

# A SELF-CONSISTENT METHOD IN THE STUDY OF THE MAGNETIC PROPERTIES OF RARE-EARTH INTERMETALLIC COMPOUNDS (\*)

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*ABSTRACT* — The magnetic properties of many Rare Earth compounds can be understood in terms of a crystal field and an exchange interaction. We give an account of the calculation of the direction of magnetization and crystalline electric field in  $\text{NdAl}_2$ . The exchange interaction in this compound is expressed by means of an isotropic molecular field proportional to the magnetization.

The magnetic properties of Rare-Earth metals, salts and alloys have been studied by many investigators, both experimentally and theoretically, in the last few years.

These works have been reviewed by Taylor [1] and by Buschow [2]. The magnetic properties of the Rare-Earths are due to the 4f electronic shell, shielded by the  $5s^2 5p^6$  conduction electrons. The spin-orbit coupling in the Rare Earth group is sufficiently strong to give rise to a total angular momentum  $J$  whose magnitude is not altered by the crystalline electric field.

The exchange interaction between the Rare-Earth spins is an indirect one via the conduction electrons. It is the competition between the crystalline electric field and the exchange interaction which gives rise to many interesting properties in the Rare-Earth metals, salts and alloys. In the metals, mainly in the heavy Rare-Earths, the exchange interaction is at least one order of magnitude stronger than the crystalline electric field, which is responsible for the complex orientations of the magnetic moments without altering their values; the magnetic moment at 0 K will have the value  $g\mu_B J/\text{ion}$ , where  $g$  is the Landé factor and  $\mu_B$  the Bohr

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magneton. In the salts, the crystalline electric field dominates over the exchange interaction; the crystalline electric field lifts the degeneracy of the ground state leaving a new ground state with lower degeneracy. The exchange interaction does not change, in a first order approximation, the new ground state.

In the Rare-Earth alloys the crystalline electric field and the exchange interaction are comparable. The exchange interaction mixes the different crystalline states, originating a new ground state whose magnetic moment is usually bigger than that of the crystalline ground state. Among Rare-Earth alloys many present a crystalline field with cubic symmetry. The magnetic properties of these alloys are not very difficult to analyse if we can assume an exchange interaction represented by a molecular field proportional to the magnetization.

We assume, very generally, that the magnetization may have one of the three possible directions  $[1, 0, 0]$ ,  $[1, 1, 0]$ ,  $[1, 1, 1]$  and therefore write the exchange hamiltonian as  $\mathcal{H}_{ex} = -g\mu_B \mathbf{H}_{ex} \cdot \mathbf{J}$ , where  $\mathbf{H}_{ex} = \lambda \mathbf{M}$ . We use this term to calculate the saturation moment at 0 K and therefore replace  $\mathbf{M}$  by its maximum value  $\mathbf{M}_0$ . The entire hamiltonian will have the form

$$\mathcal{H} = W \left( x \frac{O_4^0 + 5O_4^4}{F(4)} + (1 - |x|) \frac{O_6^0 - 21O_6^4}{F(6)} - c \sum_{k=1}^3 a_k J_k \right),$$

where

$$Wc a_k = g\mu_B \lambda M_{ok}$$

$$M_{ok} = Ng\mu_B \langle i | J_k | i \rangle,$$

$|i\rangle$  being the ground state of the Hamiltonian  $\mathcal{H}$ . In this expression  $O_4^0$ ,  $O_4^4$ ,  $O_6^0$  and  $O_6^4$  are angular momentum operators, and  $F(4)$  and  $F(6)$  are known coefficients [3]. The parameter  $x$  reflects the relative importance of the fourth and sixth order terms, while  $W$  is a scale factor which determines the magnitude of the crystal field splittings. Both  $x$  and  $W$  can be calculated from a point charge model, but reliable results are hardly expected. We have therefore treated both  $W$  and  $x$  as unknown quantities.

At 0 K the free energy  $F = U - TS$  is simply  $U$  and therefore the minimum of  $F$  is achieved for  $\mathbf{H}_{ex}$  parallel to  $\mathbf{J}$ , i.e. for  $\mathbf{M}_0$  parallel to  $\mathbf{J}$ . In the Hamiltonian,  $W$  can be obtained for each value of  $x$ , making use of the paramagnetic susceptibility. In the

molecular field model the paramagnetic susceptibility  $\chi$  is related to the susceptibility  $\chi_o$  which would be obtained if there was no exchange interaction, by the relations

$$M = \chi H_{\text{ext}} = \chi_o (H_{\text{ext}} + \lambda M)$$

which gives  $\chi^{-1} = \chi_o^{-1} - \lambda$ .

The inverse of the actual susceptibility  $\chi^{-1}$  (T) is therefore parallel to  $\chi_o^{-1}$  (T), since  $\lambda$  is assumed to be independent of temperature;  $\chi_o^{-1}$  can be computed for any choice of  $W$  and  $x$ , using only the crystal field hamiltonian. For each value of  $x$  a certain value of  $W$  results, which makes  $\chi^{-1}$  parallel to  $\chi_o^{-1}$ ; this same choice of  $W$  determines  $\lambda$ , since at  $T_c$ ,  $\chi^{-1} = 0$  and  $\lambda \chi_o = 1$ .

The magnetic moments for each of the directions  $[1, 0, 0]$ ,  $[1, 1, 0]$ ,  $[1, 1, 1]$  and for each value of  $x$  are only a function of  $c$ . We can therefore determine the value of  $c$  which gives the right magnetic moment at 0 K. Finally, using  $W$  and  $c$  we can calculate  $W \cdot c$ , which for the correct values of  $W$ ,  $x$  and the direction of the magnetic moment should be the same as  $Ng^2 \mu_B^2 \lambda \sqrt{\langle i | J_x | i \rangle^2 + \langle i | J_y | i \rangle^2 + \langle i | J_z | i \rangle^2}$ , where  $|i\rangle$  is the linear combination of the basis vectors  $|m\rangle$  which gives the right value of  $c$ . We have carried out the calculation as discussed above for the compound  $\text{NdAl}_2$ , for which  $J=9/2$ ,  $g=8/11$  and the crystal field has cubic symmetry [4]. We have found just one possible solution, where the direction of magnetization is along  $[1,0,0]$ , and the unperturbed crystalline ground state is a  $\Gamma_6$  doublet whose parameters are  $x \approx -0.9$ ,  $W \approx 6$  K. The molecular field coefficient determined in this way was  $Ng^2 \mu_B^2 \lambda \approx 9$  K.

These compare quite well (see table) with the values  $W \approx 6$  K,  $Ng^2 \mu_B^2 \lambda \approx 9$  K,  $x \approx -0.6$  quoted by Houmann et al [5] and used in the interpretation of their inelastic neutron experiments.

TABLE

	Unperturbed crystalline ground state	Direction of easy magnetization	Molecular parameter $Ng^2 \mu_B^2 \lambda$	Crystal Field Parameters	
				$W$	$x$
Experimental	$\Gamma_6$	$[1,0,0]$	$\approx 9$ K	$\approx 6$ K	$\approx -0.6$
Theoretical	$\Gamma_6$	$[1,0,0]$	$\approx 9$ K	$\approx 6$ K	$\approx -0.9$

We have therefore shown that a self-consistent method can be used in the interpretation of the magnetic properties of Rare-Earth alloys; the crystalline electric field and exchange interaction were treated on equal terms and the analyses were carried out for the three directions  $[1,0,0]$ ,  $[1,1,0]$  and  $[1,1,1]$ , one of which is the direction of easy magnetization.

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