

spin flip). It is precisely this difference in the sign of  $|\Delta_i|^2$  in the coherence factors which is responsible for the different temperature dependences of the kinetic coefficients (a monotonic increase of the ultrasound absorption with increasing temperature, as against a plot with a maximum below the transition point in the case of nuclear-spin relaxation). From the terms responsible to the interband scattering in (A.4) and (A.5) it is seen that the signs of  $|\Delta_i|$  and  $|\Delta_j|$  are different for the solution with  $\varphi_{12} = \pi$  than for the intraband processes, and therefore at  $g_{12} \sim g_{11}$  the temperature dependence of the sound absorption has a maximum, just as in the case of the spin-lattice relaxation rate.

We note that in the case of the second solution the system is more stable to impurity scattering, since the intraband scatterings are partially compensated by the interband scatterings (because the diagrams for the interband scattering enter with opposite signs).

Thus, the relative phase difference of the order parameters  $\Delta_{11}$  and  $\Delta_{22}$ , as a result of interband transitions, should lead to observable physical phenomena.

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## Possible types of magnetic ordering of S ions in the garnet structure

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A model approach is proposed to determine and classify, within the framework of the exchange approximation, the types of magnetic ordering that are produced when a crystal goes over into a magnetically ordered state. Within the framework of this approach, the possible types of ordering in the garnet structure are determined for the case when the magnetic and crystal-chemical cells coincide. It is shown that some magnetic structures cause tetragonal or trigonal lattice distortions due to exchange striction. All the obtained exchange structures are in correspondence with the exchange classes introduced by Andreev and Marchenko.

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The magnetic properties of antiferromagnetic garnets are the subject of a rather larger number of studies (see the review of Belov and Sokolov<sup>1</sup>). A theoretical analysis of some types of magnetic ordering in such structures was carried out by the Bertaut method<sup>2</sup> in a number of studies<sup>3-6</sup>. This method is based on an analysis of a model quadratic spin Hamiltonian. It will be shown in this paper, however, that inclusion of only the interactions that are quadratic in the spin is insufficient for a complete determination of all the possible magnetic configurations.

A preferable method that determines all the possible types of magnetic ordering in crystals with different symmetries is the expansion of the spin-density vector in irreducible representations of the symmetry group of the paramagnetic state of the crystal.<sup>7,8</sup> It is precisely by this method that von Prandl<sup>9</sup> obtained and classified by symmetry type all the possible types of magnetic configurations of the spins in the garnet structure. He took into account to an equal degree both the exchange and the relativistic interactions. At the same time, for crystals with magnetic S ions, a much sim-

pler treatment is possible on the basis of an analysis of the symmetry of only the exchange interactions.

This approach yields the disposition of the atomic spins relative to one another, but does not fix their disposition relative to the crystal axes. In addition, it is possible to construct a classification of the magnetic structure within the framework of this method. This classification differs somewhat from that assumed by Andreev and Marchenko,<sup>10</sup> although there is a one-to-one correspondence between them. The point is that the classification developed by Andreev and Marchenko<sup>10</sup> presupposes knowledge of the symmetry of the crystal lattice in the magnetically ordered state, while the classification assumed in the present paper presupposes only knowledge of the symmetry of the crystal lattice in the paramagnetic phase. For each magnetic configuration it is possible to indicate those lattice distortions which will arise in the magnetically ordered state.

In the present paper, a localized-spin model is used to obtain and classify by symmetry type all the homogeneous garnet-structure spin configurations in the that are allowed by exchange interaction.

### SYMMETRY OF EXCHANGE HAMILTONIAN

The elementary cubic cell of garnet contains eight formula units  $A_2D_3C_3O_{12}$  (A, D, C—metallic ions) and is described in the paramagnetic phase by the space group  $Ia3d-O_h^{10}$ . The oxygen ions form a body-centered cubic lattice, and the metallic ions are located in the interstices. There are three types of crystallographically nonequivalent interstices<sup>16</sup>: octahedral (a), 24 tetrahedral 24 (d), and 24 dodecahedral (c). The coordinates of the atoms in positions (a), (d), and (c) are given in Table I. (Since the cell is body-centered, the positions of the remaining atoms in the elementary cubic cell are obtained by translation through the vector  $(1/2, 1/2, 1/2)$ ).

On going from the paramagnetic state to the magnetically ordered state, the symmetry of the crystal lattice may change, but the deformations are weak and can be taken into account within the framework of perturbation theory. In the first stage of the calculations we can therefore exclude the magnetostriction interaction and assume that in the magnetically ordered state the lattice is described by the space group  $O_h^{10}$  of the paramagnetic phase.

In this paper we are interested only in homogeneous states of the system, i.e., states in which the directions of the atomic spins in translationally equivalent sites coincide. The symmetry group G of the Hamil-

tonian of such a system is obtained from the space group by identifying all the trivial translations with a single element (we assume also that the magnetic and crystallographic cells coincide). The Hamiltonian of the system is a function of the directions of the spin vectors of all the magnetic atoms inside the unit cell:

$$S_{\nu a}, S_{\mu d}, S_{\eta c}, \quad (1)$$

where the indices  $\nu, \mu,$  and  $\eta$  number respectively the atoms inside the sublattices (a), (d), and (c):  $\nu = 1, 2, \dots, 8; \mu = 1, 2, \dots, 12; \eta = 1, 2, \dots, 12$ .

Any symmetry operation  $T_g$  from the group G can be represented as a definite permutation of the atoms  $P_g$  and rotation of the atomic spins<sup>11</sup>  $O_g$ , i.e.,

$$T_g = P_g O_g, \quad (2)$$

wherein only atoms from crystallographically equivalent positions are permuted with one another.

