DETERMINATION OF MAGNETIC STRUCTURES IN hcp CRYSTALS. PART I

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The Landau and Lifshitz theory is applied to a single second-order magnetic phase transition from the paramagnetic to the magnetic state within the framework of the model of magnetic moments strictly attached to lattice points for the hcp crystal lattice. Knowing the magnetic space group in the disordered phase only, all magnetic orderings possible just below the transition temperature are found.

I. Introduction

It is the aim of the present paper to derive the magnetic orderings resulting from a single paramagnetic — magnetic second-order phase transition immediately below the Curie point. At still lower temperatures, the magnetic symmetry can change in further transitions. The present results permit us to decide which magnetic structures can exist at temperatures just below and closest to the Curie point on restricting considerations to the thermodynamical conditions of a second-order phase transition and moreover to invariance of the thermodynamical functions under symmetry operations.

II. Magnetic second-order phase transitions in crystals

As known, in a second-order phase transition the state of a crystal changes continuously, whereas its phase undergoes an abrupt change involving a modification of the crystal symmetry. As worked out by Landau, in the immediate vicinity of the transition point the molar free energy Φ of the ordered phase can be expanded in a series:

$$\Phi = \Phi_0 + \Phi_1(\varphi(\boldsymbol{r})) + \Phi_2(\varphi(\boldsymbol{r})) + \Phi_3(\varphi(\boldsymbol{r})) + \ldots, \tag{1}$$

where

Φ_0 is the molar free energy of the disordered phase,

Φ_1 — a linear functional in a density function $\varphi(\boldsymbol{r})$ in the point $\boldsymbol{r}$

Φ_2, Φ_3 — functionals of the second and third degree in a density function $\varphi(\boldsymbol{r})$ in the point $\boldsymbol{r}$,

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and $\varphi(\boldsymbol{r})$ — a density function describing the change in state of the crystal. At the transition point, the function $\varphi(\boldsymbol{r})$ is expected to vanish. As the temperature is lowered, $\varphi(\boldsymbol{r})$ grows monotonously.

The basic assumption of the Landau — Lifshitz theory resides in a description of the change in state of the crystal by means of the function $\varphi(\boldsymbol{r})$ only. For the case of the paramagnetic — magnetic phase transition the function $\varphi(\boldsymbol{r})$ can be thought of as the statistical average magnetic moment density $\boldsymbol{m}(\boldsymbol{r})$. For the model of spins strictly attached to lattice points, the function $\boldsymbol{m}(\boldsymbol{r})$ can be defined as

$$\boldsymbol{m}(\boldsymbol{r}) = \sum_{i,\alpha} S_i^{(\alpha)} \sigma_\alpha(\boldsymbol{r}_i)\, \delta(\boldsymbol{r}-\boldsymbol{r}_i), \tag{2}$$

where i labels magnetic sublattices (points of the magnetic unit cell);

$\alpha = x, y, z$;

$S_i^{(\alpha)}$ is the α — component of the statistical average magnetic moment density in the lattice point $\boldsymbol{r}_i$;

$\sigma_\alpha(\boldsymbol{r}_i)$ — a unit axial vector attached to the point $\boldsymbol{r}_i$;

$\delta(\boldsymbol{r}-\boldsymbol{r}_i)$ — the Dirac delta function.

The function $\boldsymbol{m}(\boldsymbol{r})$ describes the change in state of the crystal consisting in the emergence of magnetic ordering below the Curie temperature. Besides, the axial vector function $\boldsymbol{m}(\boldsymbol{r})$ is by definition invariant under all the symmetry operations contained in the magnetic space group G of the crystal in the magnetic phase.

Let us now define an operation of an element $\hat{g}$ belonging to the magnetic space group on the axial vector function $\boldsymbol{m}(\boldsymbol{r})$:

$$\hat{g}\boldsymbol{m}(\boldsymbol{r}) = \sum_{i,\alpha} S_i^{(\alpha)} \hat{g}\, \sigma_\alpha(\boldsymbol{r}_i) \hat{g}\delta\,(\boldsymbol{r}-\boldsymbol{r}_i),$$

and

$$\hat{R}\boldsymbol{m}(\boldsymbol{r}) = -\boldsymbol{m}(\boldsymbol{r}),$$

where $\hat{R}$ is the time reversal operator. Obviously, for the components $m_\alpha(\boldsymbol{r}) = \sum_i S_i^{(\alpha)} \delta(\boldsymbol{r}-\boldsymbol{r}_i)$, one has

$$\hat{g} m_\alpha(\boldsymbol{r}) = m_\alpha(\hat{g}^{-1}\boldsymbol{r}), \tag{3a}$$

$$\hat{R} m_\alpha(\boldsymbol{r}) = -m_\alpha(\boldsymbol{r}). \tag{3b}$$

Let us assume that, in the paramagnetic phase, the magnetic space group of the crystal is $G_0 R$, where G_0 contains all the space operations leaving the crystal invariant. Acting on the function $m_\alpha(\boldsymbol{r})$ with all the elements of the group $G_0 R$ we obtain a set of functions Q $\{m_\alpha(\hat{g}^{-1}\boldsymbol{r})\}$, $\hat{g} \in G_0 R$.

The linear manifold of functions Q is a finite-dimensional linear space L, which trans-

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forms according to a representation[1] $\hat{T}$ of the group G_0R defined by (3a) and (3b):

$$\hat{T}(g)m_\alpha(\boldsymbol{r}) = m_\alpha(\hat{g}^{-1}\boldsymbol{r}),$$

$$\hat{T}(R)m_\alpha(\boldsymbol{r}) = -m_\alpha(\boldsymbol{r}).$$

A representation $\hat{T}$ can be decomposed into a direct sum of irreducible representations $\hat{\tau}^{(p)}$ of the group G_0R: $\hat{T} = \oplus\, \gamma_p\hat{\tau}^{(p)}$ (γ_p indicates how many times $\hat{\tau}^{(p)}$ is contained in $\hat{T}$).

