

MAGNETIC PHASE TRANSITIONS IN NEODYMIUM IN A PHENOMENOLOGICAL APPROXIMATION. PART II

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The fact of two distinct temperatures of the magnetic phase transitions, occurring separately in the two crystallographic sublattices of neodymium, is interpreted using the model of a single Nd^{+3} ion placed in a metallic and magnetic matrix with electrostatic field point symmetries D_{3d} and D_{3h} .

We take into consideration the interaction of the Nd^{+3} ion with the crystal-electric field and its indirect exchange interaction with surrounding ions regarded in the MWMF (modified Weiss molecular field) approximation. The necessary condition for obtaining two distinct values of the magnetic phase transition temperature is the decoupling of the set of two self-consistent equations for the LRO (long-range order) parameters into two mutually independent equations. This decoupling is feasible since, as shown here, the two contributions to the molecular field from the other sublattice cancel out mutually.

The second condition necessary for the occurrence of two distinct transition temperatures is the differentiation of the molecular fields at corresponding sites belonging to the two sublattices. This difference is due to:

- 1) the different densities of packing of neighbours of the cubic and hexagonal sites;
- 2) the long-range nature of the indirect exchange integral (*via* the conduction electrons) which enables the more distant neighbours to play an important part;
- 3) the type of magnetic structure, exhibiting a different translational periodicity of the magnetic configuration in each sublattice;
- 4) the direct influence of the crystal field as revealed in the factors $\frac{1}{2}\beta_A^2$ and $\frac{1}{2}\beta_B^2$ in the final expressions for the magnetic phase transition temperatures.

1. Introduction

We are concerned with magnetic phase transitions of the type: order — disorder, depending on the spontaneous emergence (with decreasing temperature) of magnetic long-range order (LRO) accompanied by breakdown of symmetry.

It is the main purpose of this paper to explain semi-quantitatively the reasons for

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a peculiarity exhibited by the magnetic phase transitions of neodymium [3-5] consisting in a remarkable differentiation of the temperatures of the magnetic phase transition in each of the two crystal sublattices of the dhcp structure of neodymium. The difference amounts to 11.7°K (the magnetic phase transition in the hexagonal sublattice occurs at 19.2°K, and that of the cubic sublattice at 7.5°K [3-5]).

In the case of a phase transition of the second order, the LRO parameter decreases continuously with increasing temperature and vanishes at the transition temperature. At temperatures above the transition point, the LRO parameter vanishes ([2] and Refs therein).

For neodymium, we shall define two LRO parameters in the form of thermal averages of the total momentum operators $\langle \vec{J}_A \rangle_T$ and $\langle \vec{J}_B \rangle_T$ for the two sublattices, respectively. Long-range ordering in this system arises due to the presence of an LRO internal field \vec{H} , which substitutes the indirect exchange interaction of each particular ion with other ions of its environment [1], [2]. In our considerations, it is the modified Weiss molecular field [1], [6], [7].

The LRO parameters $\langle \vec{J}_A \rangle_T$ and $\langle \vec{J}_B \rangle_T$ are related to the internal fields $\vec{H}_{ex}^{(A)}$ and $\vec{H}_{ex}^{(B)}$ by the self-consistent equations:

$$\begin{aligned}\langle \vec{J}_A \rangle_T &= f_{F,T}^{(1)}(\vec{H}_{ex}^{(A)}(\langle \vec{J}_A \rangle_T, \langle \vec{J}_B \rangle_T)), \\ \langle \vec{J}_B \rangle_T &= f_{F,T}^{(2)}(\vec{H}_{ex}^{(B)}(\langle \vec{J}_A \rangle_T, \langle \vec{J}_B \rangle_T)).\end{aligned}\quad (1.1)$$

The indices F and T stand for the indirect exchange interaction strength and the temperature, respectively; their presence indicates that the LRO parameters depend in general on inter-ion exchange couplings and temperature.

To determine the magnetic phase transition temperatures in the MWMF approximation, we make use of Eq. (1.1). In the light of our previous considerations, the magnetic phase transition temperature is the temperature below which there exists at least one non-zero solution of Eq. (1.1) for $\langle \vec{J}_A \rangle_T$ or $\langle \vec{J}_B \rangle_T$.

In the course of our further discussion, we shall determine an explicit form of Eq. (1.1) for neodymium, in order to derive the magnetic phase transition temperatures for the two sublattices.

2. Determination of the LRO parameters for neodymium

Due to the relation of proportionality between the total angular momentum operator \hat{J} and the magnetic momentum operator $\hat{\mu}$ which holds for rare earth ions, any space ordering of thermal averages of the total angular momentum operators attached to lattice sites entails simultaneously a magnetic ordering of the crystal.

