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Theory I

Phenomenological Discussion of Magnetic Ordering in the Heavy Rare Earth Metals

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A phenomenological Hamiltonian is derived and applied to the magnetic properties of the rare-earth metals. Indirect exchange through the conduction electrons is thought to give the screw spin arrangement and the hexagonal crystal field appears to account for the anisotropy.

The elements in the second half of the rare-earth series Gd-Tm are all very similar, having a h.c.p. structure. The unfilled shell of f electrons appears to be intact and leads to interesting magnetic properties. The magnetic ordering is found by neutron diffraction to vary considerably from element to element, and most elements show several magnetic phases[1]. The most striking feature of this ordering is that the phase which appears at the highest T shows a wave-like modulation in one or more of the components of magnetic moment. In each plane the moments are always ferromagnetically aligned but there is a variation between planes. The h.c.p. structure also gives a very strong magnetic anisotropy relative to the hexagonal c axis and a weaker hexagonal anisotropy in the plane.

It can be seen that there are microscopic interactions present in these crystals which will produce these effects, and that the variation between elements is a natural consequence of the different f^n configurations involved. The essential interactions are:

1. A long-range oscillatory exchange interaction with a maximum Fourier component $\mathscr{J}(\boldsymbol{q})$ at $\boldsymbol{q}\neq 0$, which produces an ordering with a $e^{i\boldsymbol{q}\cdot\boldsymbol{r}}$ variation. The Yosida interaction[2] via the conduction electrons has this essential property and is known to give a satisfactory variation in magnitude along the series[3].

2. The crystalline electric field set up by the ionic charges, which in the hexagonal symmetry has the form[4]

$$V=V_2{}^0\alpha Y_2{}^0(\boldsymbol{J})+V_4{}^0\beta Y_4{}^0(\boldsymbol{J})+V_6{}^0\gamma Y_6{}^0(\boldsymbol{J})+V_6{}^6\gamma Y_6{}^6(\boldsymbol{J})\ .$$

The $Y_6{}^n(\boldsymbol{J})$ are operator equivalents and α, β, γ are constants of proportionality which vary with the electronic configuration. The $V_6{}^n$ have been evaluated on a model assuming $+3e$ charges on the nearest-neighbour sites. The essential features of the axial anisotropy follow if $V_2{}^0$ is largest and negative (as it is on the model). This forces the electronic charge cloud into the basal plane. The relative directions of $\boldsymbol{J}$ and the quadrupole moment are determined by the sign of α; it is $+ve$ for Tb, Dy, Ho whose moments are indeed in the plane, and $-ve$ for Er and Tm whose moments tend to be more along the c axis. The higher axial harmonics may give a tendency for the moments to point in intermediate directions—the $+ve\,V_6{}^0$ given by the model is of the right sign to produce the low T phase in Er where the moments directions lie in a cone about the c axis[1]. The $-ve\,V_6{}^6$ of the model is also of the right sign to account for the hexagonal anisotropy observed in Dy[5] and Ho.

3. More complicated interionic interactions—direct exchange and quadrupole-quadrupole interaction are rather difficult to calculate directly. Any interaction quartic in the angular momentum operators will, in the molecular field approximation, give an additional energy E quartic in the magnetisation M. Thus $E=JM^2+CM^4=J_{\rm eff}M^2$, where $J_{\rm eff}=J+CM^2$ depends on T through M. The observed variation of $\boldsymbol{q}$ with T does appear to be of this form.

A simplified model with these types of interaction can be evaluated in the Bragg-Williams approximation. There are two different cases.

A. Moments in the plane: In this case the theory is equivalent to the molecular field approximation. The high T phase is a spiral,

$$\mu_x{}^i=\mu M_x{}^i=\mu M\cos qz_i\ ;$$
$$\mu_y{}^i=\mu M_y{}^i=\mu M\sin qz_i\ ,$$

but at low T the hexagonal anisotropy causes a transition to ferromagnetism as proposed by Enz[6]. Because of the anisotropy, we have taken an effective exchange between layers of the form

$$\sum_{i,n} B_n(M_x{}^iM_x{}^{i+n}+M_y{}^iM_y{}^{i+n})\ .$$

Using the method of Yoshimori[7] and others, there is enough information in Dy to determine a 3-parameter theory, and we find $B_0=-55°\text{K}$, $B_1=170°\text{K}$, $B_2=-(57+11M^2)°\text{K}$.

B. Moments along the axis: In this case, assuming

$$\mu_z{}^i=\mu M_z{}^i=\mu M\sin(qz_i+\delta)\ ,$$

it is necessary to extend the Bragg-Williams approximation by evaluating the entropy,

$$S=Nk\ln 2-k\sum_i(1+M_z{}^i)\ln(1+M_z{}^i)$$
$$+(1-M_z{}^i)\ln(1-M_z{}^i)\ ,$$

which can be done in closed form. Minimising the free energy gives

$$M=2[(1-kT/2A)(kT/2A)]^{1/2}\ ,$$

when an Ising interaction between layers $\sum_{i,n} A_nM_z{}^iM_z{}^{i+n}$ has been used. This phase has a Néel temperature,

$$T_N=2A=A_0+2A_1\cos\alpha+2A_2\cos 2\alpha\ .$$

It is stable because the comparative low energy has been compensated by a large entropy to lower the free energy at high T. At $T=\frac{1}{2}T_N$, M reaches its maximum value, and below this it tends to an antiphase domain structure. At some temperature, above or below $\frac{1}{2}T_N$ depending on the A's, the free energy of the ferromagnetic state becomes lower. In Er, although the planar components also order[1], there are three rather similar phases, and one may determine

$$A_0=-40°\text{K}\ ,\quad A_1=80°\text{K}\ ,\quad A_2=-30°\text{K}\ .$$

These are quite similar to the B's in Dy and lend some credence to the model, although the $-ve$ sign of A_0 and B_0 seems a little unreasonable. Thus a relatively simple phenomenological theory can account for all the major variations of magnetic order in these metals. More fundamental theories using spin waves have been proposed by Yosida and Miwa, and Kaplan[8] although they are more difficult to evaluate in detail. The theory of ferromagnetic resonance derived from our general hamiltonian has been given by Cooper, Elliott, Nettel and Suhl. This and the details of the work described here will be published in detail in the Physical Review.

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8 See the corresponding articles in these proceedings.