Magnetic structures and excitations in rare-earth metals

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Overview of the talk

- Introduction
- Long-period magnetic structures in the heavy rare-earth metals
- Magnetic excitations in the heavy rare-earth metals
- Films and superlattices of rare-earth metals
- Structures and excitations in the light rare earths, Pr and Nd
- Conclusions

- The magnetic and superconducting properties of the rare-earth nickel borocarbides are discussed by Morten Ring Eskildsen at this conference.

Introduction

Hamiltonian:

$$\mathcal{H} = \mathcal{H}_{\rm CF} + \mathcal{H}_{\rm me} + \mathcal{H}_{\rm RKKY} + \mathcal{H}_Z + \mathcal{H}_T + \cdots$$

The crystal-field Hamiltonian of the rare-earth ions is due to the different non-spherical distribution of the localized *4f*-electrons in the different $|m_J>$ states of the ground-state J-multiplet,

$$\mathcal{H}_{\rm CF} + \mathcal{H}_{\rm me} = \sum B_l^m O_l^m(\boldsymbol{J}_i) + \sum B_l^m(\alpha\beta) O_l^m(\boldsymbol{J}_i)\epsilon_{\alpha\beta}$$

The CF anisotropy terms are determined by symmetry (l= 2, 4, 6; m = 0, 6 in the hcp-lattice), and the last magnetoelastic term becomes important if the symmetry is reduced.

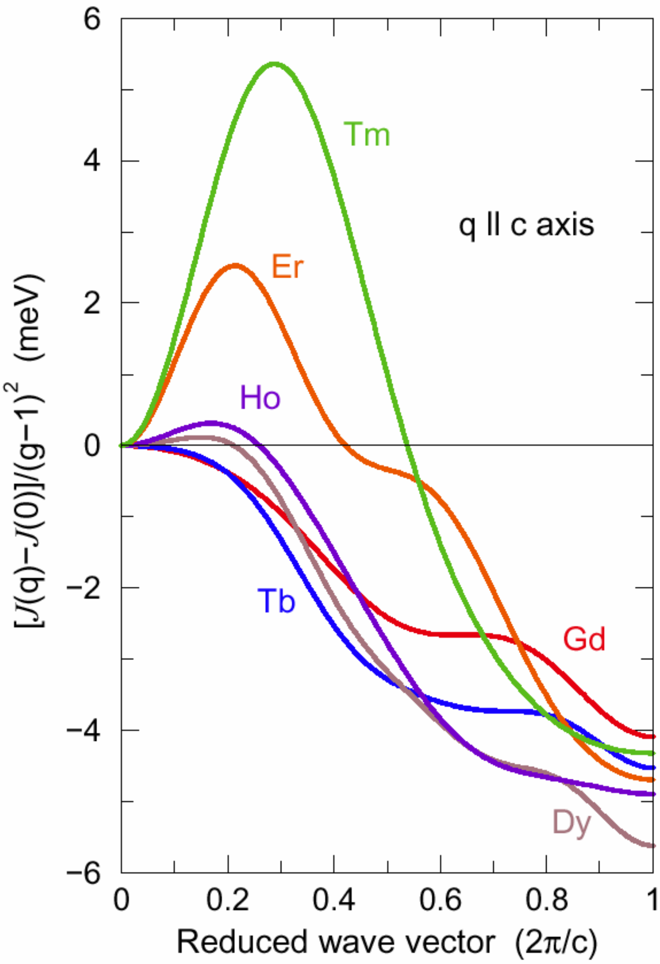
The exchange interaction between the conduction electrons and the *4f*-electrons leads effectively to a Heisenberg coupling between the *4f*-moments, the RKKY-interaction:

$$\mathcal{H}_{\rm RKKY} = -\tfrac{1}{2}\sum_{i,j} \mathcal{J}(ij) \boldsymbol{J}_i \cdot \boldsymbol{J}_j$$

where the Fourier transform of the coupling is determined by the susceptibility of the conduction electrons

$$\mathcal{J}(\boldsymbol{q}) = \frac{V(g-1)^2}{N\mu_B^2} \sum_{\boldsymbol{\tau}} \left[|I(\boldsymbol{q}+\boldsymbol{\tau})|^2 \chi_{\rm c.el.}(\boldsymbol{q}+\boldsymbol{\tau}) - \sum_{\boldsymbol{q}'} |I(\boldsymbol{q}'+\boldsymbol{\tau})|^2 \chi_{\rm c.el.}(\boldsymbol{q}'+\boldsymbol{\tau}) \right]$$

$\boldsymbol{\tau}$ is a reciprocal lattice vector, and g is the Landé-factor.



The figure shows the exchange coupling, $[\mathcal{J}(\boldsymbol{q})\text{-}\mathcal{J}(\boldsymbol{0})]/(g\text{-}1)^2$ with $\boldsymbol{q}$ parallel to the c axis, as determined experimentally in the magnetic heavy rare-earth metals (hcp-lattices) from the magnetic excitation (spin-wave) energies.