We proceed now to obtain the canonical thermal averages of the observables \hat{J}^x , \hat{J}^y and \hat{J}^z at both types of crystal site of neodymium metal. We resort to our previous results concerning the energy levels of the f^3 electron configuration in the crystal-electric and molecular fields (see [7]).

1) Nd^{+3} ion at the cubic sites ([7, 8])

The energy values as well as the wave functions $|\psi_1^{(c)}\rangle$ and $|\psi_2^{(c)}\rangle$ of the ground Kramers doublet states in the molecular field were derived in [7]. The quantum-mechanical averages of \hat{J}_c^x , \hat{J}_c^y and \hat{J}_c^z in these states are equal to:

$$\begin{aligned}\langle \psi_1^{(c)} | \hat{J}_c^x | \psi_1^{(c)} \rangle &= -\frac{1}{2} |\beta_c| \cos \varphi_c, \\ \langle \psi_1^{(c)} | \hat{J}_c^y | \psi_1^{(c)} \rangle &= -\frac{1}{2} |\beta_c| \sin \varphi_c, \\ \langle \psi_1^{(c)} | \hat{J}_c^z | \psi_1^{(c)} \rangle &= 0;\end{aligned}\quad (2.1a)$$

$$\begin{aligned}\langle \psi_2^{(c)} | \hat{J}_c^x | \psi_2^{(c)} \rangle &= \frac{1}{2} |\beta_c| \cos \varphi_c, \\ \langle \psi_2^{(c)} | \hat{J}_c^y | \psi_2^{(c)} \rangle &= \frac{1}{2} |\beta_c| \sin \varphi_c, \\ \langle \psi_2^{(c)} | \hat{J}_c^z | \psi_2^{(c)} \rangle &= 0,\end{aligned}\quad (2.1b)$$

where "c" stands for the cubic position, φ_c is the spherical angle between the hexagonal axis \vec{a}_1 in the basal plane and the projection of the molecular field $\vec{H}_{ex}^{(c)}$ on this plane

$$\beta_c = \frac{1}{2} c_1^{(\lambda)^2} (J + M_1) + c_2^{(\lambda)} c_3^{(\lambda)} \sqrt{(J + M_2)(J + M_3)} \quad [7],$$

$$J = 9/2,$$

M_1 , M_2 and M_3 are the magnetic quantum numbers of the ${}^4I_{9/2}$ term; for the ground state Kramers doublet, within the point charge model of the crystal-electric field, they are equal to: $M_1 = \frac{1}{2}$, $M_2 = -\frac{5}{2}$, $M_3 = \frac{7}{2}$;

$c_1^{(\lambda)}$, $c_2^{(\lambda)}$, $c_3^{(\lambda)}$ are the wave coefficients of the doublet (see [8]).

Resorting to the well-known formula for the canonical thermal average of an observable, we obtain, on restricting our considerations to the ground Kramers doublet:

$$\begin{aligned}\langle \hat{J}_c^x(\vec{R}_n) \rangle_T &= -\frac{1}{2} |\beta_c| \text{th} \frac{\omega^{(c)}(\vec{R}_n)}{kT} \cos \varphi_c, \\ \langle \hat{J}_c^y(\vec{R}_n) \rangle_T &= -\frac{1}{2} |\beta_c| \text{th} \frac{\omega^{(c)}(\vec{R}_n)}{kT} \sin \varphi_c,\end{aligned}\quad (2.2a)$$

where \vec{R}_n are position vectors of sites belonging to the cubic sublattices A and A' with respect to sites chosen as the centres of these sublattices;

$$\omega^{(c)} = g\mu_B |\beta_c| H_{ex}^{(c)}(\vec{R}_n) \quad (\text{see [7]}).$$

2) Nd^{+3} ions at the hexagonal sites

The energy values and wave functions of the two lowest levels in the molecular field are given in [7].

Similarly, as in the case of the cubic site, we have:

$$\begin{aligned}\langle \psi_1^{(h)} | \hat{J}_h^x | \psi_1^{(h)} \rangle &= \frac{1}{2} |\beta_h| \cos \varphi_h, \\ \langle \psi_1^{(h)} | \hat{J}_h^y | \psi_1^{(h)} \rangle &= \frac{1}{2} |\beta_h| \sin \varphi_h, \\ \langle \psi_1^{(h)} | \hat{J}_h^z | \psi_1^{(h)} \rangle &= 0;\end{aligned}\quad (2.3a)$$

