Spectrum of collective excitations of a ferromagnet with single-ion anisotropy in a magnetic field

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A diagram technique for spin operators is developed as applied to magnets with one-ion anisotropy. The technique permits an exact treatment of one-ion anisotropy of arbitrary symmetry in the presence of a magnetic field of arbitrary direction. The system ground state, whose type depends on the magnitude and direction of the external field, is investigated, as is also the spectrum of the collective excitations in each of the possible phases.

1. INTRODUCTION

This paper is devoted to the development of a systematic method for microscopic investigation of one-ion anisotropy (OA) at arbitrary symmetry and arbitrary value of the OA, and at arbitrary direction and magnitude of the external magnetic field. Also investigated are the ground state and the spectrum of the collective oscillations.

What distinguishes this problem?

The uniaxial problem—uniaxial OA and longitudinal field—has been quite thoroughly investigated (e.g., Refs. 1-8): the main properties of such magnets are close to the properties of an isotropic ferromagnet (FM) or of a ferromagnet with exchange anisotropy, although certain quantitative differences are present. Radical changes occur when a definite noncollinearity is present, viz., a field directed at an angle to the OA axis or a more complicated symmetry of the OA. In this case various types of ground state are possible--- nonmagnetic, collinear ferromagnetic, and canted ferromagnetic; these are realized in different field intervals that depend on the ratio of the OA and exchange constants (see Ref. 9 and Sec. 4 of the present paper). Entirely unusual properties compared with FM without OA are possessed in this case not only by the nonmagnetic phase, but also by ferromagnetic structures, since a quadrupole order is imposed in them (in the presence of a second-order OA). The latter, e.g., prevents magnetic saturation from being reached at T = 0 (Sec. 4) and leads also to other singularities. The collective-oscillation spectrum also changes. Not only the number of branches and the dispersion law, but also the character of the oscillations becomes different. These oscillations are in general not spin waves but superpositions of oscillations of spin operators and operators that are nonlinear in the spin variables (see Sec. 4).

The microscopic description of such a noncollinear case encounters a number difficulties, which were noted, for example, in Refs. 9–15. These can be avoided if there exists some small parameter, say a small transverse component of the magnetic field,¹⁰ a weak deviation of the OA symmetry from uniaxial,¹¹ or (according to chronologically the latest study¹²) an arbitrary magnetic field but a small uniaxial anisotropy.

At arbitrary values of the parameters, particularly arbitrary OA constants, the problems remains unsolved.¹⁾ Yet many known magnetic compounds, based in particular on rare-earth ions, have OA constants comparable with or even larger than the exchange integrals. The existence of a large group of substances whose diverse properties cannot be described by the existing theories was in fact the stimulus for the present study.

The approach developed is a generalization of the Vaks-Larkin-Pikin diagram technique¹⁷ as applied to magnets with OA. This technique permits accurate treatment of OA of arbitrary symmetry and magnitude in the presence of a field of arbitrary direction and magnitude. This is accomplished by expanding the basis of the operators that describe the state of an individual site, viz., by transforming from spin operators comprising the Lie SU(2) algebra to an assembly of spin, quadrupole, ..., 2S-pole operators that make up a Lie SU(2S + 1) algebra¹⁴ (S is the value of the spin in the site). In Sec. 2 is constructed for the uniaxial case a diagram technique that is convenient for further generalization, given in Sec. 3, to an arbitrary case.

In Sec. 4 are investigated all types of ground state and the spectra of the collective excitations for the case S = 1 at arbitrary temperatures, in the lowest approximation for the irreducible part.