Other two-ion interaction terms in the Hamiltonian:

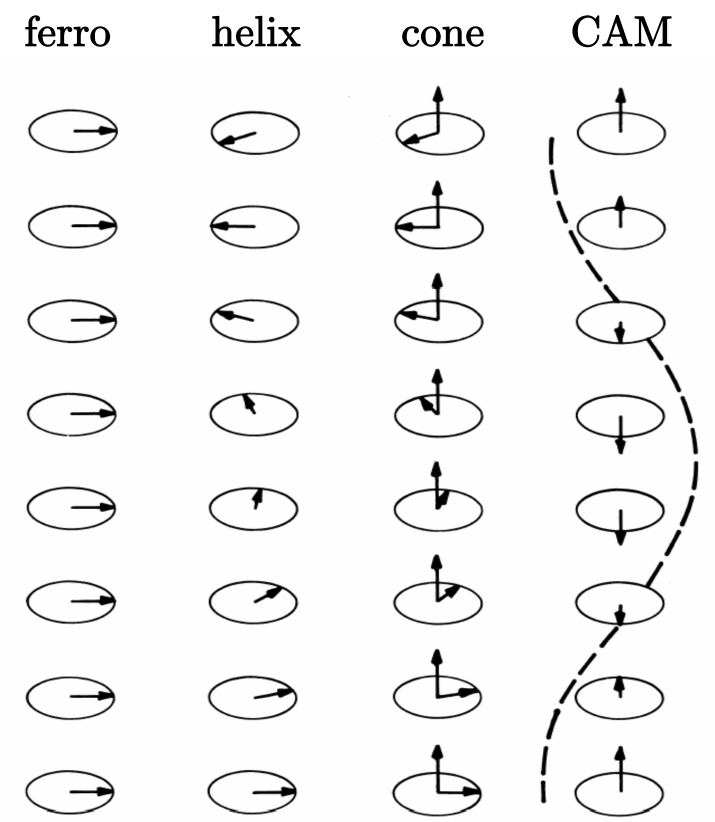
$$\mathcal{H}_T = \frac{1}{2}\sum_{ij}\sum_{ll'mm'} K_{ll'}^{mm'} O_l^m(\boldsymbol{J}_i) O_{l'}^{m'}(\boldsymbol{J}_j)$$

Among which the most spectacular ones are the *trigonal* terms with $m + m' = 3$. These terms distinguish between the two hexagonal sublattices in the hcp-structure and lead to a coupling of acoustic and optical excitations propagating in the c direction.

Long-period magnetic structures in the heavy rare earths

W.C. Koehler (in *Magnetic Properties of Rare Earth Metals*, ed. R.J. Elliott, Plenum Press, 1972):

"...the full panoply of exotic spin configurations in these metals."