$$\begin{aligned}
\langle \psi_2^{(h)} | \hat{J}_h^x | \psi_2^{(h)} \rangle &= -\frac{1}{2} |\beta_h| \cos \varphi_h, \\
\langle \psi_2^{(h)} | \hat{J}_h^y | \psi_2^{(h)} \rangle &= -\frac{1}{2} |\beta_h| \sin \varphi_h, \\
\langle \psi_2^{(h)} | \hat{J}_h^z | \psi_2^{(h)} \rangle &= 0,
\end{aligned}
\tag{2.3b}$$

where “ h ” stands for the hexagonal position, φ_h is the spherical angle between the projection of the vector $H_{ex}^{(h)}$ on the basal plane and the \vec{a}_1 -axis,

$$\beta_h = c_1^{(\lambda)} c_2^{(\lambda)} \sqrt{(J + M_1)(J + M_2)}, \quad (\text{see [7]})$$

$$J = 9/2,$$

$M_1 = -\frac{5}{2}$, $M_3 = \frac{7}{2}$ within the point charge model of the crystal-electric field,

$c_1^{(\lambda)}$, $c_2^{(\lambda)}$ are the wave coefficients of the ground Kramers doublet for D_{3h} symmetry of the crystal field [8].

The quantum-mechanical averages of \hat{J}_h^z vanish since the molecular field $\vec{H}_{ex}^{(h)}$ takes the direction perpendicular to the \vec{c} -axis [7].

The canonical thermal averages of the observables \hat{J}_h^x and \hat{J}_h^y are written as follows:

$$\langle \hat{J}_h^x(\vec{R}_n) \rangle_T = \frac{1}{2} |\beta_h| \text{th} \frac{\omega^{(h)}(\vec{R}_n)}{kT} \cos \varphi_h, \tag{2.4a}$$

$$\langle \hat{J}_h^y(\vec{R}_n) \rangle_T = \frac{1}{2} |\beta_h| \text{th} \frac{\omega^{(h)}(\vec{R}_n)}{kT} \sin \varphi_h, \tag{2.4b}$$

where \vec{R}_n are position vectors of hexagonal sites with respect to the centres of the sublattices B and C , respectively.

Let us now introduce the following notation:

$$|\langle \vec{J}_c(\vec{R}_n) \rangle_T| = \frac{1}{2} |\beta_c| \text{th} \frac{\omega^{(c)}(\vec{R}_n)}{kT}, \tag{2.5a}$$

$$|\langle \vec{J}_h(\vec{R}_n) \rangle_T| = \frac{1}{2} |\beta_h| \text{th} \frac{\omega^{(h)}(\vec{R}_n)}{kT}. \tag{2.5b}$$

As seen, the thermal averages of the total angular momentum components (see: (2.2) and (2.4)) were derived in the single-ion approximation. However, space correlation of the moments was taken into account by way of the molecular field, which varies from one site to another.

3. Explicit forms of the relations: $|\langle \vec{J}_A \rangle_T| = f_{FT}^{(1)}(\vec{H}_{ex}^{(A)})$, and $|\langle \vec{J}_B \rangle_T| = f_{FT}^{(2)}(\vec{H}_{ex}^{(B)})$

At this point of our discussion, we resort to available experimental information on the magnetic structure of neodymium [3–5] on the basis of which the following of translational dependence of the averaged moments can be assumed for the distinct sublattices of neodymium.

1) Cubic sublattices (A and A'):

$$\langle \hat{J}(\vec{R}_{A_n}) \rangle_T = \langle \hat{J}_{A_1} \rangle_T \cos(\vec{k}_c, \vec{R}_{A_1 A_n}), \quad (3.1a)$$

$$\langle \hat{J}(\vec{R}_{A'_n}) \rangle_T = \langle \hat{J}_{A'_1} \rangle_T \cos((\vec{k}_c, \vec{R}_{A'_1 A'_n}) + \gamma_{A'_1}), \quad (3.1b)$$

where $\langle \hat{J}_{A_1} \rangle_T$ and $\langle \hat{J}_{A'_1} \rangle_T$ stand for the thermal averages of the total angular momentum operators attached to the sites: A_1 and A' with largest moments of magnetic structure in the respective cubic sublattice;

$\langle \hat{J}(\vec{R}_{A_n}) \rangle_T$, $\langle \hat{J}(\vec{R}_{A'_n}) \rangle_T$ are thermal averages of the \hat{J} operators, localized in the cubic sites A_n , A'_n ;

\vec{R}_{A_n} , $\vec{R}_{A'_n}$ are position vectors of cubic sites belonging to the sublattices A and A' with respect to the central sites A_1 and A'_1 , respectively;

$\vec{k}_c = \mu_2 \vec{b}_1$ is the wave vector of the cubic sublattice [3], [4] (\vec{b}_1 — the reciprocal lattice vector);

$\gamma_{A'_1}$ is the difference in phase between the sinusoidally modulated configuration of the sublattice A and the sinusoidally modulated configuration of the sublattice A' (the difference is taken between site A_1 and one of its nearest neighbours in sublattice A').