2. DIAGRAM TECHNIQUE FOR THE UNIAXIAL CASE

Consider a ferromagnet with isotropic exchange and uniaxial OA, which is described in the presence of a longitudinal field by the Hamiltonian

$$H = H^{ex} + H^a + H^h, \tag{1}$$

$$H^{ex} = -\frac{1}{2} \sum_{ij} J_{ij} \mathbf{S}_i \mathbf{S}_j, \quad H^a = d \sum_i (S_i^z)^2, \quad H^h = -h \sum_i S_i^z.$$

Since the purpose of the formalism developed is an exact treatment of the OA, we include the one-ion Hamiltonians H^a and H^h in the zeroth Hamiltonian

$$H_{0} = -\tilde{h} \sum_{i} S_{i}^{z} + d \sum_{i} (S_{i}^{z})^{2},$$

$$\tilde{h} = h + J \langle S^{z} \rangle, \quad J = \sum_{j} J_{ij}.$$
(2)

We introduce the temperature Green's functions of the spin operators $S^{\alpha}(\tau)$ in the Heisenberg representation with imaginary time. These operators determine, as usual, the thermodynamic and kinetic characteristics of the system at finite T. We express them in the interaction representation with the zeroth Hamiltonian (2) in terms of the temperature scattering matrix, in analogy with Ref. 18. The problem of calculating the contribution of *n*-th order in H^{int} reduces then to the calculation of the mean values, with the zeroth Hamiltonian, of the *T*-product of a certain number of spin operators in the interaction representation. The calculation of mean values containing only the operators $S^{z}(\tau)$ is not a complicated task, since the latter do not depend on the time: $S^{z}(\tau) = S^{z}$. Calculation of mean values containing the off-diagonal operators $S^{\mp}(\tau)$ entails difficulties due to noncompliance with the Wick theorem for spin operators^{17,18} in the presence of OA in H_{0} , owing to the peculiar time dependence of $S^{\mp}(\tau)$ (Ref. 7):

$$S^{\mp}(\tau) = \exp\left[\left(\pm \hbar - d - 2dS^{z}\right)\tau\right]S^{\mp}.$$
(3)

The reason for this situation is that in the presence of OA the basis of the operators that determine the state of an individual site becomes larger: it includes not only the spin operators, but also the tensor operators O_l^m of the quadrupole (l = 2), octupole $(l = 3), \ldots 2S$ -pole moments (of second, third, $\ldots 2S$ th power in the spin operators). This corresponds to a transformation from the Lie SU(2) algebra in the isotroopic case to the Lie SU(2S + 1) algebra in the presence of OA.¹³⁻¹⁵

It is found in this approach that in the case of the zeroth Hamiltonian (2) Wick's theorem is satisfied for certain operators that make up another complete set of generators of the SU(2S + 1) algebra and are linearly connected with the operators O_1^m , viz., Hubbard's operators X^{pq} , which are represented by matrices containing zeros everywhere except for a single unity element at the intersection of the *q*th column and the *p*th row, and describe the transition, at the site, from the state *q* to the state *p* (in our case, when the states of the site are characterized by the projection of the spin on the *z* axis, the parameters *p* and *q* take on the values $p, q = -S, -S + 1, \ldots, S$. For details on the operators O_1^m and the Hubbard operators see, e.g., Refs. 15, 19, and 20):

$$\langle TX_{1}^{p_{iq_{i}}}(\tau_{1})\dots X_{0}^{p_{q}}(\tau)\dots X_{n}^{p_{n}q_{n}}(\tau_{n})\rangle_{0}$$

$$=\sum_{i=1}^{n} K_{0i}^{p_{q}}(\tau-\tau_{i})$$

$$\times \langle TX_{1}^{p_{iq_{i}}}(\tau_{1})\dots [X_{i}^{p_{iq_{i}}}, X_{0}^{p_{q}}]_{\tau_{i}}\dots X_{n}^{p_{n}q_{n}}(\tau_{n})\rangle_{0}, \quad (4)$$

where

$$K_{p_{i}}^{p_{q}}(\tau-\tau_{i}) = \delta_{0i}K^{p_{q}}(\tau-\tau_{i}),$$

$$K^{p_{q}}(\tau-\tau_{i}) = \exp[(E_{p}-E_{q})(\tau-\tau_{i})] \begin{cases} 1+n_{p_{i}}, & \tau < \tau_{i} \\ n_{p_{q}}, & \tau > \tau_{i} \end{cases}, \quad (5)$$