The linear space L decomposes into orthogonal irreducible subspaces L_p transforming according to the representations $\tau^{(p)}$. Bases in all orthogonal subspaces L_p form a basis of the space L. The function $m_\alpha(\boldsymbol{r})$ as a function of the space L can be expanded in terms of the elements of this basis:

$$m_\alpha(\boldsymbol{r}) = \sum c'^{(p)}_{\alpha,k}\varphi^{(p)}_k(\boldsymbol{r}), \tag{4}$$

where p labels the irreducible representations of the group G_0R contained in the representation $\hat{T}$ (strictly speaking, p denotes a set of two indices: the star $\{\boldsymbol{k}\}$, and the number n labelling irreducible representations relating to the star $\{\boldsymbol{k}\}$);

k is the label of a basic vector in a subspace L_p transforming according to the representation $\hat{\tau}^{(p)}$.

Within the framework of the model of spins strictly attached to lattice points, it is possible to expand uniquely the function $m_\alpha(\boldsymbol{r})$ with coefficients $c'^{(p)}_{\alpha,k}$ constant throughout the crystal. It should be noted that an arbitrary density function cannot be uniquely expanded in terms of a finite number of functions unless their coefficients vary throughout the crystal and a special restriction has to be placed on any spatial variation of the $c'^{(p)}_{\alpha,k}$ within the crystal.

Since the function $m_\alpha(\boldsymbol{r})$ takes only real values, the subspaces L_p transform according to the real irreducible representations $\hat{\tau}^{(p)}$ or, if the representations $\hat{\tau}^{(p)}$ are non-real, we combine them with their complex-conjugate representations to obtain so-called physically inrreducible representations $\hat{T} = \hat{\tau}^{(p)} \oplus \hat{\tau}^{(p)*}$. In the basis defined above the axial vector function $\boldsymbol{m}(\boldsymbol{r})$ can be expressed as follows:

$$\boldsymbol{m}(\boldsymbol{r}) = \sum_{p,k,\alpha} c'^{(p)}_{\alpha,k}\sigma_\alpha\varphi^{(p)}_k(\boldsymbol{r}).$$

Under the element $\hat{g}$ of the group G_0R the functions $\varphi^{(p)}_k(\boldsymbol{r})$ transform according to the representation $\hat{\tau}^{(p)}$, and the axial vectors σ_α — according to the axial vector representation $\hat{\sigma}$. Then, the expressions $\sigma_\alpha\varphi^{(p)}_k(\boldsymbol{r})$ transform according to the direct product of the axial vector, representation $\hat{\sigma}$ and the irreducible representation $\hat{\tau}^{(p)}$. Such a product is in general a reducible representation of the group G_0R. For our further considerations we decompose the direct product $\hat{\sigma}\times\hat{\tau}^{(p)}$ into irreducible representations of the group G_0R and, simul-

[1] Operators of any representation of the group G_0R can be defined as follows:

1) $\hat{T}(g) = \dot{\hat{T}}(g)$

2) $\hat{T}(gR) = -\dot{\hat{T}}(g)$,

where $\hat{T}$ is an operator of a representation of the magnetic space group, while $\dot{\hat{T}}$ is an operator of a representation of the usual space group.

taneously the linear space containing elements of the form $\sigma_\alpha \varphi_k^{(p)}(\boldsymbol{r})$ can be decomposed into a direct sum of invariant mutually orthogonal subspaces. Choosing in each of these subspaces an arbitrary orthogonal basis, we obtain a basis in our space. The function $\boldsymbol{m}(\boldsymbol{r})$ is then expanded in terms of the elements of this basis:

$$\boldsymbol{m}(\boldsymbol{r}) = \sum_{i,l} c_l^{(i)} \boldsymbol{\varphi}_l^{(i)}(\boldsymbol{r}), \tag{5}$$

where i labels the irreducible representation;

$\boldsymbol{\varphi}_l^{(i)}(\boldsymbol{r})$ is a basis vector in the subspace transforming according to the irreducible representation $\hat{\tau}^{(i)}$. The coefficients of the expansion (5) are, simply, appropriate combinations of the coefficients of the expansion (4). We consider the basis functions as fixed, and the coefficients $c_l^{(i)}$ transform under the operations of $G_0 R$.

In the immediate vicinity of the Curie point the coefficients $c_l^{(i)}$ take arbitrarily small values and for temperatures $T \geqslant T_C$ are equal to zero.

Let us go back to the expansion of the molar free energy in terms of the vector function $\boldsymbol{m}(\boldsymbol{r})$ describing the magnetically ordered state. One should keep in mind that the free energy is invariant with respect to all symmetry operations and, moreover, under the time reversal operator. As known, for each representation $\hat{\tau}^{(i)}$ we can construct one quadratic invariant of the form $\sum_l |c_l^{(i)}|^2$. In general, the higher-order invariants cannot be written in so simple a form. Therefore, we can write

$$\Phi = \Phi_0 + \sum_i A^i \sum_l |c_l^{(i)}|^2 + \sum_j B_j f_j^{(4)}(c_l) + \sum_k B'_k f_k^{(6)}(c_l) + \ldots$$

where the $f^{(4)}, f^{(6)}, \ldots$ are invariants of order 4, 6, ... constructed from the coefficients $c_l^{(i)}$ transforming according to the irreducible representations of the group $G_0 R$. The equilibrium state of the system corresponds to that set of values of the $c_l^{(i)}$ which minimizes Φ at a given temperature and pressure. The disordered phase is characterized by $c^{(i)} \equiv 0$. Below the Curie temperature, a non-zero magnetic moment density appears, and therefore not all the $c_l^{(i)}$ vanish so that at least one of the A^i changes its sign to negative at the transition point. It is easily shown that the A^i related with the quadratic invariant in the expansion of Φ takes the form $A^i = A\,(T - T_C)$. Since our considerations are restricted to the immediate vicinity of the transition point, we can neglect mixed invariants transforming according to products of irreducible representations (of order 2 and higher).