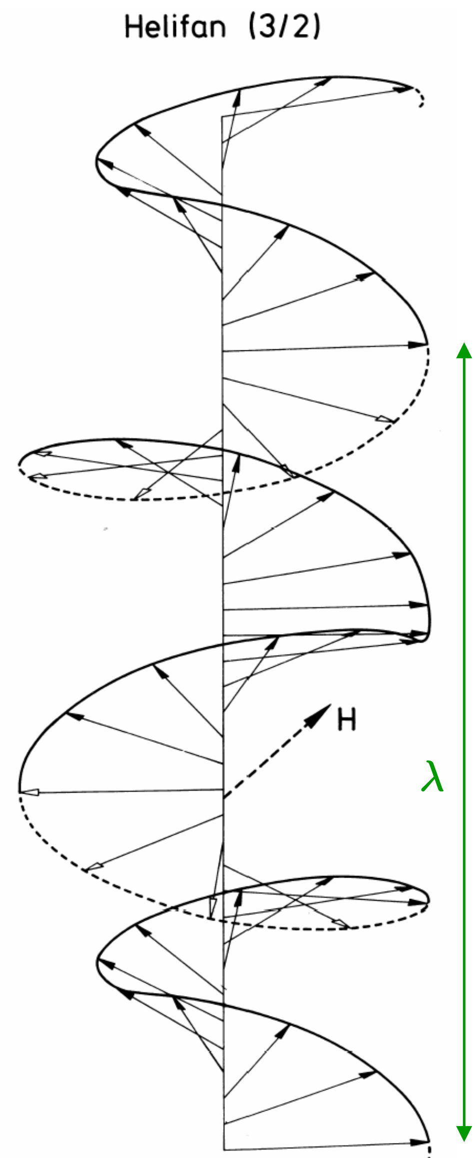


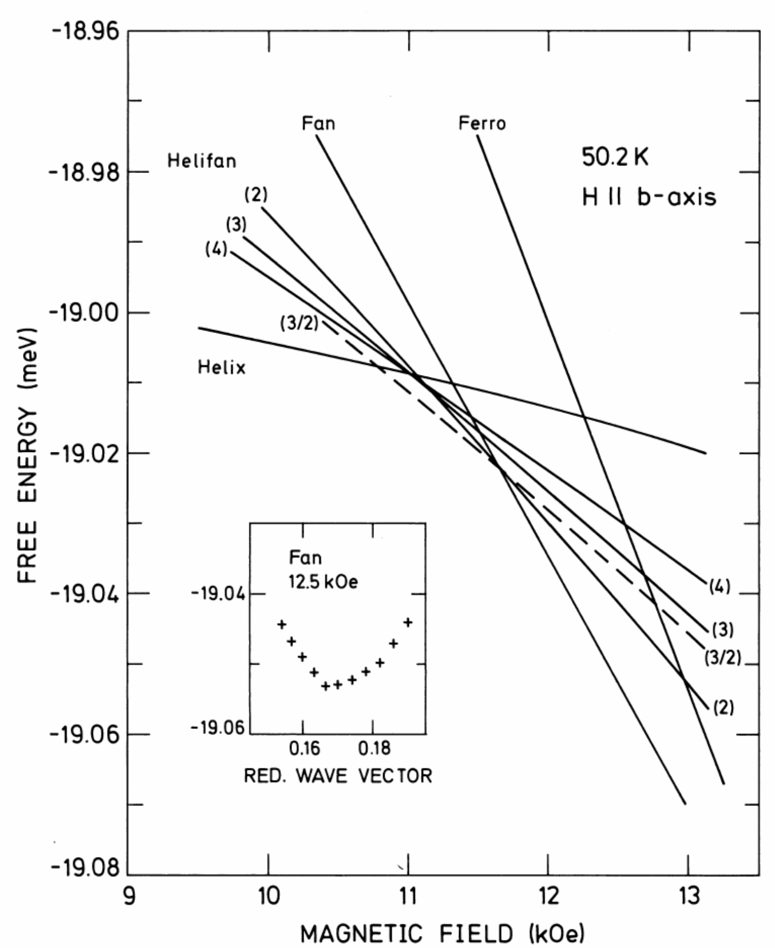
Since 1972 a rich variety of more complex magnetic structures has been found by neutron and magnetic x-ray scattering:

- The 'helifan' in Ho.
- The 'wobbling' cycloidal structure in Er.
- The 'tilted helix (or cycloid)' in the phase diagrams of Ho-Tm and Ho-Er thin film alloys.
- 'Spin-slip' and other commensurable structures in Ho and Er.
- Multiple-$\boldsymbol{q}$ structures in Nd.

The helifan (3/2) in Ho

Mean-field calculations indicate that the helifan(3/2) in Ho is stable at intermediate values of a field applied in the basal plane. The extra phase has been detected in resistivity, X-ray- and neutron-diffraction studies. The calculated scattering intensities of the helifan(3/2) agree well with the observations. The changes of the ordering wave vector observed at the transitions are also reproduced by the model.

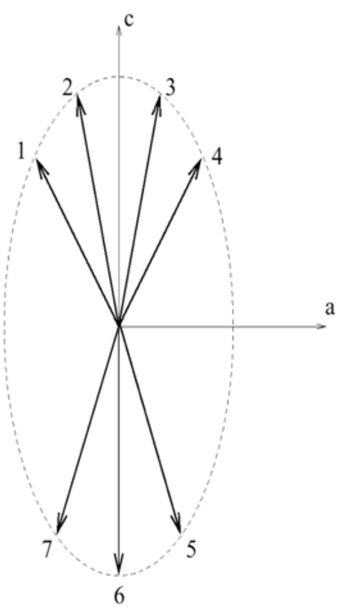




The period of the helifan(3/2) is three times longer than the period of the corresponding helix.

The `wobbling´cycloid in Er

Erbium orders in the c-axis modulated CAM structure at T_N = 84 K with an ordering vector $\boldsymbol{Q}$ close to 2/7 $\boldsymbol{c}^*$. Below T_N' = 52 K one component in the basal plane (along the a axis) is ordered at the same wave vector as the c component leading to an elliptically cycloidal structure. At T_C = 18 K there is a transition to the c-axis cone phase.