2) Hexagonal sublattices (B and C)

$$\langle \hat{J}(\vec{R}_{B_n}) \rangle_T = \langle \hat{J}_B \rangle_T \cos((\vec{k}_h, \vec{R}_{B_1 B_n}) + \gamma_{B_1}), \quad (3.2a)$$

$$\langle \hat{J}(\vec{R}_{C_n}) \rangle_T = \langle \hat{J}_C \rangle_T \cos((\vec{k}_h, \vec{R}_{C_1 C_n}) + \gamma_{C_1}), \quad (3.2b)$$

where B and C denote the sites with largest moments of periodic magnetic structures in the respective hexagonal sublattice;

$\vec{k}_h = \mu_1 \vec{b}_1$ is the wave vector of the hexagonal sublattices [3], [4];

γ_{B_1} , γ_{C_1} are differences in phase between the sinusoidally modulated magnetic structure in sublattice A and the sinusoidally modulated magnetic structures of the sublattices B and C , respectively (the phase differences are taken between site A and its nearest neighbours chosen in sublattices B and C , *i. e.* the sites B_1 and C_1).

As seen from Eqs (3.1) and (3.2), the temperature dependence of the LRO parameters is determined by their "space amplitudes" *i. e.* the quantities $\langle \hat{J}_{A_1} \rangle_T$, $\langle \hat{J}_{A'_1} \rangle_T$, $\langle \hat{J}_B \rangle_T$, $\langle \hat{J}_C \rangle_T$. Since we are mainly interested in the temperature dependence of the LRO parameters, we shall restrict our discussion to these amplitudes only. The reason for this simplification is inherent in our problem, which is aimed at the determination of the temperatures at which the LRO parameters become equal to zero.

From Eqs (2.5a) and (2.5b), where the index "c" stands for A_1 and A'_1 , and "h" for B_1 and C_1 , and on resorting to the formulas for the exchange energies $\omega^{(c)}$ and $\omega^{(h)}$ expressed as functions of the molecular fields at the appropriate sites, we obtain finally the following explicit forms of the relations:

$$|\langle \hat{J}_A \rangle_T| = f_{FT}^{(1)}(\vec{H}_{ex}^{(A)}), \quad |\langle \hat{J}_B \rangle_T| = f_{FT}^{(2)}(\vec{H}_{ex}^{(B)}).$$

1) Cubic sites:

$$|\langle \hat{J}_{A_1} \rangle_T| = \frac{1}{2} |\beta_A| \operatorname{th} \frac{g\mu_B |\beta_A| H_{ex}(\vec{R}_{A_1})}{kT}, \quad (3.3a)$$

$$|\langle \hat{J}_{A'_1} \rangle_T| = \frac{1}{2} |\beta_A| \operatorname{th} \frac{g\mu_B |\beta_A| H_{ex}(\vec{R}_{A'_1})}{kT}, \quad (3.3b)$$

where the factor $|\beta_A|$ is the same for all cubic sites.

2) Hexagonal sites:

$$|\langle \hat{J}_{B_1} \rangle_T| = \frac{1}{2} |\beta_B| \operatorname{th} \frac{g\mu_B |\beta_B| H_{ex}(\vec{R}_{B_1})}{kT}, \quad (3.4a)$$

$$|\langle \hat{J}_{C_1} \rangle_T| = \frac{1}{2} |\beta_B| \operatorname{th} \frac{g\mu_B |\beta_B| H_{ex}(\vec{R}_{C_1})}{kT}, \quad (3.4b)$$

where the factor $|\beta_B|$ is the same for all hexagonal sites.

4. Explicit forms of the relations:

$$H_{ex}^{(A)} = H_{ex}^{(A)}(\langle \hat{J}_A \rangle_T, \langle \hat{J}_B \rangle_T) \text{ and } H_{ex}^{(B)} = H_{ex}^{(B)}(\langle \hat{J}_A \rangle_T, \langle \hat{J}_B \rangle_T)$$

The molecular field at a particular site is a linear combination of the thermal averages of the total angular momentum operators attached to the neighbouring sites. The coefficients of the linear combination are directly proportional to the indirect exchange integrals for the appropriate distances. The register of neighbours for the sites of the two types in dhcp structure indicates that cubic and hexagonal sites differ in the density of packing of their neighbours. This difference becomes apparent already at the distance c , at which the cubic site has two neighbours belonging to the same sublattice, whereas the hexagonal site has no neighbours at all at this distance.

With regard to the preceding and other information on the neighbourhood of the sites, we can write the formulas for the molecular fields at the sites: A_1 , A'_1 , B_1 and C_1 , limiting our considerations to the distance c .