We thus have (dropping the index of the representation $\tau^{(i)}$):

$$\Phi = \Phi_0 + A(T - T_C) \sum_l |c_l|^2 + \sum_j B_j f_j^{(4)}(c_l) + \sum_k B'_k f_k^{(6)}(c_l) + \ldots \tag{6}$$

and

$$\boldsymbol{m}(\boldsymbol{r}) = \sum_l c_l \boldsymbol{\varphi}_l(\boldsymbol{r}). \tag{7}$$

By miniziming the free energy Φ considered as a function of the coefficients c_l we can determine the relations between the coefficients c_l and, moreover, examine the symmetry of the function $\boldsymbol{m}(\boldsymbol{r})$. Having found the function $\boldsymbol{m}(\boldsymbol{r})$, we easily find the magnetic space group taken by the crystal below the Curie point. In order to obtain this group, we have

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to determine all the elements of the group G_0R that leave unchanged the function $\boldsymbol{m}(\boldsymbol{r})$. As seen, the magnetic symmetry group of the ordered phase (G) is necessarily a subgroup of the magnetic symmetry group of the disordered phase (G_0R): $G \subset G_0R$.

Since the axial vector function of the magnetic moment density was expanded with coefficients constant throughout the crystal, we omit the Landau–Lifshitz criterion of activity imposed on those irreducible representations of the group G_0R for which nonzero $c_i^{(i)}$ can occur. This criterion eliminates all the representations whose antisymmetric direct product contains in its decomposition the vector representation of G_0. It should be noted that the Landau–Lifshitz criterion is related with the assumption of slow spatial variation of the coefficients $c_i^{(i)}$. This restriction need not be used with respect to the axial vector magnetic moment density function $\boldsymbol{m}(\boldsymbol{r})$ within the framework of the model of spins strictly attached to lattice points bacause of the special deltoidal form taken by $\boldsymbol{m}(\boldsymbol{r})$ in this model, and moreover because of the finite number of constant components $S_i^{(\alpha)}$ (see: Eq. (2)).

III. Magnetic moment vectors at sites of the magnetic unit cell

In order to solve the problem of the magnetic moment axial vectors attached to the sites of the magnetic unit cell, one has to construct a axial vector function with a finite number of parameters which can vary arbitrarily, expressed in the space of real positions in the crystal.

Under the element of G (the magnetic space group exhibited by the crystal in the ordered phase), this function transforms according to the unit representation of G.

The above procedure can be carried out as follows: let us assume a magnetic moment attached to an arbitrary site $\boldsymbol{r}_i$ of the mangetic unit cell:

$$\boldsymbol{m}(\boldsymbol{r}_i) = m_x\sigma_x(\boldsymbol{r}_i) + m_y\sigma_y(\boldsymbol{r}_i) + m_z\sigma_z(\boldsymbol{r}_i).$$

Under the element $\hat{g} \in G$ the axial vectors $\sigma_\alpha(\alpha = x, y, z)$ rotate and the magnetic moments change their positions. If the sum consisting of all functions of the form $\hat{g}\boldsymbol{m}(\boldsymbol{r}_i)$, where $\hat{g} \in G$, contains components related to all the sites of the magnetic unit cell, the problem is solved. If not, we choose any other arbitrary function $\boldsymbol{m}'(\boldsymbol{r}_j)$:

$$\boldsymbol{m}'(\boldsymbol{r}_j) = m'_x\sigma_x(\boldsymbol{r}_j) + m'_y\sigma_y(\boldsymbol{r}_j) + m'_z\sigma_z(\boldsymbol{r}_j),$$

where $\boldsymbol{r}_j \neq \hat{g}^{-1}\boldsymbol{r}_i (\hat{g} \in G)$.

The procedure must then be repeated. The sum containing all functions of the form $\hat{g}\boldsymbol{m}'(\boldsymbol{r}_j)$ $(\hat{g} \in G)$ we add to the sum obtained previously. The finally obtained axial vector function contains magnetic moments relating to all the sites of the magnetic unit cell.

IV. Application

Let us assume that magnetic ions (atoms) are attached to the sites of both sublattices: A and B (see: Fig. 1). The space group D_{6h}^4 belongs to the class D_{6h}. Each element of the group D_{6h}^4 represents a product of the translation

$$t_{m_1\boldsymbol{a}_1+m_2\boldsymbol{a}_2+m_3\boldsymbol{a}_3}(m_1, m_2, m_3 = 0, \pm 1, \ldots; \boldsymbol{a}_3 \perp \boldsymbol{a}_1, \boldsymbol{a}_2)$$

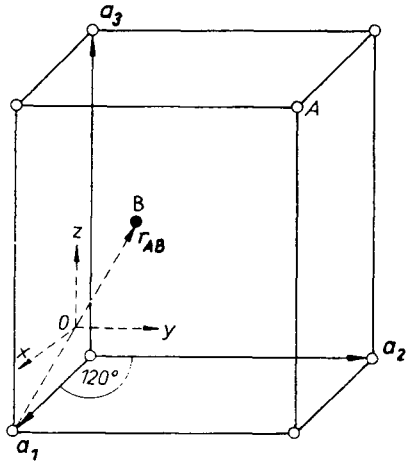

Fig. 1. ○ A sublattice side, ● B sublattice side. $\boldsymbol{a}_3 \perp \boldsymbol{a}_1, \boldsymbol{a}_2$; $\boldsymbol{r}_{AB} = \frac{1}{3}\boldsymbol{a}_1 - \frac{1}{3}\boldsymbol{a}_2 + \frac{1}{2}\boldsymbol{a}_3$

and one of the following 24 elements:

$$e,\ t_\alpha C_6,\ C_6^2,\ t_\alpha C_6^3,\ C_6^4,\ t_\alpha C_6^5,\ u_1,\ t_\alpha u_2,\ u_3,\ t_\alpha u_4,\ u_5,\ t_\alpha u_6,\ I,\ t_\alpha S_6^4,\ S_6^5,\ t_\alpha \sigma_h,\ S_6,\ t_\alpha S_6^2,\ \sigma_4,\ t_\alpha \sigma_5,\ \sigma_6,$$

$$t_\alpha \sigma_1,\ \sigma_2,\ t_\alpha \sigma_3$$

where

$$\boldsymbol{\alpha} = \frac{1}{2}\boldsymbol{a}_3$$

$C_6, C_6^2, \ldots, C_6^5$ — rotations about the axis $\boldsymbol{a}_3$ by all integer multiples of the angle $\pi/3$;
σ_h — a reflection in the horizontal plane;
$S_6^{(k)} = \sigma_h C_6^{(k)}$ — $(k = 1, 2, \ldots 5)$;
$u_1, \ldots, u_6$ — horizontal twofold axes;
$\sigma_1, \ldots, \sigma_6$ — vertical reflection planes.