We now consider the exchange Hamiltonian  $\mathcal{H}_e$  of our system. Being a function of scalar products of atomic spins,  $\mathcal{H}_e$  is invariant to rotations in spin space. Consequently the operations  $T_g$  transform the Hamiltonian  $\mathcal{H}_e$  in the following manner:

$$\mathcal{H}_e \rightarrow \mathcal{H}_e' = (P_g^{-1} O_g^{-1}) \mathcal{H}_e (O_g P_g) = P_g^{-1} \mathcal{H}_e P_g. \quad (3)$$

In other words, an analysis of the symmetry of the exchange Hamiltonian reduces to an analysis of the symmetry relative only to permutations of the atoms  $P_g$  (we designate the corresponding group by  $\mathcal{P}$ ). Since each operation  $T_g$  from the group G corresponds to an operation  $P_g$  from  $\mathcal{P}$  and, as it turns out, none of the permutations  $P_g$  (with exception of the unitary one) leaves all the atoms in their places, it follows that the group  $\mathcal{P}$  is isomorphic to the group G, which in turn is isomorphic to the point group  $O_h$ . Consequently, it is possible to classify the magnetic structures in the present case in accordance with the irreducible representations of the group  $O_h$ —the crystal class of the lattice.

All the foregoing can be explained also in a somewhat different manner. We denote by  $U$  the group of three-dimensional rotations in spin space, and by  $R$  the operation of time reversal, which is the operation of inversion of the spin space. Since the exchange Hamiltonian is invariant to the groups  $G, U,$  and  $R,$  and since the operators  $O_g$  form a subgroup of group  $U,$  it is clear that the complete symmetry group of the exchange Hamiltonian  $\mathcal{T}$  can be represented as a direct product of the groups  $\mathcal{P}, U,$  and  $R$ :

$$\mathcal{T} = \mathcal{P} \times U \times R.$$

The possible magnetic configurations can be classified also with the aid of irreducible representations of the group  $\mathcal{T}$ , which in turn are direct products of the irreducible representations of  $\mathcal{P}$  and  $U \times R$ . It is clear that any vector in spin space is transformed in accordance with the vector representation  $Y_1^m$  of the group of three-dimensional rotations, meaning in practice that the direction of this vector in space is specified. Thus, to determine the symmetry of the magnetic ordering in the exchange approximation it suffices to indicate only the irreducible representation of the group  $\mathcal{P}$  in ac-

TABLE I. Coordinates of magnetic atoms in garnet structure (in fractions of the unit cell)

N	(a)	(d)	(c)	N	(a)	(d)	(c)
1	0, 0, 0	0, 1/4, 3/8	0, 1/4, 1/8	7	3/4, 3/4, 1/4	0, 1/4, 7/8	0, 1/4, 7/8
2	1/2, 1/2, 0	3/4, 0, 1/4	1/8, 0, 1/4	8	1/4, 3/4, 3/4	7/8, 0, 1/4	3/8, 0, 1/4
3	0, 1/4, 1/2	1/4, 3/8, 0	1/4, 1/8, 0	9		1/4, 7/8, 0	1/4, 7/8, 0
4	1/2, 0, 1/2	0, 3/4, 3/8	0, 3/4, 3/8	10		0, 3/4, 3/8	0, 3/4, 3/8
5	1/4, 1/4, 1/4	1/8, 0, 3/4	3/8, 0, 3/4	11		3/8, 0, 3/4	1/8, 0, 3/4
6	3/4, 1/4, 3/4	3/4, 1/8, 0	3/4, 3/8, 0	12		3/4, 3/8, 0	3/4, 3/8, 0

cordance with which the coordinates of the magnetic atoms are transformed.<sup>1)</sup>

Under the action of the operations  $P_g$ , the vectors (1) are transformed in the following manner:

$$P_g S_{va} = \sum_{v'} P_{vv'}^{(a)}(g^{-1}) S_{v'a}, \quad P_g S_{\mu d} = \sum_{\mu'} P_{\mu\mu'}^{(d)}(g^{-1}) S_{\mu'd},$$

$$P_g S_{\eta c} = \sum_{\eta'} P_{\eta\eta'}^{(c)}(g^{-1}) S_{\eta'c}. \quad (4)$$

The matrices  $P$  form a representation, constructed on the vectors (1), of the group  $\mathcal{P}$ . This representation is reducible and can be expanded into irreducible parts, an important factor being that it is possible to reduce separately the representations for the sublattices ( $a$ ), ( $d$ ), and ( $c$ ). We denote the total exchange representation by the letter  $\Gamma$ , and the representations for the sublattices by the letters  $\Gamma_a$ ,  $\Gamma_d$ , and  $\Gamma_c$ . By virtue of the foregoing we have

$$\Gamma = \Gamma_a + \Gamma_d + \Gamma_c. \quad (5)$$

It is known that the generating elements of the symmetry group  $O_h^{10}$  are three mutually perpendicular four-fold screw axes, which correspond in our approach to three permutation operators  $P_4^{(x)}$ ,  $P_4^{(y)}$ ,  $P_4^{(z)}$ , and to an inversion operation, which corresponds to the permutation  $P_i$ . The action of these four operators is illustrated in Table II. The representations  $\Gamma_a$ ,  $\Gamma_d$ , and  $\Gamma_c$  can be expanded in irreducible representations of  $O_h$ . Using the data of Table II, we can show that

$$\begin{aligned} \Gamma_a &= \Gamma_1 + \Gamma_2 + \Gamma_3 + \Gamma_5, \\ \Gamma_d &= \Gamma_1 + \Gamma_1' + \Gamma_3 + \Gamma_3' + \Gamma_5 + \Gamma_5', \\ \Gamma_c &= \Gamma_1 + \Gamma_1' + \Gamma_3 + \Gamma_3' + \Gamma_5 + \Gamma_5'. \end{aligned} \quad (6)$$

We use here the standard Bethe notation for the irreducible representations of the point group  $O_h$  (Ref. 12) and, as usual,  $\Gamma$  denotes an even representation and  $\Gamma'$  an odd one. The representations  $\Gamma_1$  and  $\Gamma_2$  are one-dimensional ( $\Gamma_1$ -identity),  $\Gamma_3$  is two-dimensional, and  $\Gamma_4$  and  $\Gamma_5$  are three-dimensional ( $\Gamma_4$  is the  $x, y, z$  vector representation).