$$n_{p_{q}} = [\exp(E_{p}-E_{q})-1]^{-1}.$$

The Wick theorem holds for the Hubbard operators because they have a non-operator dependence on the time if H_0 is of the form (2):

$$X^{pq}(\tau) = \{ \exp[(E_p - E_q)\tau] \} X^{pq}, \tag{6}$$

where E_p and E_q are the eigenenergies of the Hamiltonian H_0 and correspond to states with spin projections m = p and m = q.

Wick's theorem was used in Ref. 4, where a diagram

technique was developed for Hubbard operators as applied to a uniaxial ferromagnet in a longitudinal field. We regard this technique as too detailed for the case when the ion-ion interaction has, as usual, a spin-spin character, i.e., there are no interactions of higher order in the spin operators. In this case the interactions couple only three vertices, corresponding to the operators S^+ , S^- , and S^z out of the $(2S + 1)^2 - 1$ vertices connected with all the operators of the corresponding Lie algebra and contained in the approach of Ref. 4. Furthermore, all the physical characteristics are determined as a rule by means of spin operators.

It is therefore expedient, in our opinion, to project the total $[(2S + 1)^2 - 1]$ -dimensional space on the three-dimensional subspace of the spin operators and reformulate for the latter the VLP diagram technique with account taken of the properties of the complete Lie algebra. This idea is implemented below for the simplest nontrivial case S = 1.

For S = 1, the frequencies $E_p - E_q$ in (6) are given by $\varepsilon_2 = E_{-1} - E_0 = \hbar + d$, $\varepsilon_1 = E_0 - E_1 = \hbar - d$, $\varepsilon_3 = E_{-1} - E_1 = 2\hbar$.
(7)

Accordingly, there are three independent off-diagonal Hubbard operators $X^{-10}(\tau)$, $X^{01}(\tau)$, $X^{-11}(\tau)$ and three different Green's functions

$$K^{i}(\tau - \tau_{i}) \equiv K^{0i}(\tau - \tau_{i}), \quad K^{2}(\tau - \tau_{i}) \equiv K^{-i0}(\tau - \tau_{i}),$$

$$K^{3}(\tau - \tau_{i}) \equiv K^{-ii}(\tau - \tau_{i}).$$
(8)

Besides the aforementioned operators, the complete set of the generators of the SU(3) algebra include also the Hermitian-adjoint operators $X^{0-1}(\tau), X^{10}(\tau), X^{1-1}(\tau)$ and two diagonal operators X^{11} and X^{-1-1} , which are independent of time according to (6). These eight Hubbard operators are linearly connected with the eight tensor operators O_l^m (spin $O_1^m \equiv S^m$ at l = 1 and quadrupole $O_2^m \equiv O^m$ at l = 2, Ref. $15l^{2}$ by the formulas

$$S^{-}(\tau) = X^{-10}(\tau) + X^{01}(\tau),$$

$$O^{-1}(\tau) = -(S^{z}S^{-} + S^{-}S^{z})(\tau) = X^{-10}(\tau) - X^{01}(\tau),$$

$$O^{-2}(\tau) = (S^{-})^{2}(\tau) = X^{-11}(\tau),$$

$$S^{z} = X^{11} - X^{-1-1}, \quad O^{0} = (S^{z})^{2} - \frac{2}{3} = (X^{11} + X^{-1-1}) - \frac{2}{3}.$$
 (9)