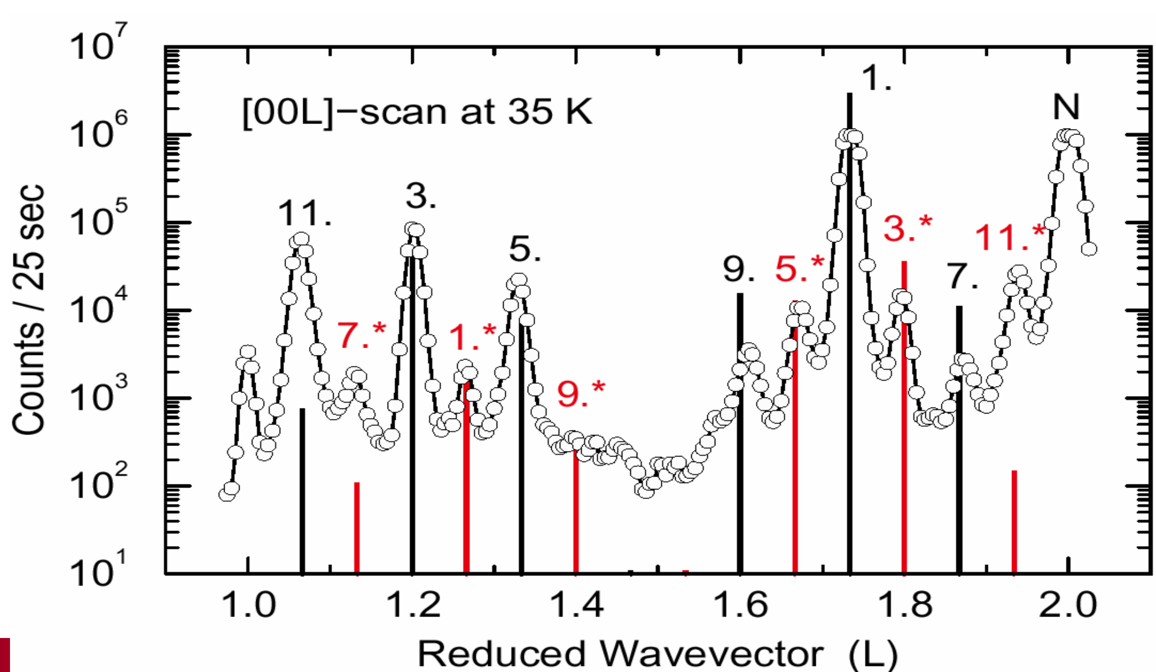


The [00L] scan at 35 K shows that the basal plane moments are ordered with the commensurable wave vector $\boldsymbol{Q}$= 4/15 $\boldsymbol{c}^*$, corresponding to the (4443) structure. Scattering peaks are observed at

$$q \;= (2p+1)Q + 2nc^*$$

$$q^* = (2p+1)Q + (2n+1)\; c^*$$

The odd harmonics at q^* involves odd multiples of c^*, which may only be introduced by the trigonal interactions implying an ordering of the b component perpendicular to the cycloidal plane.



The phase diagrams of Tm-Ho and Er-Ho thin-film alloys

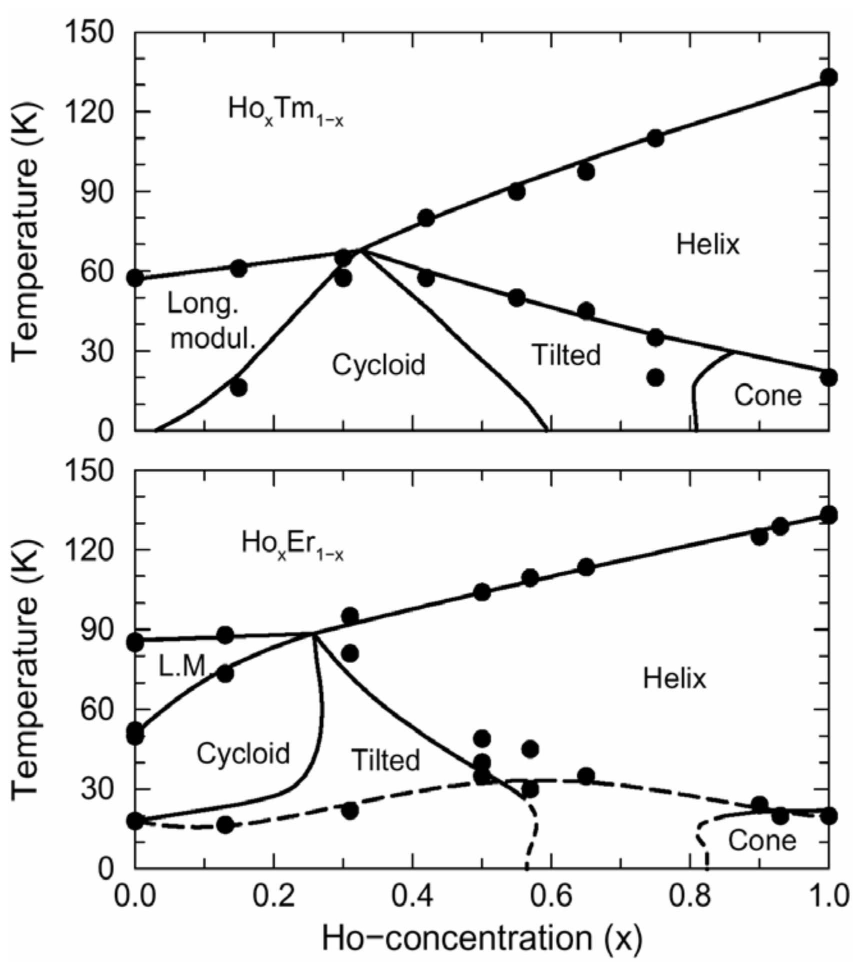
The technique of molecular-beam-epitaxy has open up a new field: the study of rare-earth superlattices and films. It has been used for a neutron-diffraction study of films of Ho-Tm and Ho-Er random alloys.