The reduction (6) is effected in standard fashion by changing over from the basis made up of the vectors  $S_{va}$ ,  $S_{\mu d}$ ,  $S_{\eta c}$  to their linear combinations that transform in accordance with irreducible representations of the

TABLE II. Action of the permutation operators  $P_g$  on the positions of the magnetic atoms

Octahedral positions (a)										
$E$	1	2	3	4	5	6	7	8		
$P_4^{(x)}$	7	8	6	5	3	4	2	1		
$P_4^{(y)}$	8	5	6	7	4	1	2	3		
$P_4^{(z)}$	6	8	5	7	2	4	1	3		
$P_i$	1	2	3	4	5	6	7	8		

  

Tetrahedral positions (d)												
$E$	1	2	3	4	5	6	7	8	9	10	11	12
$P_4^{(x)}$	9	11	10	12	8	7	3	5	4	6	2	1
$P_4^{(y)}$	11	7	12	8	10	9	5	1	6	2	4	3
$P_4^{(z)}$	10	12	8	7	9	11	4	6	2	1	3	5
$P_i$	10	11	12	7	8	9	4	5	6	1	2	3

  

Dodecahedral positions (c)												
$E$	1	2	3	4	5	6	7	8	9	10	11	12
$P_4^{(x)}$	3	5	4	6	2	1	9	11	10	12	8	7
$P_4^{(y)}$	5	1	6	2	4	3	11	7	12	8	10	9
$P_4^{(z)}$	4	6	2	1	3	5	10	12	8	7	9	11
$P_i$	10	11	12	7	8	9	4	5	6	1	2	3

group  $O_h$ . We shall denote these combinations by the letters  $A_\nu, D_\mu, C_\eta$ . To obtain them we must apply the projection operator

$$Q_{\nu\alpha}^{(\alpha)} = \sum_g D_{\nu\alpha}^{(\alpha)}(g) P_g \quad (7)$$

to an arbitrary vector of the old basis.<sup>12</sup> Here  $\alpha$  is the number of the irreducible representation,  $D_{\nu\alpha}^{(\alpha)}(g)$  are the matrices of the irreducible representations of the group  $O_h$ , and the action of the operator  $P_g$  is given in Table II. The sum in (7) is taken over all the 48 operations of the group  $O_h$ . As a result of the action of the operator (7) we obtain the following irreducible vectors:<sup>2)</sup>

$$A_\nu = \sum_{v'} U_{vv'}^{(\alpha)} S_{v'a}, \quad D_\mu = \sum_{\mu'} U_{\mu\mu'}^{(d)} S_{\mu'd},$$

$$C_\eta = \sum_{\eta'} U_{\eta\eta'}^{(c)} S_{\eta'c}. \quad (8)$$

The expressions for the matrices  $\hat{U}$  are given in Table III, which gives also the classification of the vectors  $A$ ,  $D$ , and  $C$  in accordance with the irreducible representations. For convenience we have not normalized the irreducible vectors, so that the matrices  $\hat{U}$  are not unitary. The transformation inverse to (8) is of the form

$$S_{va} = \sum_{\nu'} U_{\nu v}^{(\alpha)} \frac{A_{\nu'}}{\zeta_{\nu'}}, \quad S_{\mu d} = \sum_{\mu'} U_{\mu\mu'}^{(d)} D_{\mu'}. \quad (9)$$

Using (6) and the data of Table III, we can find and classify by symmetry types of magnetic ordering of the ions in different sublattices. We shall say that a system is in a pure  $\Gamma_\alpha$  state if only vectors that transform in accordance with  $\alpha$ -th irreducible representation differ from zero.

TABLE III. Matrix of transition to the irreducible vectors

Octahedral positions (a)												
$\Gamma_1$	1	1	1	1	1	1	1	1	1	1	1	1
$\Gamma_2$	1	1	1	1	-1	-1	-1	-1	-1	-1	-1	-1
$\Gamma_3$	1	1	-1	-1	1	1	-1	-1	1	1	-1	-1
$\Gamma_4$	1	-1	1	-1	1	-1	1	-1	1	-1	1	-1
$\Gamma_5$	1	-1	-1	1	1	-1	-1	1	-1	1	1	-1
$\Gamma_6$	1	-1	-1	-1	-1	1	1	-1	-1	1	1	-1
$(x, y, z)$	1	-1	-1	-1	1	-1	1	-1	1	-1	1	-1

  

Tetrahedral positions (d), $\gamma = \sqrt{3}$												
$\Gamma_1$	1	1	1	1	1	1	1	1	1	1	1	1
$\Gamma_2$	0	1	-1	0	1	-1	0	1	-1	0	1	-1
$\Gamma_3$	2	-1	-1	2	-1	-1	2	-1	-1	2	-1	-1
$\Gamma_4$	0	1	0	0	-1	0	0	-1	0	0	1	0
$\Gamma_5$	0	0	1	0	0	-1	0	0	-1	0	0	1
$\Gamma_6$	1	0	0	-1	0	0	-1	0	0	1	0	0
$\Gamma_7$	1	1	1	1	1	1	-1	-1	-1	-1	-1	-1
$\Gamma_8$	2	-1	-1	2	-1	-1	-2	1	1	-2	1	1
$\Gamma_9$	0	-1	1	0	-1	1	0	1	-1	0	1	-1
$\Gamma_{10}$	0	1	0	0	-1	0	0	1	0	0	-1	0
$\Gamma_{11}$	0	0	1	0	0	-1	0	0	1	0	0	-1
$\Gamma_{12}$	1	0	0	-1	0	0	1	0	0	-1	0	0