Let us return to the sought mean values. Expressing $S_0^-(\tau)$ in terms of the Hubbard operators and using (4) as applied to the case S = 1, we get

$$\langle TS_{1}^{\alpha_{1}}(\tau_{1})\dots S_{0}^{-}(\tau)\dots S_{n}^{\alpha_{n}}(\tau_{n})\rangle_{0}$$

$$=\sum_{i=1}^{n} \{K_{0i}^{1}(\tau-\tau_{i})\langle TS_{1}^{\alpha_{1}}(\tau_{1})\dots [S_{i}^{\alpha_{1}}, X_{0}^{01}]_{\tau_{1}}\dots S_{n}^{\alpha_{n}}(\tau_{n})\rangle_{0}$$

$$+K_{0i}^{2}(\tau-\tau_{i})\langle TS_{1}^{\alpha_{1}}(\tau_{1})\dots [S_{i}^{\alpha_{t}}, X_{0}^{-10}]_{\tau_{i}},\dots S_{n}^{\alpha_{n}}(\tau_{n})\rangle_{0}\}.$$
(10)

We shall need later on the explicit form the commutation relations for the two Hubbard operators X^{01} and X^{-10} , and for the third off-diagonal Hubbard operator X^{-11} with the spin operators

$$[X^{-10}, S^{+}] = \frac{1}{2} (S^{z} - 3O^{0}),$$

$$[X^{01}, S^{+}] = \frac{1}{2} (S^{z} + 3O^{0}), \quad [X^{-11}, S^{+}] = X^{01} - X^{-10}, \quad (11)$$

$$[X^{-10}, S^{-}] = -[X^{01}, S^{-}] = X^{-11} = O^{-2}, \quad [X^{-11}, S^{-}] = 0,$$

$$[X^{-10}, S^{z}] = X^{-10}, \quad [X^{01}, S^{z}] = X^{01}, \quad [X^{-11}, S^{z}] = X^{-11}.$$

The described scheme for calculating the mean values of the spin operators is equivalent to carrying out all the possible pairings of the spin operators in accordance with new rules that can be deduced from the general rule

$$\widehat{X^{pq}(\tau_i)O_l}^m(\tau_j) = -K^{pq}(\tau_i - \tau_j)[X^{pq}, O_l^m]_{\tau_j}.$$
 (12)

In particular, one can realize the following normal pairings

$$\begin{split} \hat{S}_{i}^{-}(\tau_{i})\hat{S}_{j}^{+}(\tau_{j}) = & -\frac{i}{2}K_{ij}^{-1}(\tau_{i}-\tau_{j})\left(S_{j}^{z}+3O_{j}^{0}\right) \\ & -\frac{i}{2}K_{ij}^{-2}(\tau_{i}-\tau_{j})\left(S_{j}^{z}-3O_{j}^{0}\right), \end{split}$$
(13)

$$\widehat{S_{i}^{-}(\tau_{i})S_{j}^{2}(\tau_{j})} = -K_{ij}^{4}(\tau_{i}-\tau_{j})X_{j}^{01}(\tau_{j}) - K_{ij}^{2}(\tau_{i}-\tau_{j})X_{j}^{-10}(\tau_{j}),$$

$$\widehat{S_{i}^{-}(\tau_{j})O_{j}^{0}(\tau_{j})} = -K_{ij}^{4}(\tau_{i}-\tau_{j})X_{j}^{04}(\tau_{j}) + K_{ij}^{2}(\tau_{i}-\tau_{j})X_{j}^{-10}(\tau_{j})$$

and the anomalous pairing

$$\widehat{S_{i}^{-}(\tau_{i})}\widehat{S}_{j}^{-}(\tau_{j}) = [K_{ij}^{+1}(\tau_{i}-\tau_{j})-K_{ij}^{-2}(\tau_{i}-\tau_{j})]X_{j}^{-1}(\tau_{j}), \qquad (14)$$

which follow from the equations (12) for pairing the Hubbard operators and from the commutation relations (11).