1) For the cubic sites we have:

$$\begin{aligned} \vec{H}_{ex}(\vec{R}_{A_1}) = & -\frac{1}{g\mu_B} F_{A_1}^A \langle \hat{J}_{A_1} \rangle_T - \frac{1}{g\mu_B} F_{A_1}^B \langle \hat{J}_B \rangle_T - \\ & - \frac{1}{g\mu_B} F_{A_1}^C \langle \hat{J}_C \rangle_T - \frac{1}{g\mu_B} F_{A_1}^{A'} \langle \hat{J}_{A'} \rangle_T, \end{aligned} \quad (4.1)$$

$$\begin{aligned} \vec{H}_{ex}(\vec{R}_{A'_1}) = & -\frac{1}{g\mu_B} F_{A'_1}^A \langle \hat{J}_{A'} \rangle_T - \frac{1}{g\mu_B} F_{A'_1}^B \langle \hat{J}_B \rangle_T - \\ & - \frac{1}{g\mu_B} F_{A'_1}^C \langle \hat{J}_C \rangle_T - \frac{1}{g\mu_B} F_{A'_1}^A \langle \hat{J}_{A_1} \rangle_T, \end{aligned} \quad (4.2)$$

where

$$\begin{aligned}
 F_{A_1}^A &= F_1 \sum_n^{(1)} \cos(\vec{k}_c, \vec{R}_{A_1 A_n}), \\
 F_{A_1}^B &= F_1 \sum_n^{(1)} \cos((\vec{k}_h, \vec{R}_{A_1 B_n} - \vec{R}_{A_1 B_1}) + \gamma_{B_1}) + F_2 \sum_n^{(2)} \cos((\vec{k}_h, \vec{R}_{A_1 B_n} - \vec{R}_{A_1 B_1}) + \gamma_{B_1}), \\
 F_{A_1}^C &= F_1 \sum_n^{(1)} \cos((\vec{k}_h, \vec{R}_{A_1 C_n} - \vec{R}_{A_1 C_1}) + \gamma_{C_1}) + F_2 \sum_n^{(2)} \cos((\vec{k}_h, \vec{R}_{A_1 C_n} - \vec{R}_{A_1 C_1}) + \gamma_{C_1}), \\
 F_{A_1}^{A'} &= F_3 \sum_n^{(3)} \cos((\vec{k}_c, \vec{R}_{A_1 A'_n} - \vec{R}_{A_1 A'_1}) + \gamma_{A'}); \\
 F_{A_1}^{A'} &= F_1 \sum_n^{(1)} \cos((\vec{k}_c, \vec{R}_{A_1 A'_n}) + \gamma_{A'}),
 \end{aligned} \tag{4.1a}$$

$$\begin{aligned}
 F_{A'_1}^B &= F_1 \sum_n^{(1)} \cos((\vec{k}_h, \vec{R}_{A'_1 B_n} - \vec{R}_{A'_1 B_1}) + \gamma_{B_1}) + F_2 \sum_n^{(2)} \cos((\vec{k}_h, \vec{R}_{A'_1 B_n} - \vec{R}_{A'_1 B_1}) + \gamma_{B_1}), \\
 F_{A'_1}^C &= F_1 \sum_n^{(1)} \cos((\vec{k}_h, \vec{R}_{A'_1 C_n} - \vec{R}_{A'_1 C_1}) + \gamma_{C_1}) + F_2 \sum_n^{(2)} \cos((\vec{k}_h, \vec{R}_{A'_1 C_n} - \vec{R}_{A'_1 C_1}) + \gamma_{C_1}), \\
 F_{A'_1}^A &= F_3 \sum_n^{(3)} \cos(\vec{k}_c, \vec{R}_{A'_1 A_n} - \vec{R}_{A'_1 A_1}),
 \end{aligned} \tag{4.2a}$$

where F_1, F_2, F_3 are indirect exchange integrals at the distances: $a, a\sqrt{2}$ and $c \cong 1.63 a$, respectively; $\Sigma^{(i)}$ denotes the summation over the coordination sphere "i".