The magnetic space group $D_{6h}^4 R$ is the direct product of the space group D_{6h}^4 and the group R consisting of two elements e and R.

We now proceed to discuss all the non-equivalent irreducible representations of the space group D_{6h}^4. As known, its irreducible representations are related with 17 stars (Kovalev 1961). In Part I of this paper, it is our intention to discuss the representation relating to the star $\{\boldsymbol{k}_{12}\}$ (see: Kovalev 1961), where $\boldsymbol{k}_{12} = \frac{1}{2}\boldsymbol{b}_1$ (where $\boldsymbol{b}_1, \boldsymbol{b}_2, \boldsymbol{b}_3$ are basic vectors of the reciprocal lattice

$$\boldsymbol{b}_1 = \frac{[\boldsymbol{a}_2, \boldsymbol{a}_3]}{(\boldsymbol{a}_1, [\boldsymbol{a}_2, \boldsymbol{a}_3])}, \quad \boldsymbol{b}_2 = \frac{[\boldsymbol{a}_3, \boldsymbol{a}_1]}{(\boldsymbol{a}_1, [\boldsymbol{a}_2, \boldsymbol{a}_3])}, \quad \boldsymbol{b}_3 = \frac{[\boldsymbol{a}_1, \boldsymbol{a}_2]}{(\boldsymbol{a}_1, [\boldsymbol{a}_2, \boldsymbol{a}_3])}.$$

The set of inequivalent irreducible representations related with the star $\{\boldsymbol{k}_{12}\}$ consists of 8 representations $\tau^{(i)}$ $(i = 1, 2, \ldots 8)$ all of them real.

We require for $T < T_C$ that the magnetic moment density shall be non-zero in crystal lattice points. This means that the representation $\tau^{(i)}$ according to which the components $m_\alpha(\boldsymbol{r})$ ($\alpha = x, y, z$) transform under the element $\hat{g} \in G_0 R$ must contain in its decomposition the unit representation of the point subgroup $\mathcal{H}$ of the group G_0, with the centre of the group occurring at a lattice point (see: Kovalev 1963). The above criterion is fulfilled for the representations $\tau^{(1)}$ and $\tau^{(8)}$. Hence, the axial vector function of the magnetic moment density transforms under $\hat{g} \in G_0 R$ according to the simple product $\sigma \times \tau^{(1)}$ or $\sigma \times \tau^{(8)}$. Decomposing these products into simple sums consisting of irreducible representations of the group D_{6h}^4, we find that these sums contain representations[2] $\tau^{(i)}$, where $i = 2, 3, 4, 5, 6, 7$. All the representations $\tau^{(i)}$ are three-dimensional. After simple derivations, one obtains that the formula (6) takes the same form for all the $\tau^{(i)}$:

$$\Phi = \Phi_0 + A(T - T_C)(c_1^2 + c_2^2 + c_3^2) + B_1(c_1^2 + c_2^2 + c_3^2)^2 + B_2(c_1^4 + c_2^4 + c_3^4) + \ldots,$$

where c_1, c_2, c_3 are coefficients in the expansion of the magnetic moment density function (see: Eq. (7)).

The axial vector function of the magnetic moment density can be expressed as follows (for all $\tau^{(i)}$):

$$\boldsymbol{m}(\boldsymbol{r}) = c_1 \boldsymbol{\varphi}_1(\boldsymbol{r}) + c_2 \boldsymbol{\varphi}_2(\boldsymbol{r}) + c_3 \boldsymbol{\varphi}_3(\boldsymbol{r}).$$

Examining the minimum value of the expression Φ with respect to three independent variables c_1, c_2, c_3 we obtain a more particularized form of the function $\boldsymbol{m}(\boldsymbol{r})$. Our results take the following form:

$$\text{I) 1) } \boldsymbol{m}(\boldsymbol{r}) = \pm c\boldsymbol{\varphi}_1(\boldsymbol{r}),$$

$$\text{2) } \boldsymbol{m}(\boldsymbol{r}) = \pm c\boldsymbol{\varphi}_2(\boldsymbol{r}),$$

$$\text{3) } \boldsymbol{m}(\boldsymbol{r}) = \pm c\boldsymbol{\varphi}_3(\boldsymbol{r}),$$

and

$$\text{II) 1) } \boldsymbol{m}(\boldsymbol{r}) = \pm c[\boldsymbol{\varphi}_1(\boldsymbol{r}) + \boldsymbol{\varphi}_2(\boldsymbol{r}) + \boldsymbol{\varphi}_3(\boldsymbol{r})],$$

$$\text{2) } \boldsymbol{m}(\boldsymbol{r}) = \pm c[-\boldsymbol{\varphi}_1(\boldsymbol{r}) + \boldsymbol{\varphi}_2(\boldsymbol{r}) + \boldsymbol{\varphi}_3(\boldsymbol{r})],$$

$$\text{3) } \boldsymbol{m}(\boldsymbol{r}) = \pm c[\boldsymbol{\varphi}_1(\boldsymbol{r}) - \boldsymbol{\varphi}_2(\boldsymbol{r}) + \boldsymbol{\varphi}_3(\boldsymbol{r})],$$

$$\text{4) } \boldsymbol{m}(\boldsymbol{r}) = \pm c[\boldsymbol{\varphi}_1(\boldsymbol{r}) + \boldsymbol{\varphi}_2(\boldsymbol{r}) - \boldsymbol{\varphi}_3(\boldsymbol{r})].$$

Each of the above solutions is related to all the representations $\tau^{(i)}$, where $i = 2, 3, 4, 5, 6, 7$.

Information on the magnetic orderings related with the solutions[3] I is tabulated below.