In the calculation of the corrections to the paired Green's functions, there can be encountered the simplest vertex blocks shown in Fig. 1. In these blocks an empty circle and a circle with a point in it correspond to the diagonal operators S^z and O^0 , while circles with + and - signs correspond to the operators S^+ and S^- . Each type of vertex block is a graphic representation of the mean value, with zeroth Hamiltonian, of a *T*-product of the corresponding operators in the coordinate representation, among which are paired those operators that correspond to vertices joined by lines; the arrow indicates the direction of the time. Just as in the VLP technique, the only nonzero blocks are those containing the equal number of operators S^- and S^+ . Corresponding to these diagrams are the analytic expressions

a)
$$-\frac{1}{2}K_{12}^{i}(\tau_{1}-\tau_{2})\langle S^{z}+3O^{0}\rangle_{0}-\frac{1}{2}K_{12}^{2}(\tau_{1}-\tau_{2})\langle S^{z}-3O^{0}\rangle_{0};$$
b)
$$\frac{1}{2}K_{12}^{i}(\tau_{1}-\tau_{2})K_{23}^{i}(\tau_{2}-\tau_{3})\langle S^{z}+3O^{0}\rangle_{0}$$

$$+\frac{1}{2}K_{12}^{2}(\tau_{1}-\tau_{2})K_{23}^{2}(\tau_{2}-\tau_{3})\langle S^{z}-3O^{0}\rangle_{0};$$
c)
$$\frac{1}{2}K_{12}^{i}(\tau_{1}-\tau_{2})K_{23}^{i}(\tau_{2}-\tau_{3})\langle S^{z}+3O^{0}\rangle_{0}$$

$$-\frac{1}{2}K_{12}^{2}(\tau_{1}-\tau_{2})K_{23}^{2}(\tau_{2}-\tau_{3})\langle S^{z}-3O^{0}\rangle_{0};$$
d)
$$-\frac{1}{2}[2K_{12}^{i}(\tau_{1}-\tau_{2})-K_{12}^{2}(\tau_{1}-\tau_{2})]K_{32}^{i}(\tau_{3}-\tau_{2})K_{24}^{i}(\tau_{2}-\tau_{4})$$

$$\times \langle S^{z}+3O^{0}\rangle_{0}$$
(15)

$$\begin{array}{l} -\frac{i}{2} \left[2K_{12}{}^{2}(\tau_{1}-\tau_{2})-K_{12}{}^{4}(\tau_{1}-\tau_{2}) \right] K_{32}{}^{2}(\tau_{3}-\tau_{2}) K_{24}{}^{2}(\tau_{2}-\tau_{4}) \\ \times \langle S^{z}-3O^{0}\rangle_{0}, \\ \text{e)} \left[K_{13}{}^{4}(\tau_{1}-\tau_{3})-K_{13}{}^{2}(\tau_{1}-\tau_{3}) \right] K_{34}{}^{3}(\tau_{3}-\tau_{4}) \\ \times \left[\frac{i}{2}K_{42}{}^{1}(\tau_{4}-\tau_{2}) \langle S^{z}+3O^{0}\rangle_{0} - \frac{i}{2}K_{42}{}^{2}(\tau_{4}-\tau_{2}) \langle S^{z}-3O^{0}\rangle_{0} \right]; \\ \text{g)}, \text{ h)} -\frac{i}{2}K_{12}{}^{1}(\tau_{1}-\tau_{2}) K_{23}{}^{1}(\tau_{2}-\tau_{3}) K_{34}{}^{1}(\tau_{3}-\tau_{4}) \langle S^{z}+3O^{0}\rangle_{0} \\ -\frac{i}{2}K_{12}{}^{2}(\tau_{1}-\tau_{2}) K_{23}{}^{2}(\tau_{2}-\tau_{3}) K_{34}{}^{2}(\tau_{3}-\tau_{4}) \langle S^{z}+3O^{0}\rangle_{0}; \\ \text{f)} -\frac{i}{2}K_{12}{}^{1}(\tau_{1}-\tau_{2}) K_{23}{}^{1}(\tau_{2}-\tau_{3}) K_{34}{}^{1}(\tau_{3}-\tau_{4}) \langle S^{z}+3O^{0}\rangle_{0} \\ +\frac{i}{2}K_{12}{}^{2}(\tau_{1}-\tau_{2}) K_{23}{}^{2}(\tau_{2}-\tau_{3}) K_{34}{}^{2}(\tau_{3}-\tau_{4}) \langle S^{z}-3O^{0}\rangle_{0}. \end{array}$$