2) For the hexagonal sites we have:

$$\vec{H}_{ex}(\vec{R}_{B_1}) = -\frac{1}{g\mu_B} F_{B_1}^B \langle \hat{J}_B \rangle_T - \frac{1}{g\mu_B} F_{B_1}^A \langle \hat{J}_{A_1} \rangle_T - \frac{1}{g\mu_B} F_{B_1}^{A'} \langle \hat{J}_{A'} \rangle_T \tag{4.3}$$

and

$$\vec{H}_{ex}(\vec{R}_{C_1}) = -\frac{1}{g\mu_B} F_{C_1}^C \langle \hat{J}_C \rangle_T - \frac{1}{g\mu_B} F_{C_1}^A \langle \hat{J}_{A_1} \rangle_T - \frac{1}{g\mu_B} F_{C_1}^{A'} \langle \hat{J}_{A'} \rangle_T, \tag{4.4}$$

where

$$\begin{aligned}
 F_{B_1}^B &= F_1 \sum_n^{(1)} \cos((\vec{k}_h, \vec{R}_{B_1 B_n}) + \gamma_{B_1}), \\
 F_{B_1}^A &= F_1 \sum_n^{(1)} \cos(\vec{k}_c, \vec{R}_{B_1 A_n} - \vec{R}_{B_1 A_1}) + F_2 \sum_n^{(2)} \cos(\vec{k}_c, \vec{R}_{B_1 A_n} - \vec{R}_{B_1 A_1}), \\
 F_{B_1}^{A'} &= F_1 \sum_n^{(1)} \cos((\vec{k}_c, \vec{R}_{B_1 A'_n} - \vec{R}_{B_1 A'_1}) + \gamma_{A'}) + F_2 \sum_n^{(2)} \cos((\vec{k}_c, \vec{R}_{B_1 A'_n} - \vec{R}_{B_1 A'_1}) + \gamma_{A'}); \\
 F_{C_1}^C &= F_1 \sum_n^{(1)} \cos((\vec{k}_h, \vec{R}_{C_1 C_n}) + \gamma_{C_1}),
 \end{aligned} \tag{4.3a}$$

$$F_{C_1}^A = F_1 \sum_n^{(1)} \cos(\vec{k}_c, \vec{R}_{C_1 A_n} - \vec{R}_{C_1 A_1}) + F_2 \sum_n^{(2)} \cos(\vec{k}_c, \vec{R}_{C_1 A_n} - \vec{R}_{C_1 A_1}),$$

$$F_{C_1}^{A'} = F_1 \sum_n^{(1)} \cos((\vec{k}_c, \vec{R}_{C_1 A'_n} - \vec{R}_{C_1 A'_1}) + \gamma_{A'_1}) + F_2 \sum_n^{(2)} \cos((\vec{k}_c, \vec{R}_{C_1 A'_n} - \vec{R}_{C_1 A'_1}) + \gamma_{A'_1}),$$
(4.4a)

where F_1, F_2 are indirect exchange integrals at the distances a and $a\sqrt{2}$, respectively; $\Sigma^{(i)}$ stands for the summation over the coordination sphere “ i ”.

The vector equations (4.1)–(4.4) may be rewritten in Cartesian components on insertion of the averaged components of the total angular momentum operators from Eqs (2.2a), (2.2b), (2.4a), (2.4b) using the notation (2.5a) and (2.5b). It should be kept in mind, moreover, that at any particular site, the magnetic moments takes the direction of the molecular field \vec{H}_{ex} [6].

Let us revert to the experimental data on the magnetic structure in neodymium [3–5]. It is easy to notice that for the magnetic ordering exhibited by neodymium, the following relations are fulfilled:

$$|\langle \hat{J}_{A_1} \rangle_T| = |\langle \hat{J}_{A'_1} \rangle_T|, \quad \varphi_A = \varphi_{A'}, \quad \gamma_{A'_1} = \pi;$$

$$|\langle \hat{J}_B \rangle_T| = |\langle \hat{J}_C \rangle_T|, \quad \varphi_B = \varphi_C, \quad \gamma_{C_1} = \gamma_{B_1} + \pi.$$
(4.5)

Taking into consideration the environments of the particular sites, we can write due to Eq. (4.5):

$$F_{A'_1}^A = -F_{A_1}^A, \quad F_{A_1}^A = -F_{A'_1}^A, \quad F_{A_1}^C = -F_{A_1}^B, \quad F_{A'_1}^C = -F_{A'_1}^B;$$

$$F_{C_1}^C = -F_{B_1}^B, \quad F_{B_1}^A = -F_{B_1}^A, \quad F_{C_1}^A = -F_{C_1}^A.$$
(4.6)

Finally, we come to the following formulas for the components of the molecular field \vec{H}_{ex} :

1) Cubic sites

$$H_{ex}^x(\vec{R}_{A_1}) = -\frac{1}{g\mu_B} |\langle \vec{J}_{A_1} \rangle_T| (F_{A_1}^A + F_{A'_1}^A) \cos \varphi_A,$$

$$H_{ex}^y(\vec{R}_{A_1}) = -\frac{1}{g\mu_B} |\langle \vec{J}_{A_1} \rangle_T| (F_{A_1}^A + F_{A'_1}^A) \sin \varphi_A,$$

$$H_{ex}^z(\vec{R}_{A_1}) = 0$$
(4.7a)

and

$$H_{ex}^x(\vec{R}_{A'_1}) = -H_{ex}^x(\vec{R}_{A_1}),$$

$$H_{ex}^y(\vec{R}_{A'_1}) = -H_{ex}^y(\vec{R}_{A_1}),$$

$$H_{ex}^z(\vec{R}_{A'_1}) = 0.$$
(4.7b)