From our results it follows that the solutions of type I lead to antiferromagnetic collinear orderings. The magnetic structures I 1 for $i = 3$ and I 3 for $i = 5$ are shown in Figs 2*a* and 2*b*, which represent projections of the magnetic unit cells on the plane of the vectors $\boldsymbol{a}_1$, $\boldsymbol{a}_2$ (basic vectors of the hcp lattice). In Fig. 2*a* one can see the ferromagnetic planes

[2] The representations $\tau^{(i)}(i = 2, 3, 4, \ldots, 7)$ fulfil the Landau–Lifshitz criterion of activity.

[3] The results contained in Table I 1 are consistent with those obtained by Kovalev (see: Kovalev 1963).

TABLE I 1

Index of the representation $\tau^{(i)}$	Magnetic space group G	Basic vectors of the magnetic unit cell	Magnetic moments attached to the particular sites of the magnetic unit cell			
2	$D_{2h}^{6}\left(\frac{1}{2}\boldsymbol{a}_1' + \frac{1}{2}\boldsymbol{a}_2'\middle\vert R\right)$	$\boldsymbol{a}_2, -2\boldsymbol{a}_1-\boldsymbol{a}_2, \boldsymbol{a}_3$	$m_x, 0, 0$	$-m_x, 0, 0$	$m_x, 0, 0$	$-m_x, 0, 0$
3	$D_{2h}^{16}\left(\frac{1}{2}\boldsymbol{a}_2' + \frac{1}{2}\boldsymbol{a}_3'\middle\vert R\right)$	$\boldsymbol{a}_3, 2\boldsymbol{a}_1+\boldsymbol{a}_2, \boldsymbol{a}_2$	$0, 0, m_z$	$0, 0, m_z$	$0, 0, -m_z$	$0, 0, -m_z$
4	$D_{2h}^{16}\left(\frac{1}{2}\boldsymbol{a}_1' + \frac{1}{2}\boldsymbol{a}_3'\middle\vert R\right)$	$\boldsymbol{a}_2, \boldsymbol{a}_3, 2\boldsymbol{a}_1+\boldsymbol{a}_2$	$0, m_y, 0$	$0, -m_y, 0$	$0, m_y, 0$	$0, -m_y, 0$
5	$D_{2h}^{12}\left(\frac{1}{2}\boldsymbol{a}_1' + \frac{1}{2}\boldsymbol{a}_3'\middle\vert R\right)$	$-2\boldsymbol{a}_1-\boldsymbol{a}_2, \boldsymbol{a}_3, \boldsymbol{a}_2$	$0, m_y, 0$	$0, -m_y, 0$	$0, m_y, 0$	$0, -m_y, 0$
6	$D_{2h}^{11}\left(\frac{1}{2}\boldsymbol{a}_2' + \frac{1}{2}\boldsymbol{a}_3'\middle\vert R\right)$	$\boldsymbol{a}_3, 2\boldsymbol{a}_1+\boldsymbol{a}_2, \boldsymbol{a}_2$	$0, 0, m_z$,	$0, 0, m_z$	$0, 0, -m_z$	$0, 0, -m_z$
7	$D_{2h}^{14}\left(\frac{1}{2}\boldsymbol{a}_2' + \frac{1}{2}\boldsymbol{a}_3'\middle\vert R\right)$	$\boldsymbol{a}_3, -\boldsymbol{a}_2, 2\boldsymbol{a}_1+\boldsymbol{a}_2$	$m_x, 0, 0$	$-m_x, 0, 0$	$m_x, 0, 0$	$-m_x, 0, 0$

TABLE I 2

Index of the representations $\tau^{(i)}$	Magnetic space group G	Basic vectors of the magnetic unit cell	Magnetic moments attached to the particular sites of the magnetic unit cell			
2	$D_{2h}^{6}\left(\frac{1}{2}\boldsymbol{a}_1' + \frac{1}{2}\boldsymbol{a}_2'\middle\vert R\right)$	$\boldsymbol{a}_1, \boldsymbol{a}_1+2\boldsymbol{a}_2, \boldsymbol{a}_3$	$0, m_y, 0$	$0, -m_y, 0$	$0, m_y, 0$	$0, -m_y, 0$
3	$D_{2h}^{16}\left(\frac{1}{2}\boldsymbol{a}_2' + \frac{1}{2}\boldsymbol{a}_3'\middle\vert R\right)$	$\boldsymbol{a}_3, -\boldsymbol{a}_1-2\boldsymbol{a}_2, \boldsymbol{a}_1$	$0, 0, m_z$	$0, 0, m_z$	$0, 0, -m_z$	$0, 0, -m_z$
4	$D_{2h}^{16}\left(\frac{1}{2}\boldsymbol{a}_1' + \frac{1}{2}\boldsymbol{a}_3'\middle\vert R\right)$	$-\boldsymbol{a}_1, \boldsymbol{a}_3, \boldsymbol{a}_1+2\boldsymbol{a}_2$	$m_x, 0, 0$	$-m_x, 0, 0$	$m_x, 0, 0$	$-m_x, 0, 0$
5	$D_{2h}^{12}\left(\frac{1}{2}\boldsymbol{a}_2' + \frac{1}{2}\boldsymbol{a}_3'\middle\vert R\right)$	$\boldsymbol{a}_3, -\boldsymbol{a}_1-2\boldsymbol{a}_2, \boldsymbol{a}_1$	$m_x, 0, 0$	$-m_x, 0, 0$	$m_x, 0, 0$	$-m_x, 0, 0$
6	$D_{2h}^{11}\left(\frac{1}{2}\boldsymbol{a}_2' + \frac{1}{2}\boldsymbol{a}_3'\middle\vert R\right)$	$\boldsymbol{a}_3, -\boldsymbol{a}_1-2\boldsymbol{a}_2, \boldsymbol{a}_1$	$0, 0, m_z$	$0, 0, m_z$	$0, 0, -m_z$	$0, 0, -m_z$
7	$D_{2h}^{14}\left(\frac{1}{2}\boldsymbol{a}_2' + \frac{1}{2}\boldsymbol{a}_3'\middle\vert R\right)$	$\boldsymbol{a}_3, \boldsymbol{a}_1, \boldsymbol{a}_1+2\boldsymbol{a}_2$	$0, m_y, 0$	$0, -m_y, 0$	$0, m_y, 0$	$0, -m_y, 0$