The c axis is the easy axis in Tm and Er but is the hard one in Ho, and the results show for the first time the presence of

- the *tilted helix (cycloid)*

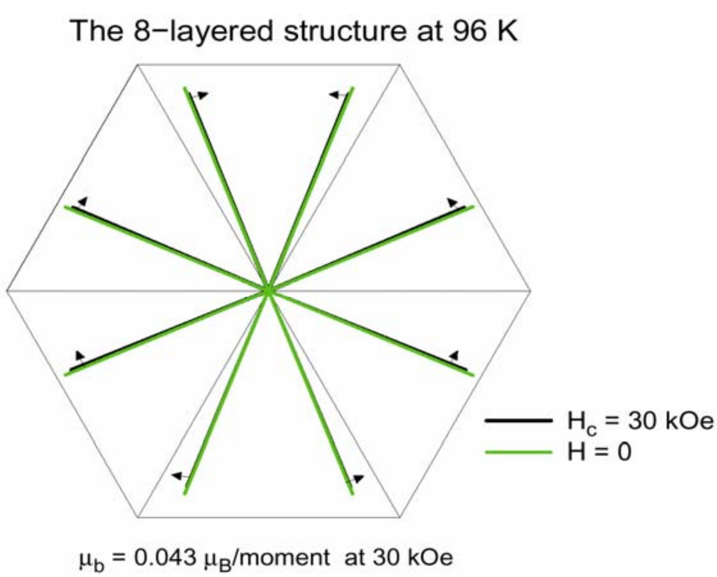
and

- a *pentacritical point*



The commensurable structures in Ho

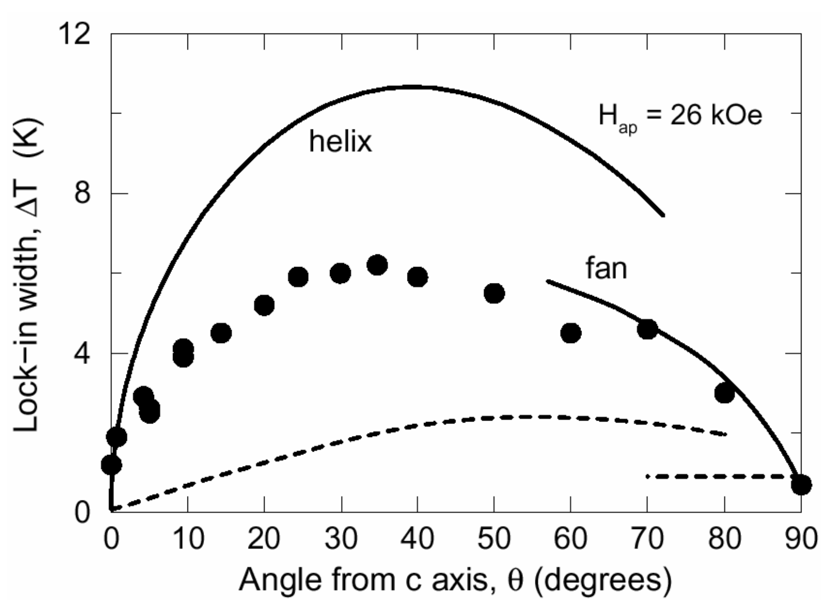
A great number of commensurable "spin-slip" structures are observed at low temperatures in Ho. The distortions of the helical structures are due to the hexagonal anisotropy. Around 100 K this anisotropy is negligible. Nevertheless, the 8-layered structure shows a strong lock-in in the presence of a field which has both a *c*- and a basal-plane component.



This lock-in effect is produced by a trigonal coupling. To first order:

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

and in the case of $Q = 1/4\ c^*$ it induces a (3+1)th higher harmonic translated an odd integer times c*, i.e. a ferromagnetic component.