It can be seen from them that a solid line on a diagram does not stand for a single zeroth Green's function, as in the VLP technique, but to a definite linear combination of three zeroth Green's functions. We note also the appearance of new vertex blocks compared with the VLP technique. These are, in particular, the block e), which corresponds to pairing of the operators $S_1^{-}(\tau_1)$ and $S_3^{-}(\tau_3)$, and the blocks c), h), and f), which contain the quadrupole diagonal operator O^0 . In the calculation of the corrections to the series for the Green's functions, the latter appear only as intermediate elements for the construction of more complicated blocks, each of which can be obtained by combination of the simplest blocks shown in Fig. 1.

The pairings leave us with mean values containing only the time-independent diagonal operators S^z and O^0 . The prescription for their calculation is similar to that in the VLP technique for mean values containing only the operators S^z . Namely, differentiating the expression for the zeroth partition function

$$Z_0 = \operatorname{Sp} \exp (yS^z + \Gamma \cdot 3O^0), \quad y = \beta \tilde{h}, \quad \Gamma = -\beta d/3, \quad \beta = 1/kT,$$
(16)

with respect to the parameters y and Γ we obtain

$$\sigma = \langle S^r \rangle_0 = \frac{1}{Z_0} \frac{\partial Z_0}{\partial y}, \quad \lambda \equiv 3 \langle O^0 \rangle_0 = \frac{1}{Z_0} \frac{\partial Z_0}{\partial \Gamma}. \quad (17)$$

Further differentiation yields equations analogous to those of Ref. 18:

$$\langle S^{z}\rangle_{0} = \sigma, \quad 3\langle O^{0}\rangle_{0} = \lambda,$$

$$\langle S^{z}(\mathbf{r}_{1})S^{z}(\mathbf{r}_{2})\rangle_{0} = \sigma^{2} + \delta_{12}\sigma_{y}', \quad \langle 3O^{0}(\mathbf{r}_{1})3O^{0}(\mathbf{r}_{2})\rangle_{0} = \lambda^{2} + \delta_{12}\lambda_{\Gamma}',$$

$$\langle S^{z}(\mathbf{r}_{1})3O^{0}(\mathbf{r}_{2})\rangle_{0} = \langle 3O^{0}(\mathbf{r}_{1})S^{z}(\mathbf{r}_{2})\rangle_{0} = \sigma\lambda + \delta_{12}\sigma_{\Gamma}', \dots$$
(18)

Explicit formulas for the parameters σ and λ , the first derivatives σ'_y , λ'_r , $\sigma'_r = \lambda'_y$, and the higher-order derivatives can be easily from (16) and (17) (the former coincide with Eqs. (11) of Ref. 9).

We retain for the mean values (18) the graphic representation used in the VLP technique, i.e., we assign to the terms

FIG. 1.

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containing a definite number of δ symbols and connect individual vertices into definite combinations an oval (block) that encloses these vertices and their connecting lines. The only difference is that in the VLP technique a block that contains not even one broken-up element corresponds to a factor σ (or b in the notation of Refs. 17 and 18), a block containing one broken-up element corresponds to $-\sigma'_{\nu}(b')$, one containing two elements to $-\sigma''_{\nu}(b''),...,$ whereas in our case these factors are certain linear combinations of σ and λ if there are no broken-up elements in the block, linear combinations of the first derivatives σ'_{ν} , λ'_{Γ} , $\sigma'_{\Gamma} = \lambda'_{\nu}$, if there is one such element, and so forth. The form of the linear combination corresponding to a block depends on the vertices that it combines. For example, the diagrams of Fig. 1, in which there are no broken-up elements, correspond in Eqs. (15) to the linear combinations $\langle S^z + 3O^0 \rangle_0 \equiv \sigma + \lambda$.