TABLE I 3

Index of the representations $\tau^{(i)}$	Magnetic space group G	Basic vectors of the magnetic unit cell	Magnetic moments attached to the particular sites of the magnetic unit cell			
2	$D_{2h}^{6}\left(\frac{1}{2}\boldsymbol{a}_1'+\frac{1}{2}\boldsymbol{a}_2'\vert R\right)$	$\boldsymbol{a}_1+\boldsymbol{a}_2, -\boldsymbol{a}_1+\boldsymbol{a}_2, \boldsymbol{a}_3$	$0, m_y, 0$	$0, -m_y, 0$	$0, m_y, 0$	$0, -m_x, 0$
3	$D_{2h}^{16}\left(\frac{1}{2}\boldsymbol{a}_2'+\frac{1}{2}\boldsymbol{a}_3'\vert R\right)$	$\boldsymbol{a}_3, \boldsymbol{a}_1-\boldsymbol{a}_2, \boldsymbol{a}_1+\boldsymbol{a}_2$	$0, 0, m_z$	$0, 0, m_z$	$0, 0, -m_z$	$0, 0, -m_z$
4	$D_{2h}^{16}\left(\frac{1}{2}\boldsymbol{a}_1'+\frac{1}{2}\boldsymbol{a}_3'\vert R\right)$	$-\boldsymbol{a}_1-\boldsymbol{a}_2, \boldsymbol{a}_3, -\boldsymbol{a}_1+\boldsymbol{a}_2$	$m_x, 0, 0$	$-m_x, 0, 0$	$m_x, 0, 0$	$-m_x, 0, 0$
5	$D_{2h}^{12}\left(\frac{1}{2}\boldsymbol{a}_2'+\frac{1}{2}\boldsymbol{a}_3'\vert R\right)$	$\boldsymbol{a}_3, \boldsymbol{a}_1-\boldsymbol{a}_2, \boldsymbol{a}_1+\boldsymbol{a}_2$	$m_x, 0, 0$	$-m_x, 0, 0$	$m_x, 0, 0$	$-m_x, 0, 0$
6	$D_{2h}^{11}\left(\frac{1}{2}\boldsymbol{a}_2'+\frac{1}{2}\boldsymbol{a}_3'\vert R\right)$	$\boldsymbol{a}_3, \boldsymbol{a}_1-\boldsymbol{a}_2, \boldsymbol{a}_1+\boldsymbol{a}_2$	$0, 0, m_z$	$0, 0, m_z$	$0, 0, -m_z$	$0, 0, -m_z$
7	$D_{2h}^{14}\left(\frac{1}{2}\boldsymbol{a}_2'+\frac{1}{2}\boldsymbol{a}_3'\vert R\right)$	$\boldsymbol{a}_3, \boldsymbol{a}_1+\boldsymbol{a}_2, -\boldsymbol{a}_1+\boldsymbol{a}_2$	$0, m_y, 0$	$0, -m_y, 0$	$0, m_y, 0$	$0, -m_y, 0$

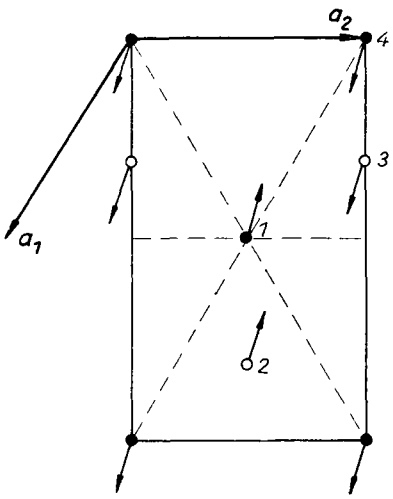

Fig. 2a

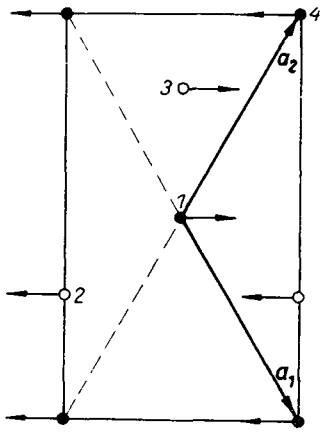

Fig. 2b

Fig. 2a. Projection (001). The sites represented by solid circles are situated in the plane of the drawing whereas those represented by open circles are located in the plane parallel to the drawing but displaced along the $\boldsymbol{a}_3$ vector by $\frac{1}{2}\boldsymbol{a}_3$ with respect to the plane of the drawing

Fig. 2b. See Fig. 2a for explaining

mutually parallel and perpendicular to the plane of the figure; moreover, in the direction of the basic vector $\boldsymbol{a}_2$, the planes have the ordering $+-+-\ldots$, whereas in the direction perpendicular to $\boldsymbol{a}_2$ the ordering is $++--\ldots$ In Fig. 2*b*, the situation is analogous. The ferromagnetic mutually parallel planes perpendicular to the plane of the drawing are indicated. In the direction of the vector $\boldsymbol{a}_1+\boldsymbol{a}_2$ the planes take the ordering $+-+-\ldots$, whereas in the direction perpendicular thereto their ordering is $++--++\ldots$.

Let us now proceed to discuss the magnetic orderings arising below the Curie point in relation with the magnetic moment density functions II. First, we shall find the magnetic ordering to which the density function of the form $\boldsymbol{m}(\boldsymbol{r}) = \pm c[\boldsymbol{\varphi}_1^{(2)}(\boldsymbol{r})+\boldsymbol{\varphi}_2^{(2)}(\boldsymbol{r})+\boldsymbol{\varphi}_3^{(2)}(\boldsymbol{r})]$ is related (see: II 1, $i = 2$). In the immediate vicinity of the Curie point such an ordering belongs to the magnetic space group $G = D_6^6(I|R)$ (I is the inversion operator). The basic vectors of the magnetic unit cell take the form $\boldsymbol{a}_1' = 2\boldsymbol{a}_1$, $\boldsymbol{a}_2' = 2\boldsymbol{a}_2$ and $\boldsymbol{a}_3' = \boldsymbol{a}_3$.