  

Dodecahedral positions (c)												
$\Gamma_1$	1	1	1	1	1	1	1	1	1	1	1	1
$\Gamma_2$	0	1	-1	0	1	-1	0	1	-1	0	1	-1
$\Gamma_3$	2	-1	-1	2	-1	-1	2	-1	-1	2	-1	-1
$\Gamma_4$	0	1	0	0	-1	0	0	-1	0	0	1	0
$\Gamma_5$	0	0	1	0	0	-1	0	0	-1	0	0	1
$\Gamma_6$	1	0	0	-1	0	0	-1	0	0	1	0	0
$\Gamma_7$	1	1	1	1	1	1	-1	-1	-1	-1	-1	-1
$\Gamma_8$	0	1	-1	0	1	-1	0	1	-1	0	1	-1
$\Gamma_9$	2	-1	-1	2	-1	-1	-2	1	1	-2	1	1
$\Gamma_{10}$	0	1	0	0	-1	0	0	1	0	0	-1	0
$\Gamma_{11}$	0	0	1	0	0	-1	0	0	1	0	0	-1
$\Gamma_{12}$	1	0	0	-1	0	0	1	0	0	-1	0	0

It is seen from (6) that four pure states are possible in an octahedral sublattice and six different states each are possible in a tetrahedral and dodecahedral sublattice. We consider them separately.

## MAGNETIC ORDERING IN OCTAHEDRA

1. Configuration  $\Gamma_1$ —ferromagnetic ordering, or spins directed to the same side.

2. Configuration  $\Gamma_2$ —collinear antiferromagnetic ordering, with

$$S_{1a}=S_{2a}=S_{3a}=S_{4a}=-S_{5a}=-S_{6a}=-S_{7a}=-S_{8a}. \quad (10)$$

3. Configuration  $\Gamma_4$ —antiferromagnetic ordering. In this case the following relations hold between the spins:

$$\begin{aligned} S_{3a}=-S_{1a}, \quad S_{6a}=-S_{2a}, \quad S_{7a}=-S_{3a}, \quad S_{8a}=-S_{4a}, \\ S_{1a}+S_{2a}+S_{3a}+S_{4a}=0, \end{aligned} \quad (11)$$

i.e., the symmetry admits of a definite leeway in the disposition of the spins 1, 2, 3, 4; the only requirement is that their total angular momentum be equal to zero. To eliminate the leeway it is necessary to resort to some concrete expression for the exchange energy of the system. We shall use it somewhat later.

4. Configuration  $\Gamma_5$ —antiferromagnetic ordering. The relations between the spins are

$$\begin{aligned} S_{3a}=S_{1a}, \quad S_{6a}=S_{2a}, \quad S_{7a}=S_{3a}, \quad S_{8a}=S_{4a}, \\ S_{1a}+S_{2a}+S_{3a}+S_{4a}=0. \end{aligned} \quad (12)$$

Just as in the preceding case, the relative disposition of the spins 1, 2, 3, and 4 is not completely fixed.

To choose between the different configurations we must compare their exchange energy. We consider a Heisenberg Hamiltonian of a garnet with magnetic atoms in the sublattice (a):

$$\mathcal{H} = \frac{1}{2} \sum_{\nu, \nu'} \sum_{f, f'} J_{\nu\nu'}^{(a)} (\mathbf{R}_f - \mathbf{R}_{f'}) S_{\nu a}(\mathbf{R}_f) S_{\nu' a}(\mathbf{R}_{f'}); \quad (13)$$

the index  $f$  numbers here the unit cells. Since we are interested in the energy of the homogeneous state, we obtain for the energy per unit cell of the crystal, putting  $S_{\nu a}(\mathbf{R}_f) = S_{\nu a}(\mathbf{R}_{f'}) \equiv S_{\nu a}$  in (13),

$$\mathcal{H}^{(a)} = \frac{1}{2} \sum_{\nu, \nu'} J_{\nu\nu'}^{(a)} S_{\nu a} S_{\nu' a}, \quad (14)$$

$$J_{\nu, \nu'}^{(a)} = \sum_{f, f'} J_{\nu\nu'}^{(a)} (\mathbf{R}_f - \mathbf{R}_{f'}), \quad (15)$$

where  $J_{\nu, \nu'}^{(a)}$  is the energy of the interaction of the atom  $\nu$  with all the atoms  $\nu'$  from all the cells.

Not all the exchange interactions in (15) are independent. It can be shown, by using Table II, that there are only four independent parameters:

$$\begin{aligned} J_1^{(a)} \equiv J_{1,3}^{(a)}, \quad J_2^{(a)} \equiv J_{1,2}^{(a)} = J_{1,3}^{(a)} = J_{1,4}^{(a)}, \\ J_3^{(a)} \equiv J_{1,5}^{(a)} = J_{1,7}^{(a)} = J_{1,8}^{(a)}, \quad J_4^{(a)} \equiv J_{1,4}^{(a)}. \end{aligned} \quad (16)$$

Changing from the vectors  $S_{\nu a}$  to the irreducible vectors  $\mathbf{A}_\nu$  with the aid of (9), we get

$$\mathcal{H}^{(a)} = \frac{1}{2} \lambda_{1a} \mathbf{A}_1^2 + \frac{1}{2} \lambda_{2a} \mathbf{A}_2^2 + \frac{1}{2} \lambda_{3a} (\mathbf{A}_5^2 + \mathbf{A}_7^2 + \mathbf{A}_8^2) + \frac{1}{2} \lambda_{4a} (\mathbf{A}_3^2 + \mathbf{A}_4^2 + \mathbf{A}_5^2), \quad (17)$$

where the coefficients  $\lambda_a$  are connected with the exchange integrals (16) by the following relations:

$$\begin{aligned} \lambda_{1a} &= \frac{1}{8} [J_1^{(a)} + 3J_2^{(a)} + 3J_3^{(a)} + J_4^{(a)}], \\ \lambda_{2a} &= -\frac{1}{8} [J_1^{(a)} - 3J_2^{(a)} + 3J_3^{(a)} - J_4^{(a)}], \\ \lambda_{3a} &= -\frac{1}{8} [J_1^{(a)} + J_2^{(a)} - J_3^{(a)} - J_4^{(a)}], \quad \lambda_{4a} = \frac{1}{8} [J_1^{(a)} - J_2^{(a)} - J_3^{(a)} + J_4^{(a)}]. \end{aligned} \quad (18)$$