3. DIAGRAM TECHNIQUE FOR THE CASE OF AN OA OF ARBITRARY SYMMETRY AND OF AN ARBITRARILY DIRECTED EXTERNAL FIELD

We consider a situation in which the OA has arbitrary symmetry and the external field has arbitrary direction. In this case the eigenstates of the zeroth Hamiltonian do not coincide with the eigenstates of the operator S^{z} and therefore cannot be numbered in accord with the projection of the angular momentum on the z axis. The Hubbard operators (6) have accordingly no longer the simple operator representation on which the generalized Wick theorem (4) was based.

It is easy to perceive that the Wick theorem can be formulated in a form similar to (4) in the general case of an arbitrary S and an arbitrary form of H_0 for those SU(2S + 1) algebra operators which describe transitions between different eigenstates of the Hamiltonian H_0 . The only difficulty lies in identifying the quantum number that labels the eigenstates of H_0 in the case when they do not coincide with the eigenstates of any of the three angular-momentum operators.

This difficulty can be circumvented by transforming, through rotations in $[(2S + 1)^2 - 1]$ -dimensional space, to those coordinates in which H_0 is diagonal¹⁴:

$$\tilde{H}_{0} = -\tilde{\hbar} \sum_{i} \tilde{S}_{i}^{\,i} + \tilde{d}_{2} \sum_{i} \tilde{O}_{2}^{\,0}(i) + \ldots + \tilde{d}_{28} \sum_{i} \tilde{O}_{28}^{\,0}(i),$$

where $\tilde{O}_{2}^{0},...,\tilde{O}_{2S}^{0}$ are the quadrupole and ... the 2S-pole diagonal operators in the rotated space, the latter marked with a superior tilde. The eigenstates of the zeroth Hamiltonian H_0 in terms of these coordinates is characterized, just as in the uniaxial case, by the projection of the spin on the z axis, so that Wick's theorem can be formulated in terms of these coordinates for the same Hubbard operators as in the preceding section.

We implement the described program for the simplest nontrivial case S = 1, when the OA of the most general form is described by the Hamiltonian

$$H^{a} = d \sum_{i} (S_{i}^{z})^{2} + E \sum_{i} [(S_{i}^{x})^{2} - (S_{i}^{y})^{2}].$$
(19)

Let the exchange be isotropic as before, and let the external field direction be arbitrary:

$$H^{h} = -h_{\parallel} \sum_{i} S_{i}^{z} - h_{\perp} \sum_{i} S_{i}^{x}.$$
⁽²⁰⁾

 H_0 takes accordingly the form

$$H_{0} = -(h_{\parallel} + J \langle S^{r} \rangle) \sum_{i} S_{i}^{z} - (h_{\perp} + J \langle S^{r} \rangle)$$

$$\times \sum_{i} S_{i}^{z} + d \sum_{i} O_{i}^{0} + E \sum_{i} (O_{i}^{2} + O_{i}^{-2}).$$
(21)

The unitary transformations (rotations in (8-dimensional space) that diagonalize it were obtained in Ref. 14:

$$U = \exp \left[\varphi \left(S^{+} + S^{-} \right) / \sqrt{2} \right]$$

$$\times \exp \left[K \left(O^{1} + O^{-1} \right) / \sqrt{2} \right] \exp \left[L \left(O^{2} - O^{-2} \right) \right].$$