In the ensuing Table, the components of the statistical average magnetic moments attached to the particular points of the magnetic unit cell are given.

TABLE 1 a

Components of average magnetic moments

Number of the site	1	2	3	4	5	6	7	8
m_x	m	$\frac{1}{2}m$	$-\frac{1}{2}m$	$-m$	$-\frac{1}{2}m$	$\frac{1}{2}m$	0	0
m_y	0	$-\frac{\sqrt{3}}{2}m$	$-\frac{\sqrt{3}}{2}m$	0	$\frac{\sqrt{3}}{2}m$	$\frac{\sqrt{3}}{2}m$	0	0
m_z	0	0	0	0	0	0	0	0

m denotes an arbitrary constant value.

It is easily seen that the values of the magnetic moments attached to the unit cell sites 1, 2, 3, 4, 5, 6 are equal to one another, whereas those attached to the sites 7, 8 are indetically zero.

This magnetic ordering is shown in Fig. 3 in the form of the projection of the magnetic unit cell on the plane perpendicular to the basic vector $\boldsymbol{a}_3$ of the hcp lattice. The magnetic moments are indicated by means of arrows. This structure exhibits a ferrimagnetic character.

A result analogous to the above one is obtained for the magnetic moment density II 1 with $i = 4$. In this case the magnetic space group of the structure arising just below the Curie temperature is $G = C_{6v}^4(I|R)$. The magnetic unit cell has the same basic vectors as previously and moreover analogous statistical average magnetic moments are attached to the sites of the magnetic unit cell. Two magnetic moments take zero values. In the planes perpendicular to the basic vector $\boldsymbol{a}_3$ the magnetic moments in the particular sites are rotated about the $\boldsymbol{a}_3$ vector with respect to one another. The angles between them amount to 120°.

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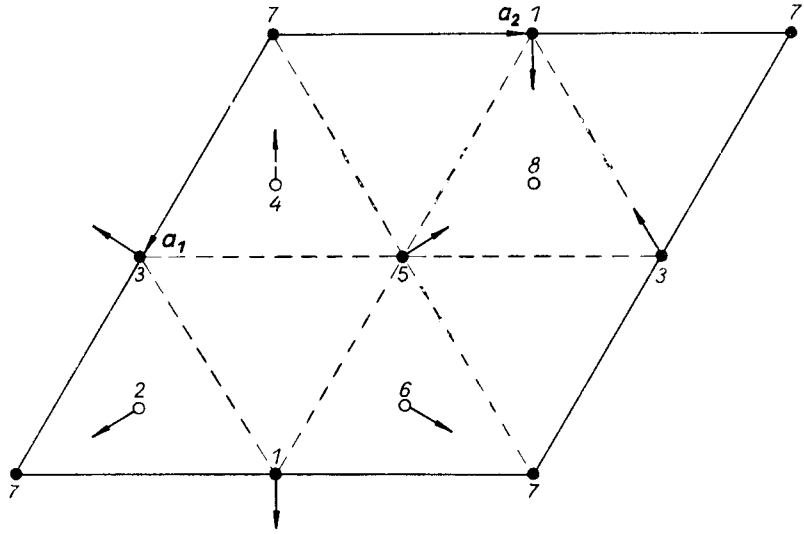

Fig. 3. See Fig. 2*a* for explaining

The relative values of the magnetic moments with the exception of the two zero magnetic moments are equal to one another.

For the magnetic moment density in the form given by the solution II 1 with $i = 3 : \boldsymbol{m}(\boldsymbol{r}) = \pm c[\boldsymbol{\varphi}_1^{(3)}(\boldsymbol{r}) + \boldsymbol{\varphi}_2^{(3)}(\boldsymbol{r}) + \boldsymbol{\varphi}_3^{(3)}(\boldsymbol{r})]$ one obtains the magnetic space groups $G = C_{6h}^2(u|R)$ (u is a twofold axis perpendicular to the axis C_6). The basic vectors of the magnetic unit cell take the values $\boldsymbol{a}_1' = 2\boldsymbol{a}_1$, $\boldsymbol{a}_2' = 2\boldsymbol{a}_2$, $\boldsymbol{a}_3' = \boldsymbol{a}_3$. The magnetic moments attached to the sites of the magnetic unit cell are presented in the following Table.

TABLE I b

Magnetic moments of the magnetic unit cell

Number of the site	1	2	3	4	5	6	7	8
m_x	0	0	0	0	0	0	0	0
m_y	0	0	0	0	0	0	0	0
m_z	m	m	m	m	m	m	m'	m'

m, m' take arbitrary constant values.

This is a typical ferrimagnetic structure (see: Fig. 4). Two magnetic moments attached to two sites of the magnetic unit cell take values m' different from that m of the remaining six magnetic moments.

Ferrimagnetic structure is also obtained when the magnetic moment density function takes the form as for II 1 : $i = 5$. In this case the magnetic space group exhibited by the crystal below the transition point is $G = C_{2h}^1\left(\frac{1}{2}a_2' \middle| Ru\right)$. The basic vectors of the magnetic