It is now easy to obtain the energies of the different configurations:

$$\begin{aligned} \mathcal{H}_2(\Gamma_1) &= 32\lambda_{1a} S_a^2, \quad \mathcal{H}_2(\Gamma_2) = 32\lambda_{2a} S_a^2, \\ \mathcal{H}_2(\Gamma_4) &= 32\lambda_{3a} S_a^2, \quad \mathcal{H}_2(\Gamma_5) = 32\lambda_{4a} S_a^2, \end{aligned} \quad (19)$$

where  $S_a$  is the ion spin in the position (a). Thus, in the approximation quadratic in the spins, both the configuration  $\Gamma_4$  and the configuration  $\Gamma_5$  are degenerate [their energies are independent of the different spin-configuration deformations that are allowed by relations (11) and (12)].

To find the relative arrangement of the atoms, bi-quadratic exchange must be added to the Hamiltonian (14). Terms of fourth order in the vectors  $\mathbf{A}_\nu$  appear then in (17). We shall not write them out, and present only those terms which influence the energy of the pure configurations  $\Gamma_4$  and  $\Gamma_5$  and depend on the relative arrangement of the spins:

$$\mathcal{H}_4^{(a)} = L_{4a} (\mathbf{A}_6^4 + \mathbf{A}_7^4 + \mathbf{A}_8^4) + L_{5a} (\mathbf{A}_3^4 + \mathbf{A}_4^4 + \mathbf{A}_5^4). \quad (20)$$

For the configuration  $\Gamma_4$  we obtain, taking (11) into account,

$$\begin{aligned} \mathcal{H}_4(\Gamma_4) &= 2^8 L_{4a} \{2S_a^4 + (S_{1a}S_{2a})^2 + (S_{1a}S_{3a})^2 \\ &+ (S_{1a}S_{4a})^2 + (S_{2a}S_{3a})^2 + (S_{2a}S_{4a})^2 + (S_{3a}S_{4a})^2\}. \end{aligned} \quad (21)$$

If  $L_{4a}$  is negative, then the energy minimum corresponds to three collinear configurations:

$$\begin{aligned} \Gamma_4^{(1)}: S_{1a}=S_{2a}=-S_{3a}=-S_{4a}, \quad \mathbf{A}_6=4S_{1a}, \quad \mathbf{A}_7=\mathbf{A}_8=0; \\ \Gamma_4^{(2)}: S_{1a}=-S_{2a}=S_{3a}=-S_{4a}, \quad \mathbf{A}_7=4S_{1a}, \quad \mathbf{A}_6=\mathbf{A}_8=0; \\ \Gamma_4^{(3)}: S_{1a}=-S_{2a}=-S_{3a}=S_{4a}, \quad \mathbf{A}_8=4S_{1a}, \quad \mathbf{A}_6=\mathbf{A}_7=0; \\ \mathcal{H}_4^{(a)}(\Gamma_4^{(1,2,3)}) = 2^{12} L_{4a} S_a^4. \end{aligned} \quad (22)$$

On the other hand if  $L_{4a}$  is positive, then the spins are directed along the four different body diagonals of the cube, and furthermore in such a way that the condition (11) is satisfied. It is easily seen that two nonequivalent spin arrangements exist here:  $\Gamma_4^{(4)}$  and  $\Gamma_4^{(5)}$ , with

$$\mathcal{H}_4^{(a)}(\Gamma_4^{(4,5)}) = \frac{1}{2} 2^{12} L_{4a} S_a^4.$$

In both configurations, the vectors  $\mathbf{A}_6$ ,  $\mathbf{A}_7$ , and  $\mathbf{A}_8$  are equal to one another in magnitude and are mutually perpendicular.

For the configuration  $\Gamma_5$  we obtain an expression which is perfectly analogous to (21), with  $L_{4a}$  replaced by  $L_{5a}$ . We then obtain either the three collinear possibilities (22) or the two three-dimensional ones, but with allowance for (12). Only one irreducible vector,  $\mathbf{A}_3$  or  $\mathbf{A}_4$  or  $\mathbf{A}_5$ , respectively, differs then from zero in the configurations  $\Gamma_5^{(1)}$ ,  $\Gamma_5^{(2)}$ ,  $\Gamma_5^{(3)}$ , while in configurations  $\Gamma_5^{(4)}$  and  $\Gamma_5^{(5)}$  all the three vectors  $\mathbf{A}_3$ ,  $\mathbf{A}_4$ , and  $\mathbf{A}_5$  are equal in magnitude and mutually perpendicular. But all these structures are compatible in symmetry with the group  $O_h$ . This means that allowance for the magnetostriction interaction for such structures leads to a distortion of the lattice and to a lowering of its symmetry. To obtain these distortions we shall use

the ideas of Andreev and Marchenko,<sup>10</sup> namely that the lattice symmetry in the magnetically ordered state should coincide with the symmetry of any spin scalar. Consequently, the lattice symmetry will be determined by the symmetry of quantities of the type  $A_{\nu}$ ,  $A_{\nu'}$ , set up for the given configuration.