2) Hexagonal sites

$$\begin{aligned}
 H_{ex}^x(\vec{R}_{B_1}) &= -\frac{1}{g\mu_B} |\langle \hat{J}_B \rangle_T| F_{B_1}^B \cos \varphi_B, \\
 H_{ex}^y(\vec{R}_{B_1}) &= -\frac{1}{g\mu_B} |\langle \hat{J}_B \rangle_T| F_{B_1}^B \sin \varphi_B, \\
 H_{ex}^z(\vec{R}_{B_1}) &= 0
 \end{aligned} \tag{4.8a}$$

and

$$\begin{aligned}
 H_{ex}^x(\vec{R}_{C_1}) &= -H_{ex}^x(\vec{R}_{B_1}), \\
 H_{ex}^y(\vec{R}_{C_1}) &= -H_{ex}^y(\vec{R}_{B_1}), \\
 H_{ex}^z(\vec{R}_{C_1}) &= 0.
 \end{aligned} \tag{4.8b}$$

Since the molecular field components at the sites A'_1 and C_1 differ in sign only from those at the sites A_1 and B_1 , respectively, in our further consideration we shall discuss two sites only: one cubic (A_1) and one hexagonal site (B_1). The strengths of the molecular field at the sites A_1 and B_1 are equal to:

$$H_{ex}(\vec{R}_{A_1}) = \frac{1}{g\mu_B} |\langle \hat{J}_{A_1} \rangle_T| (F_{A_1}^A + F_{A_1}^{A'}), \tag{4.9a}$$

$$H_{ex}(\vec{R}_{B_1}) = \frac{1}{g\mu_B} |\langle \hat{J}_B \rangle_T| F_{B_1}^B. \tag{4.9b}$$

Eqs (4.9a) and (4.9b) are explicit forms of the relations between the LRO field and the space amplitudes of the LRO parameters. The temperature-dependence of the LRO parameter is determined by its space amplitude and, therefore, from our point of view, it suffices to examine the temperatures at which the space amplitudes of the LRO parameter vanish.

5. Determination of the magnetic phase transition temperatures

Combining Eqs (3.3a), (3.4a) with Eqs (4.9a) and (4.9b), respectively, we come finally to the explicit forms of the self-consistent equations for the LRO parameters or, strictly speaking, the space amplitudes of the LRO parameters, as follows:

$$|\langle \hat{J}_{A_1} \rangle_T| = \frac{1}{2} |\beta_A| \operatorname{th} \left(\frac{\frac{1}{2} |\beta_A| (F_{A_1}^A + F_{A_1}^{A'})}{kT} |\langle \hat{J}_{A_1} \rangle_T| \right) \tag{5.1}$$

and

$$|\langle \hat{J}_{B_1} \rangle_T| = \frac{1}{2} |\beta_B| \operatorname{th} \left(\frac{\frac{1}{2} |\beta_B| F_{B_1}^B}{kT} |\langle \hat{J}_{B_1} \rangle_T| \right). \tag{5.2}$$

On performing summations of the cosines over the appropriate sites in $F_{A_1}^A$, $F_{A_1}^{A'}$ and $F_{B_1}^B$, we obtain:

$$\begin{aligned}
 F_{A_1}^A &= 2F_1 \left(\cos \frac{4\pi}{\sqrt{3}} \mu_2 + 2 \cos \frac{2\pi}{\sqrt{3}} \mu_2 \right), \\
 F_{A_1}^{A'} &= -2F_3, \\
 F_{B_1}^B &= 2F_1 \cos \gamma_{B_1} \left(\cos \frac{4\pi}{\sqrt{3}} \mu_1 + 2 \cos \frac{2\pi}{\sqrt{3}} \mu_1 \right). \quad (5.3)
 \end{aligned}$$

Inserting Eqs (5.3) into Eqs (5.1) and (5.2), and on making use of the relations:

$$|\langle \hat{J}_{A_1} \rangle_T| = |\langle \hat{J}_A \rangle_T| \quad \text{and} \quad |\langle \hat{J}_{B_1} \rangle_T| = |\langle \hat{J}_B \rangle_T| \cos \gamma_{B_1}$$