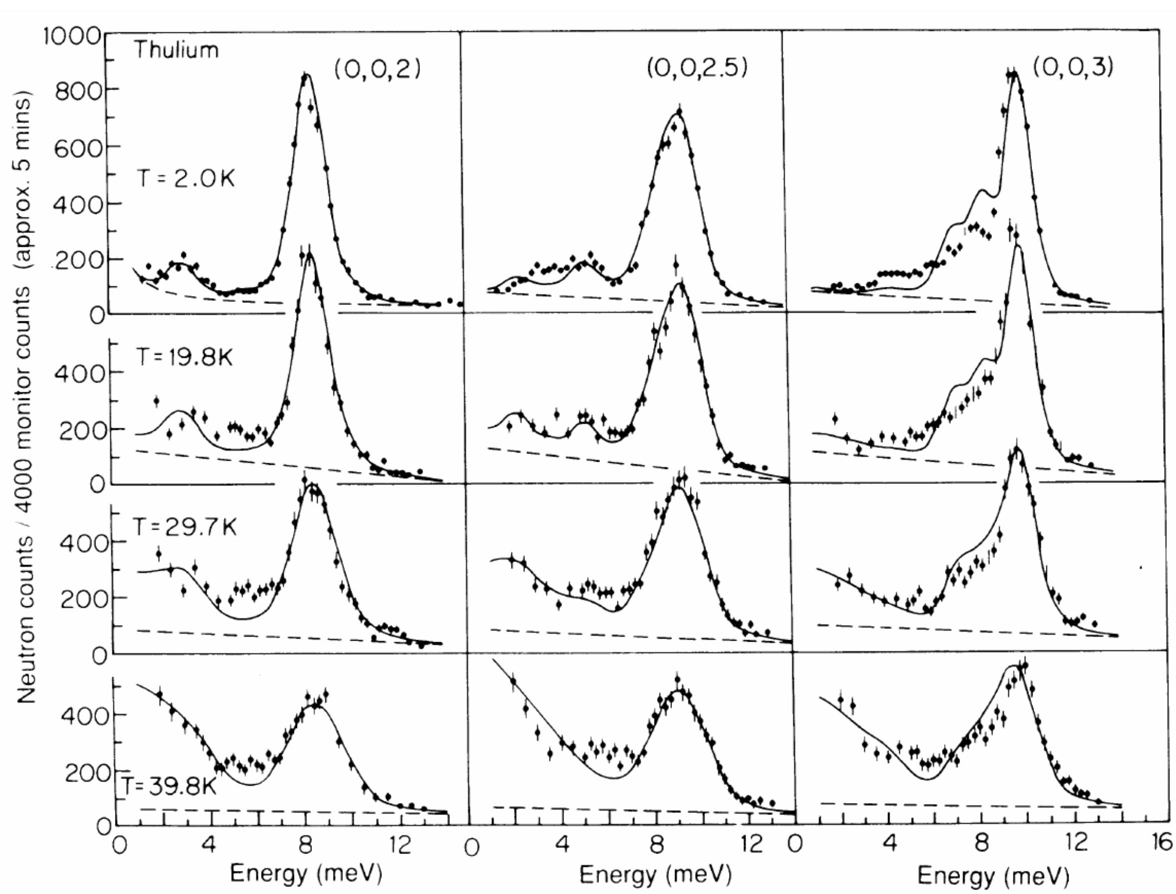


Magnetic excitations

The magnetic excitations in the heavy rare earths are well-defined spin waves at low temperatures. In Tm the RKKY-exchange interaction, being proportional to $(g\text{-}1)^2$, is weak compared to the crystal-field anisotropy energies. At low temperatures the c-axis moments are ordered in a square wave with a period of 7 hexagonal layers, i.e. 4 up and 3 down.

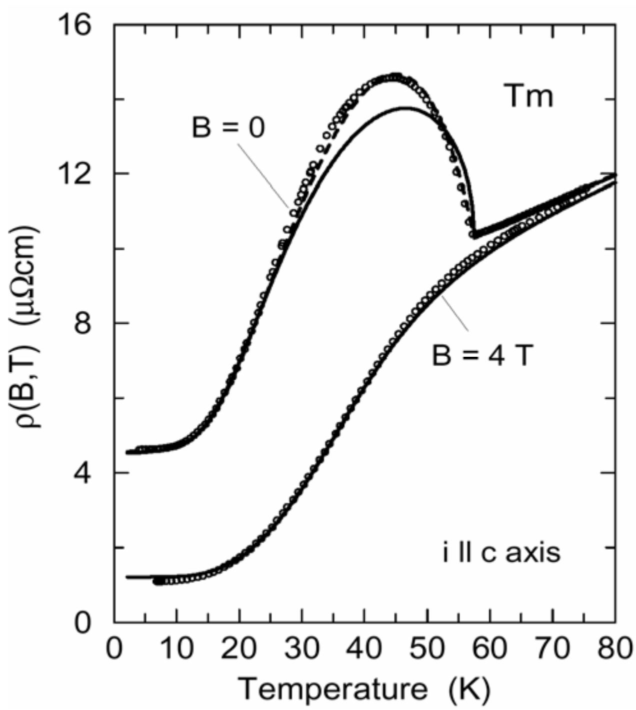
The spin waves in Tm split into 7 closely spaced energy bands.

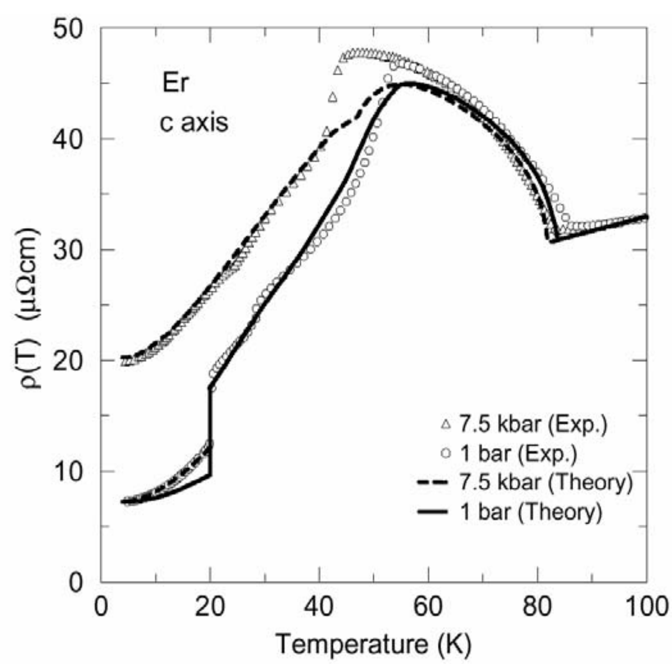
Thulium:



Magnetoresistivity

The RKKY interaction between the conduction and the 4f-electrons implies that the magnetic excitations scatter the conduction electrons. Furthermore, a periodic ordering of the magnetic moments introduces energy gaps at the new 'superzone' boundaries which may reduce strongly the density of states at the Fermi surface (the theory of Elliott and Wedgwood).





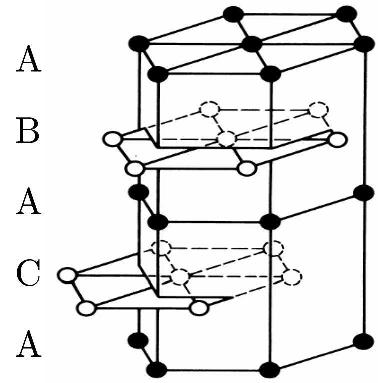
The resistivity measurements on Tm and Er give a clear illustration of the superzone effect.

The light rare-earth metals Pr and Nd

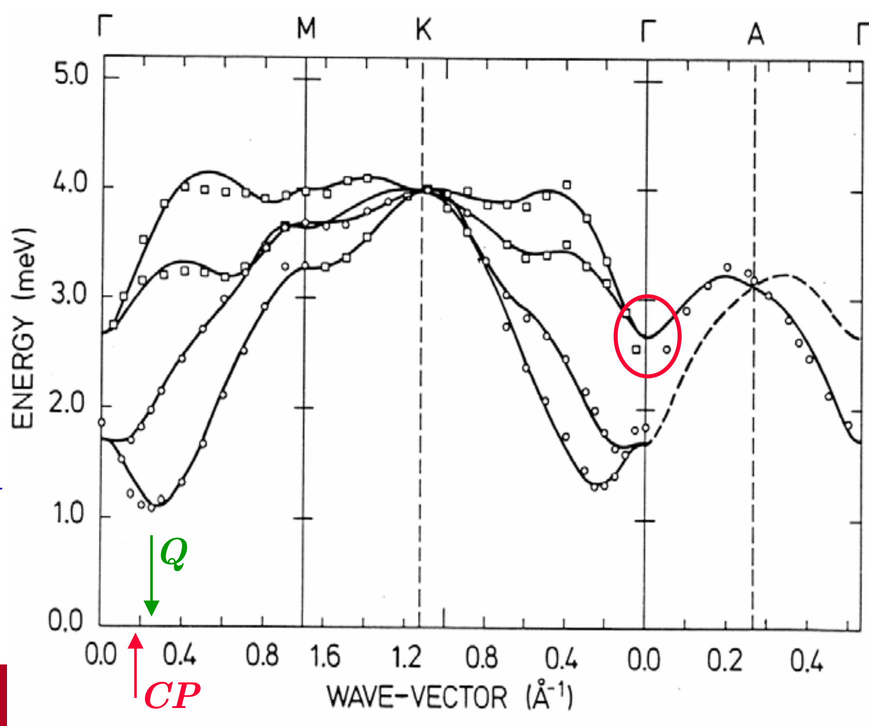
Most of the magnetic properties of the two light rare-earths are accounted for in terms of the MF/RPA-theory. However, there are two, possibly related, observations in Pr which the 'standard model' may not easily explain:

- A broad quasielastic or central peak which appears below about 15 K, in the shape of a ring in reciprocal space around Γ perpendicular to the c axis and with a radius of about 0.1 a^*.
- Two extra excitation modes hybridizing at small wave vectors with the normal magnetic modes propagating on the hexagonal and the cubic sites.

The lattice of Pr and Nd is the double hexagonal close-packed structure (dhcp) with A-sites having nearly cubic surroundings and the B and C sites with hexgonal surroundings.