As already mentioned, the vectors  $A_{\nu}$  play the same role as the multipole moments introduced in Ref. 10. This means that if the transition to the magnetically ordered state occurs without a change in the lattice symmetry, the following relations should be satisfied

$$A_{\nu} A_{\nu'} = \xi_{\alpha} \delta_{\alpha\beta} \delta_{\nu\nu'}, \quad (23)$$

where  $\alpha$  and  $\beta$  are the numbers of the irreducible representations in accordance with which the vectors  $A_{\nu}$  and  $A_{\nu'}$  transform, and  $\xi_{\alpha}$  are certain constants. It can be directly verified that the configurations  $\Gamma_1, \Gamma_2, \Gamma_4^{(4,5)}, \Gamma_5^{(4,5)}$  satisfy relations (23), i.e., a phase transition to these configurations is not accompanied by lattice distortions. The symmetry of these configurations is determined by the exchange classes  $A_{1g}, A_{2g}, F_{1g}, F_{2g}$  of the  $O_h$  group from Ref. 10.

In the configurations  $\Gamma_4^{(1,2,3)}$  and  $\Gamma_5^{(1,2,3)}$  the spin scalars have the symmetry  $D_{4h}$ , meaning that the lattice has the same symmetry. These spin configurations belong to the exchange classes  $A_{2g}$  and  $B_{1g}$  of the group  $D_{4h}$ .

## MAGNETIC ORDERING IN TETRAHEDRA

1. Configuration  $\Gamma_1$ —ferromagnetic ordering.

2. Configuration  $\Gamma_2$ —collinear antiferromagnetic ordering:

$$\begin{aligned} S_{1d} = S_{2d} = S_{3d} = S_{4d} = S_{5d} = S_{6d} \\ = -S_{7d} = -S_{8d} = -S_{9d} = -S_{10d} = -S_{11d} = -S_{12d}. \end{aligned} \quad (24)$$

3. Configuration  $\Gamma_3$ —triangular ordering:

$$\begin{aligned} S_{10d} = S_{7d} = S_{4d} = S_{1d}, \quad S_{11d} = S_{8d} = S_{3d} = S_{2d}, \\ S_{12d} = S_{9d} = S_{6d} = S_{5d}, \quad S_{1d} + S_{2d} + S_{3d} = 0. \end{aligned} \quad (25)$$

4. Configuration  $\Gamma_3'$ —triangular ordering:

$$\begin{aligned} S_{10d} = S_{7d} = -S_{4d} = -S_{1d}, \quad S_{11d} = S_{8d} = -S_{3d} = -S_{2d}, \\ S_{12d} = S_{9d} = -S_{6d} = -S_{5d}, \quad S_{1d} + S_{2d} + S_{3d} = 0. \end{aligned} \quad (26)$$

5. Configuration  $\Gamma_4$ —three antiferromagnetic sublattices:

$$\begin{aligned} S_{10d} = -S_{7d} = -S_{4d} = S_{1d}, \quad D_6 = 4S_{1d}, \\ S_{11d} = -S_{8d} = -S_{3d} = S_{2d}, \quad D_4 = 4S_{2d}, \\ S_{12d} = -S_{9d} = -S_{6d} = S_{5d}, \quad D_5 = 4S_{3d} \end{aligned} \quad (27)$$

with arbitrary directions of the antiferromagnetism axes relative to one another. Just as in the configurations  $\Gamma_4$  and  $\Gamma_5$  from the sublattice (a), the quadratic spin Hamiltonian does not fix the relative positions of these axes.

Inclusion of biquadratic exchange leads to the possible existence of either the collinear structures

$$\begin{aligned} \Gamma_4^{(1)}: S_{1d} = S_{2d} = S_{3d}, \\ \Gamma_4^{(2)}: S_{1d} = S_{2d} = -S_{3d}, \\ \Gamma_4^{(3)}: S_{1d} = -S_{2d} = S_{3d}, \\ \Gamma_4^{(4)}: S_{1d} = -S_{2d} = -S_{3d}, \end{aligned} \quad (28)$$

for which all three vectors  $D_4, D_5, D_6$  are collinear or two three-dimensional structures  $\Gamma_4^{(5)}, \Gamma_4^{(6)}$ , in which all the three antiferromagnetism vectors are perpendicular to one another:  $D_4 \perp D_5 \perp D_6$  and are equal in magnitude.

6. Configuration  $\Gamma_5'$ —three antiferromagnetic sublattices:

$$\begin{aligned} S_{10d} = -S_{7d} = S_{4d} = -S_{1d}, \quad D_{12} = 4S_{1d}, \\ S_{11d} = -S_{8d} = S_{3d} = -S_{2d}, \quad D_{10} = 4S_{2d}, \\ S_{12d} = -S_{9d} = S_{6d} = -S_{5d}, \quad D_{11} = 4S_{3d}, \end{aligned} \quad (29)$$

which are subject to everything applicable to configuration  $\Gamma_4$ .

The exchange energy per cell for the ( $d$ ) ions is given by

$$\mathcal{H}^{(d)} = \frac{1}{2} \sum_{\mu, \mu'} J_{\mu, \mu'}^{(d)} S_{\mu d} S_{\mu' d}, \quad (30)$$

and there are six different exchange parameters

$$\begin{aligned} J^{(d)} = J_{1,1}^{(d)}, \quad J_1^{(d)} = J_{1,2}^{(d)} = J_{1,3}^{(d)} = J_{1,5}^{(d)} = J_{1,6}^{(d)}, \\ J_2^{(d)} = J_{1,4}^{(d)}, \quad J_3^{(d)} = J_{1,8}^{(d)} = J_{1,9}^{(d)} = J_{1,11}^{(d)} = J_{1,12}^{(d)}, \\ J_4^{(d)} = J_{1,7}^{(d)}, \quad J_5^{(d)} = J_{1,10}^{(d)}. \end{aligned} \quad (31)$$

