment of 1.9 μ_B/ion, and a critical a-axis field of 14 kOe at 1.8 K. These numbers indicate that a distribution of hcp double-layers with a density of about one per 1000 layers, is a promising candidate for explaining the quasielastic peak in Pr. Although new normal modes would be associated with such a partial ordering, their intensities are probably all too weak to be able to explain the extra modes observed in Pr.

VI. CONCLUSIONS

The rare-earth metals display a large number of fascinating and exotic magnetic ordered structures, helifans, spin-slip and other commensurable structures, multiple-$\mathbf{q}$ structures, and distorted structures induced by trigonal couplings. The RKKY-interaction is of long range and excluding the critical properties, the MF- and RPA-theories reasonably account for many of the properties of these systems. The parameters in the magnetic Hamiltonian are adjustable. It would be desirable to calculate these parameters from first principle, and some progress has been made recently [65]. Many aspects of the band properties of the conduction electrons, in the regime where the $4f$-electrons may be treated as localized core electrons, are well understood, see e.g. Ref. [1]. This may be utilized for calculating the change of the Fermi surface areas due to the magnetic modulated moments in Tm and Er, as to be compared with the large reductions indicated by the resistivity measurements. A first-principle calculation of the RKKY interaction is a demanding task, however, a large amount of experimental data is available, and it would be valuable to get more theoretical insight into the behavior of the RKKY-interaction both in the homogeneous bulk systems and in the inhomogeneous superlattices.

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