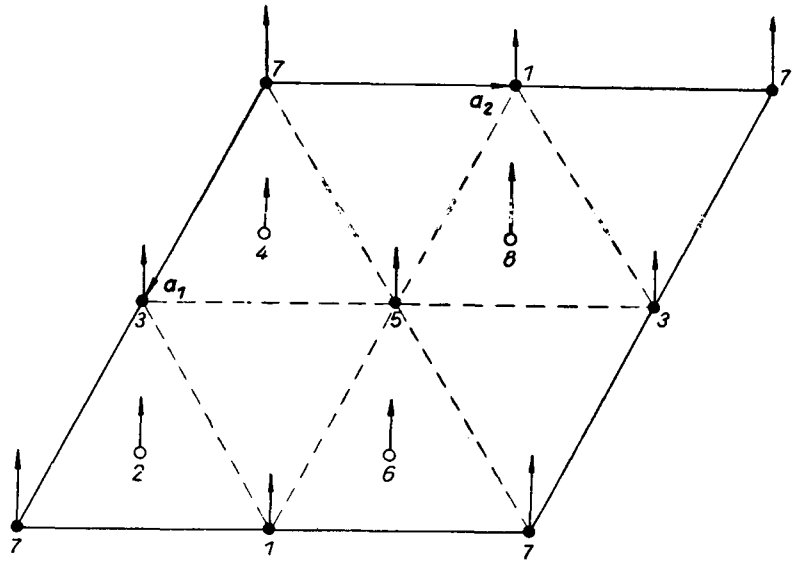

Fig. 4. See Fig. 2*a* for explaining

unit cell are considerably elongated in comparison with the preceding ones, and take the values $\boldsymbol{a}_1' = 2\boldsymbol{a}_1 + 4\boldsymbol{a}_2$, $\boldsymbol{a}_2' = \boldsymbol{a}_3$, $\boldsymbol{a}_3' = 2\boldsymbol{a}_1$. The magnetic unit cell consits of 16 magnetic sites. The magnetic moments attached to these assume four distinct values and, moreover, the x-components of the magnetic moments mutually cancel out while all the z-components can take non-zero values (the y-components all vanish).

TABLE II 2

Number of the representation $\tau^{(i)}$	Magnetic space group G	The type of magnetic structure arising in relation with the magnetic phase transition (i)
2	$D_2^2\,(I\|R)$	Ferrimagnetic
3	$C_{2h}^2\left(\frac{1}{2}\,\boldsymbol{a}_3'Ru\right)$	Ferrimagnetic
4	$C_{2v}^4\,(I\|R)$	Ferrimagnetic
5	$S_2^1\left(\frac{1}{2}\,\boldsymbol{a}_3\|RC_2\right)$	Ferrimagnetic
6	$C_s^1\,(I\|R)$	Ferrimagnetic
7	$S_2^1\left(\frac{1}{2}\,\boldsymbol{a}_3\|RC_2\right)$	Ferrimagnetic

The basic vectors of the magnetic unit cell are equal to $2\boldsymbol{a}_1+2\boldsymbol{a}_2$, $-2\boldsymbol{a}_1+2\boldsymbol{a}_2$, $\boldsymbol{a}_3$

TABLE II 3

Number of the representation $\tau^{(i)}$	Magnetic space group G	The type of magnetic structure arising in relation with the magnetic phase transition (i)
2	$D_2^2\,(I\|R)$	Ferrimagnetic
3	$C_{2h}^2\,(u\|R)$	Ferrimagnetic
4	$C_{2v}^4\,(I\|R)$	Ferrimagnetic
5	$S_2^1\left(\frac{1}{2}\,\boldsymbol{a}_3\|RC_2\right)$	Ferrimagnetic
6	$C_S^1\,(I\|R)$	Ferrimagnetic
7	$S_2^1\left(\frac{1}{2}\,\boldsymbol{a}_3\|RC_2\right)$	Ferrimagnetic

The basic vectors of the magnetic unit cell amount to $4\boldsymbol{a}_1 + 2\boldsymbol{a}_2, 2\boldsymbol{a}_2, \boldsymbol{a}_3$.

TABLE II 4

Number of the representation $\tau^{(i)}$	Magnetic space group G	The type of magnetic structure arising in relation with the magnetic phase transition (i)
2	$D_2^2\,(I\|R)$	Ferrimagnetic
3	$C_{2h}^2\,(u\|R)$	Ferrimagnetic
4	$C_{2v}^4\,(I\|R)$	Ferrimagnetic
5	$C_{2h}^1\left(\frac{1}{2}\,\boldsymbol{a}_3\|RC_2\right)$	Ferrimagnetic
6	$C_{2v}^4\,(I\|R)$	Ferrimagnetic
7	$C_{2h}^4\left(\frac{1}{2}\,\boldsymbol{a}_3\|RC_2\right)$	Ferrimagnetic

The basic vectors of the magnetic unit cell are $2\boldsymbol{a}_1, 2\boldsymbol{a}_1 + 4\boldsymbol{a}_2, \boldsymbol{a}_3$.

For the magnetic moment density function expressed in the form

$$\boldsymbol{m}(\boldsymbol{r}) = \pm c\,[\boldsymbol{\varphi}_1^{(6)}(\boldsymbol{r}) + \boldsymbol{\varphi}_2^{(6)}(\boldsymbol{r}) + \boldsymbol{\varphi}_3^{(6)}(\boldsymbol{r})]$$

(I 1 with $i = 6$) one obtains a ferrimagnetic ordering. Besides, in this case, we have full compensation of the magnetic moments belonging to one magnetic unit cell. The basic vectors of the magnetic unit cell are equal to $-\boldsymbol{a}_3$, $2\boldsymbol{a}_1 + 4\boldsymbol{a}_2$, $2\boldsymbol{a}_1$; the magnetic symmetry group exhibited by the crystal is $G = C_{2v}^4(I|R)$.

Ferrimagnetic structure is also related with the function $\boldsymbol{m}(\boldsymbol{r})$ of the type I 1 with $i = 7$. The magnetic space group of the crystal is $G = C_{2h}^4\left(\frac{1}{2}\,\boldsymbol{a}_1'\,|\,RC_2\right)$. The magnetic unit cell

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basic vectors are as follows: $\boldsymbol{a}_3$, $2\boldsymbol{a}_1$, $2\boldsymbol{a}_1+4\boldsymbol{a}_2$. The magnetic unit cell contains 16 sites, of which only 8 have non-zero magnetic moments attached to them; furhermore, these magnetic moments mutually cancel within the magnetic unit cell.

We now proceed to discuss in short the results related with a magnetic moment density of the type II 2, II 3 and II 4 for $i = 2, 3, 4, 5, 6, 7$. These results do not lead to any new magnetic orderings different from the preceding ones (II 1). The results are tabulated below.

The magnetic second-order phase transitions relating to the remaining non-equivalent irreducible representations of the magnetic space group D^4_{6h} R will be discussed in further parts of this paper. There, we shall present the final conclusions and compare some of the results with experimental data.

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