Changing over to the irreducible vectors  $D_{\mu}$ , we obtain

$$\begin{aligned} \mathcal{H}^{(d)} = \frac{1}{2} \lambda_{1d} D_1^2 + \frac{1}{2} \lambda_{2d} D_2^2 + \frac{1}{2} \lambda_{3d} (D_2^2 + D_3^2) \\ + \frac{1}{2} \lambda_{4d} (D_6^2 + D_9^2) + \frac{1}{2} \lambda_{5d} (D_4^2 + D_5^2 + D_6^2) \\ + \frac{1}{2} \lambda_{6d} (D_{10}^2 + D_{11}^2 + D_{12}^2), \end{aligned} \quad (32)$$

and the constants  $\lambda_{\mu}$  take the following form

$$\begin{aligned} \lambda_{1d} = \frac{1}{12} [4J_1^{(d)} + J_2^{(d)} + 4J_3^{(d)} + J_4^{(d)} + J_5^{(d)} + J^{(d)}], \\ \lambda_{2d} = \frac{1}{12} [4J_1^{(d)} + J_2^{(d)} - 4J_3^{(d)} - J_4^{(d)} - J_5^{(d)} + J^{(d)}], \\ \lambda_{3d} = -\frac{1}{24} [2J_1^{(d)} - J_2^{(d)} + 2J_3^{(d)} - J_4^{(d)} - J_5^{(d)} - J^{(d)}], \\ \lambda_{4d} = -\frac{1}{24} [2J_1^{(d)} - J_2^{(d)} - 2J_3^{(d)} + J_4^{(d)} + J_5^{(d)} - J^{(d)}], \\ \lambda_{5d} = -\frac{1}{4} [J_2^{(d)} + J_4^{(d)} - J_5^{(d)} - J^{(d)}], \\ \lambda_{6d} = -\frac{1}{4} [J_2^{(d)} - J_4^{(d)} + J_5^{(d)} - J^{(d)}]. \end{aligned} \quad (33)$$

It is possible to obtain the energy of the system in the different configurations:

$$\begin{aligned} \mathcal{H}^{(d)}(\Gamma_1) = 72\lambda_{1d} S_d^2, \quad \mathcal{H}^{(d)}(\Gamma_2) = 72\lambda_{2d} S_d^2, \\ \mathcal{H}^{(d)}(\Gamma_3) = 144\lambda_{3d} S_d^2, \quad \mathcal{H}^{(d)}(\Gamma_3') = 144\lambda_{3d} S_d^2, \\ \mathcal{H}^{(d)}(\Gamma_4) = 24\lambda_{4d} S_d^2, \quad \mathcal{H}^{(d)}(\Gamma_5') = 24\lambda_{6d} S_d^2. \end{aligned} \quad (34)$$

To lift the degeneracy in configurations in  $\Gamma_4$  and  $\Gamma_5'$  it is necessary to add to the Hamiltonian (32) a biquadratic exchange in the form

$$\begin{aligned} \mathcal{H}_i^{(d)} = L_i^{(d)} [(D_1 D_2)^2 + (D_1 D_3)^2 + (D_2 D_3)^2] \\ + L_5^{(d)} [(D_{10} D_{11})^2 + (D_{11} D_{12})^2 + (D_{10} D_{12})^2]. \end{aligned} \quad (35)$$

We then obtain

$$\begin{aligned} \mathcal{H}_i^{(d)}(\Gamma_4^{(1,2,3,4)}) = 3 \cdot 2^8 L_i^{(d)} S_d^4, \quad \mathcal{H}_i^{(d)}(\Gamma_4^{(5,6)}) = 0, \\ \mathcal{H}_i^{(d)}(\Gamma_5^{(1,2,3,4)}) = 3 \cdot 2^8 L_5^{(d)} S_d^4, \quad \mathcal{H}_i^{(d)}(\Gamma_5^{(5,6)}) = 0. \end{aligned} \quad (36)$$

Just as for the octahedral sublattice, it can be shown that the configurations  $\Gamma_1, \Gamma_2, \Gamma_3, \Gamma_3', \Gamma_4^{(5,6)}, \Gamma_5^{(5,6)}$  do not distort the crystal lattice and are described by the exchange classes  $A_{1g}, A_{2u}, E_g, E_u, F_{1g}, F_{2u}$  of group  $O_h$ , while the configurations  $\Gamma_4^{(1,2,3,4)}$  and  $\Gamma_5^{(1,2,3,4)}$  cause trigonal distortions of the lattice and are described by exchange classes  $A_{2g}$  and  $A_{1u}$  of the group  $D_{3d}$ .

## MAGNETIC ORDERING IN DODECAHEDRA

The possible types of ordering in the dodecahedral sublattice are analogous in many respects to the ordering in the tetragonal sublattice, and we present therefore only a very brief description of these structures with references to the preceding section.

Configuration  $\Gamma_1$ —ferromagnetic ordering.

Configuration  $\Gamma'_1$ —the spin arrangement is described by Eq. (24).

Configuration  $\Gamma_3$ —Eq. (25).

Configuration  $\Gamma'_3$ —Eq. (26).

Configuration  $\Gamma_5$ —Eqs. (27) and (28).

Configuration  $\Gamma'_5$ —Eq. (29).

The exchange energy for the cell for the (c) ions is

$$\mathcal{H}^{(c)} = \frac{1}{2} \sum J_{n,n'}^{(c)} S_{nc} S_{n'c}. \quad (37)$$

The symmetry of the exchange interactions  $J_{n,n'}^{(c)}$  is determined by relations (31).