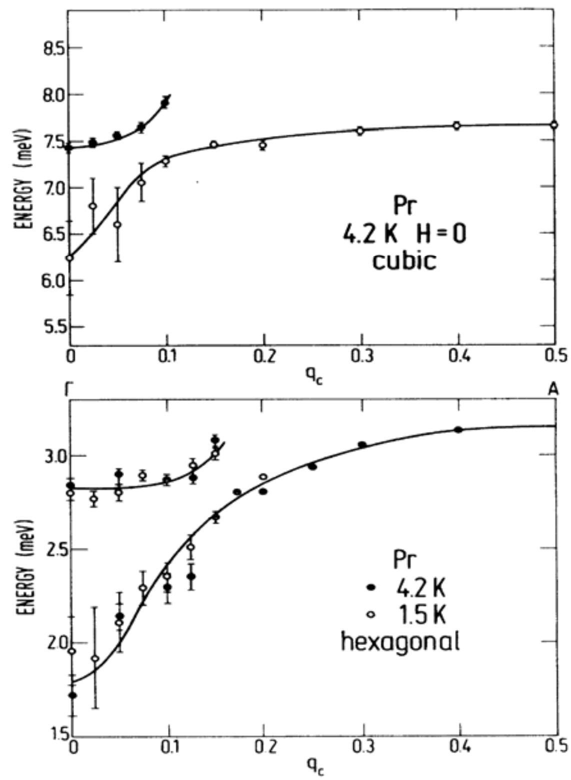


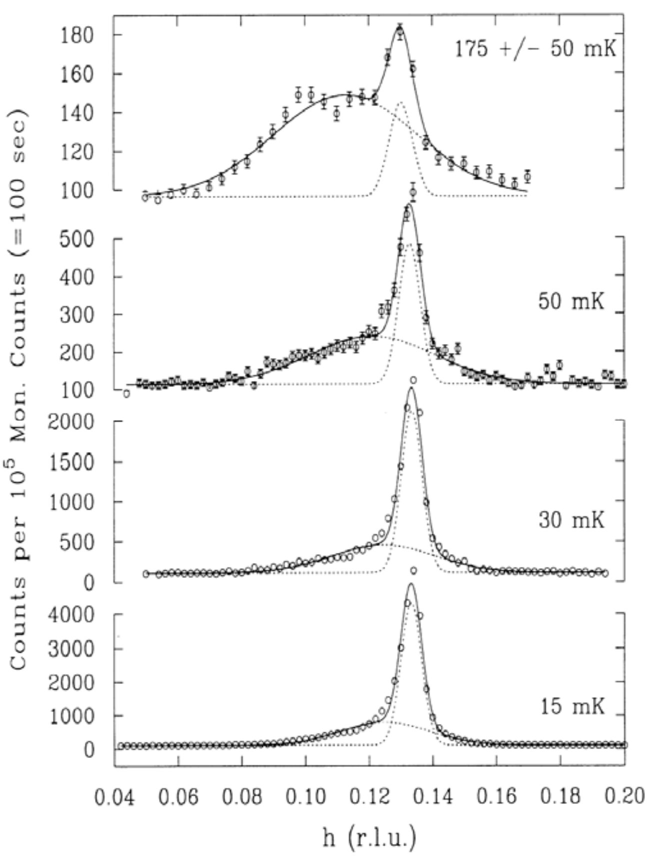
- Pr (J=4) is a singlet-ground state – excited doublet system (on the BC-sites).
- The (anisotropic) RKKY interactions is 92% of the threshold value for inducing a magnetic ordering of the BC moments.
- The system eventually orders at about 45 mK due to the hyperfine interaction.



Non-standard behavior of Pr

The linewidth of the two new excitations shows a sharp drop when applying an a-axis field of about 15 kOe at 4.2 K. The extra quasielastic peak disappears at this field suggesting a connection between the two phenomena. It is proposed that they both derive from a weak hybridization of the $4f$ and the conduction electrons.





Another possibility is the stacking faults in the $ABAC$ sequence which place two hexagonal layers as neighbors resulting in a larger value of the effective RKKY interaction within the two layers. A 25% increase of the coupling would produce a magnetic double layer with an estimated ordering temperature of 15 K and a critical field of 14 kOe (at 1.8 K). A density of one double layer per 1000 layers seems to be sufficient to explain the intensity of the quasielastic peak.

Conclusions

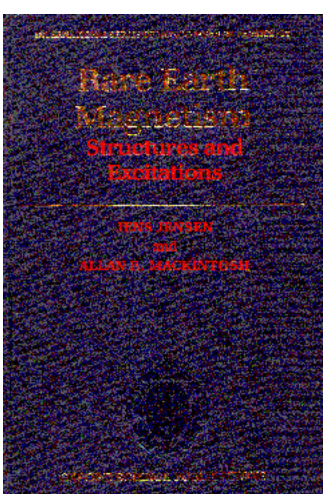
- The rare-earth metals display a large number of magnetic ordered structures: ferromagnet, helix, cone, CAM, cycloid, tilted helix (cycloid), different commensurable structures, wobbling cycloid and other distorted structures induced by the trigonal interactions, fans and helifans.

- The MF/RPA theories account reasonably well for most properties, except for the two non-standard phenomena in Pr.

- Wanted: First-principle electronic band calculations of the phenomenological crystal-field parameters and the RKKY interactions in the different metals

- A detail exposition of the theoretical and experimental understanding of the rare-earth metals is presented in

J. Jensen and A. R. Mackintosh, *Rare Earth Magnetism: Structures and Excitations* (Clarendon Press, Oxford, 1991).



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