(see: Eqs (3.1) and (3.2)), we come finally to the following relations:

$$|\langle \hat{J}_{A_1} \rangle_T| = \frac{1}{2} |\beta_A| \operatorname{th} \left(\frac{|\beta_A| \left[F_1 \left(\cos \frac{4\pi}{\sqrt{3}} \mu_2 + 2 \cos \frac{2\pi}{\sqrt{3}} \mu_2 \right) - F_3 \right]}{kT} |\langle \hat{J}_{A_1} \rangle_T| \right), \quad (5.4)$$

$$|\langle \hat{J}_{B_1} \rangle_T| = \frac{1}{2} |\beta_B| \operatorname{th} \left(\frac{|\beta_B| F_1 \left(\cos \frac{4\pi}{\sqrt{3}} \mu_1 + 2 \cos \frac{2\pi}{\sqrt{3}} \mu_1 \right)}{kT} |\langle \hat{J}_{B_1} \rangle_T| \right). \quad (5.5)$$

As seen, the system of self-consistent, mutually conjugate equations for the LRO parameters decouples into two independent equations, since within the framework of the approximation adopted by us, the exchange field at a site under consideration depends only on the LRO parameter of the sublattice to which the site belongs. The magnetic crystal is thus decoupled into two magnetic subsystems, corresponding to the crystallographic sublattices. It should moreover be noted that, on limiting ones considerations to the distance c , the sites B and C are not neighbours yet. The mutual independence of the planes B and C does not play, however, any part at all in the differentiation of the magnetic phase transition temperatures. The essential role belongs to the mutual compensation of the contributions from the other sublattice to the exchange field at any site.

On the basis of Eqs (5.4) and (5.5), we can determine those temperatures at which the space amplitudes of the LRO parameters *i.e.* the quantities $|\langle \hat{J}_{A_1} \rangle_T|$ and $|\langle \hat{J}_{B_1} \rangle_T|$ become equal to zero. In other words, at those temperatures, the space ordering of the magnetic moments spontaneously disappears *i.e.* a phase transition takes place.

The determination of the magnetic phase transition temperatures is performed according to a very well-known procedure (see: [6]).

We obtain the following results:

$$kT_{NA} = \frac{1}{2} \beta_A^2 \left[F_1 \left(\cos \frac{4\pi}{\sqrt{3}} \mu_2 + 2 \cos \frac{2\pi}{\sqrt{3}} \mu_2 \right) - F_3 \right], \quad (5.6)$$

$$kT_{NB} = \frac{1}{2} \beta_B^2 F_1 \left(\cos \frac{4\pi}{\sqrt{3}} \mu_1 + 2 \cos \frac{2\pi}{\sqrt{3}} \mu_1 \right). \quad (5.7)$$

As seen clearly from the above equations, the difference between the two magnetic phase transition temperatures of the crystallographic sublattices of neodymium is determined by the unequivalence of the crystal positions in the dhcp structure. The different packing densities of the neighbours of the cubic and the hexagonal sites give rise to the indirect exchange integral F_3 in Eq. (5.4).

The quantities μ_1 and μ_2 , which also contribute to the difference in magnetic phase transition temperatures, are directly related to the magnetic structure of neodymium (see Eqs (3.1), (3.2)).

6. Final remarks

Our conclusions are summarized as follows:

We have specified the factors leading to two different magnetic phase transition temperatures for the two sublattices of neodymium. The necessary condition for this differentiation is the splitting of the set of the self-consistent equations for the LRO parameters into two mutually independent equations. The splitting is determined by the type of magnetic structure which implies at each site the mutual cancelling out of the contributions to the molecular field from the sublattice to which the considered site does not belong. Owing to this compensation, the magnetic crystal behaves like a system consisting of two magnetic mutually decoupled subsystems, corresponding to the crystallographic cubic and hexagonal sublattices, which can thus have different temperatures of magnetic phase transition. The chief role herein is played by the single-ion magnetic anisotropy responsible for the "plane" character of the magnetic ordering (with no z -component). The single-ion anisotropy arises here due to the crystal-electric field.

The second necessary condition for the differentiation of the magnetic phase transition temperatures is the occurrence of different molecular fields at the cubic and hexagonal sites. This difference is due to the following reasons: the different density of packing of the neighbours of a cubic and a hexagonal site; the long-range nature of the indirect exchange integral which causes that the more distant neighbours still have to be taken into account; and, finally, the quantities μ_1 and μ_2 (see: (5.5) and (5.6)). The latter are directly related with the magnetic ordering. The quantities μ_1 and μ_2 determine the translational dependence of the magnetic ordering within the cubic and the hexagonal sublattice, respectively.

The direct influence of the crystal-electric field is expressed by the factors $\frac{1}{2} \beta_B^2$ and $\frac{1}{2} \beta_A^2$ for cubic and hexagonal sites, respectively.

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