Changing over in (37) to irreducible vectors, we get

$$\begin{aligned} \mathcal{H}^{(c)} = & \frac{1}{2} \lambda_{1c} C_1^2 + \frac{1}{2} \lambda_{1c} C_7^2 + \frac{1}{2} \lambda_{3c} (C_2^2 + C_3^2) \\ & + \frac{1}{2} \lambda_{3c} (C_8^2 + C_9^2) + \frac{1}{2} \lambda_{5c} (C_4^2 + C_5^2 + C_6^2) \\ & + \frac{1}{2} \lambda_{5c} (C_{10}^2 + C_{11}^2 + C_{12}^2). \end{aligned} \quad (38)$$

The constants  $\lambda_c$  are determined by relations (33), in which we must make the substitutions

$$\lambda_{1d} \rightarrow \lambda_{1c}, \quad \lambda_{2d} \rightarrow \lambda_{1c}, \quad \lambda_{3d} \rightarrow \lambda_{3c}, \quad (39)$$

$$\lambda_{3d} \rightarrow \lambda_{3c}, \quad \lambda_{4d} \rightarrow \lambda_{3c}, \quad \lambda_{5d} \rightarrow \lambda_{5c}, \quad J_V^{(d)} \rightarrow J_V^{(c)}.$$

The energy of the system in the various configurations is determined by the expressions

$$\begin{aligned} \mathcal{H}^{(c)}(\Gamma_1) &= 72\lambda_{1c} S_c^2, & \mathcal{H}^{(c)}(\Gamma'_1) &= 72\lambda_{1c} S_c^2, \\ \mathcal{H}^{(c)}(\Gamma_3) &= 144\lambda_{3c} S_c^2, & \mathcal{H}^{(c)}(\Gamma'_3) &= 144\lambda_{3c} S_c^2, \\ \mathcal{H}^{(c)}(\Gamma_5) &= 24\lambda_{5c} S_c^2, & \mathcal{H}^{(c)}(\Gamma'_5) &= 24\lambda_{5c} S_c^2. \end{aligned} \quad (40)$$

That part of the fourth Hamiltonian which stabilizes the configurations  $\Gamma_5$  and  $\Gamma'_5$  is given by

$$\begin{aligned} \mathcal{H}_4^{(c)} &= L_5^{(c)} [(C_1 C_2)^2 + (C_3 C_4)^2 + (C_5 C_6)^2] \\ &+ \bar{L}_5^{(c)} [(C_{10} C_{11})^2 + (C_{11} C_{12})^2 + (C_{12} C_{10})^2]. \end{aligned} \quad (41)$$

We then obtain

$$\begin{aligned} \mathcal{H}_4^{(c)}(\Gamma_5^{(1,2,3,4)}) &= 3 \cdot 2^8 L_5^{(c)} S_c^4, & \mathcal{H}_4^{(c)}(\Gamma_5^{(5,6)}) &= 0, \\ \mathcal{H}_4^{(c)}(\Gamma'_5^{(1,2,3,4)}) &= 3 \cdot 2^8 \bar{L}_5^{(c)} S_c^4, & \mathcal{H}_4^{(c)}(\Gamma'_5^{(5,6)}) &= 0. \end{aligned} \quad (42)$$

The configurations  $\Gamma_1, \Gamma'_1, \Gamma_3, \Gamma'_3, \Gamma_5^{(5,6)}, \Gamma'_5^{(5,6)}$  do not change the lattice symmetry and are described by the exchange classes  $A_{1g}, A_{1u}, E_g, E_u, F_{2g}, F_{2u}$  of the  $O_h$  group, while the configurations  $\Gamma_5^{(1,2,3,4)}$  and  $\Gamma'_5^{(1,2,3,4)}$  cause trigonal distortions of the lattice and will be described by the exchange classes  $A_{1g}$  and  $A_{1u}$  of the  $D_{3d}$  group.

## CONCLUSION

We have analyzed in detail and obtained the energy for only pure configurations (i. e., configurations described by one of the irreducible representations of the group  $\mathcal{P}$ ). It is possible that mixed configura-

tions can also exist and can be obtained within the framework of the present analysis. We must bear in mind here the limitations of the coexistence of different configurations; these limitations were formulated by Andreev and Marchenko<sup>10</sup> [see also Eq. (23) of the present paper].

As already noted, the configurations  $\Gamma_4, \Gamma_5, \Gamma'_4, \Gamma'_5$  are not fully defined within the framework of the quadratic Hamiltonian. We have shown that this uncertainty can be partially eliminated by taking into account a biquadratic exchange interaction. The same uncertainty can be lifted also by taking into account relativistic interactions, but this is outside the scope of the present paper. Using for the irreducible vectors  $A_\nu, D_\mu, C_\eta$ , the expressions obtained in the present paper, we can examine the question of the possible types of ordering in the case when the magnetic ions occupy two or three nonequivalent positions. It is then again possible to start from the model Hamiltonian, but when account is taken of the lattice deformations and when the coexistence of various pure configurations is considered it is necessary to invoke a relation similar to (23). The difference is that now some irreducible representations are encountered several times in the expansion of the complete representation. Let  $\Gamma_{pq}^{(\alpha)}$  be an irreducible vector that transforms in accordance with the  $p$ -th line of the  $\alpha$ -th irreducible representation of the group  $\mathcal{P}$  ( $q$  is an arbitrary number that runs through as many values as there are encounters of the  $\alpha$ -th representation in the total exchange representation  $\Gamma$ ). It can then be shown that if the given spin configuration is compatible with the lattice symmetry, the following condition should be satisfied

$$\Gamma_{pq}^{(\alpha)} \Gamma_{p'q'}^{(\alpha')} = \xi_{qq'} \delta_{\alpha\alpha'} \delta_{pp'},$$

where  $\xi_{qq'}$  are certain constants.

In conclusion, the author is grateful to V. G. Bar'yakhtar and I. M. Vitebskii for numerous helpful discussions.

<sup>1</sup>We note that besides the ordinary magnetic ordering, for which the order parameter is a vector that transforms in spin space in accordance with the representation  $Y_1^n$ , the symmetry admits also of more complicated types of ordering, whose order parameter transforms in spin space with one of the representations  $Y_q^n$  of the rotation group. The consideration of these states, however, is outside the scope of the present paper.

<sup>2</sup>We note that in our approach the vectors A, D, and C play the same role as the multipole moments in the paper of Andreev and Marchenko.<sup>10</sup>

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