The sought diagonal form of the zeroth Hamiltonian

$$\widehat{H}_{0} = U H_{0} U^{-1} = -\widetilde{\hbar} \sum_{i} \widetilde{S}_{i}^{z} + \widetilde{d} \sum_{i} \widetilde{O}_{i}^{0}$$
⁽²²⁾

is realized in 8-dimensional-space coordinates characterized by the angles φ , K, and L and satisfying Eqs. (8) of Ref. 9 (where \tilde{h} and \tilde{d} are replaced by $h(\varphi, K, L)$ and $d(\varphi, K)$, and E is replaced by ε). The same reference contains explicit equations [(7)] that relate \tilde{h} and \tilde{d} with the angles φ , K, and L with the only nonzero mean values $\sigma = \langle \tilde{S}^z \rangle_0$, $\lambda = 3 \langle \tilde{O}^0 \rangle_0$, in terms of these coordinates, as well as transcendental equations [(11)] for them.

We proceed to find the mean values of the spin operators $S^{\alpha}(\tau)$ in the initial space. We use the equalities of these mean values, defined with the Hamiltonian (21), to the mean values $\tilde{S}^{\alpha}(\tau)$, in rotated space, of the same operators defined with the uniaxial H_0 from (22). We express next the spin operators \tilde{S}^{α} in terms of the spin and quadrupole operators in the initial space

$$S^{\alpha} = \sum_{m=-1}^{1} A_{m}^{\alpha} S^{m} + \sum_{m=-2}^{2} B_{m}^{\alpha} O^{m}.$$
 (23)

(Explicit equations for the coefficients A_m^{α} and B_m^{α} can be easily obtained from the equations of Ref. 14 and are cited in the Appendix). The sought mean values of the operators in rotated space and specified with nondiagonal H_0 are expressed in terms of mean values determined with diagonal \tilde{H}_0 , from the spin and quadrupole operators $S^m(\tilde{\tau})$, $O^m(\tilde{\tau})$ in the initial space, which have a time dependence that is now determined by the uniaxial \tilde{H}_0 from (22). This connection can be represented as the existence of a relation (defined only under the mean-value sign) between the operator $S^{\alpha}(\tau)$ and the operators $S^m(\tilde{\tau})$, $O^m(\tilde{\tau})$, defined with uniaxial \tilde{H}_0 :

$$S^{\alpha}(\tau) = \sum_{m=-1}^{1} A_m{}^{\alpha} S^m(\tilde{\tau}) + \sum_{m=-2}^{2} B_m{}^{\alpha} O^m(\tilde{\tau}).$$
⁽²⁴⁾

Since the latter operators are defined in the initial space, in which the interaction is of the spin-spin type, the interaction lines connect, just as in the uniaxial case, only three types of vertices corresponding to the spin operators.

The problem of determining the mean values of spin operators with arbitrary zeroth Hamiltonian has thus been

reduced to a determination of the mean values of spin and quadrupole operators with uniaxial \tilde{H}_0 for H^{int} corresponding to the initial isotropic exchange, i.e., to the problem solved in Sec. 2.

It is useful to formulate the obtained rules for calculating mean values of spin operators in the language of pairing of these operators. It follows from (24) that such pairings can be represented as a sum of all possible pairings of the operators $S^m(\tilde{\tau})$, $O^m(\tilde{\tau})$, defined in uniaxial \tilde{H}_0 . They are given [Eqs. (13) and (14)] in Sec. 2. We present below the pairings similarly obtained for quadrupole operators:

$$\widetilde{O}_i^{-1}(\widetilde{\tau}_i) \widetilde{O}_j^2(\widetilde{\tau}_j) = K_{ij}^{-1}(\tau_i - \tau_j) X^{0-1}(\widetilde{\tau}_j) + K_{ij}^{-2}(\tau_i - \tau_j) X_j^{10}(\widetilde{\tau}_j).$$

Using the representation (24), the explicit expressions given for the coefficients A_m^{α} and B_m^{α} in the Appendix, and the pairing equations (25), (13), and (14), we can easily obtain explicitly the sought formulas for pairings of spin operators in the interaction representation with arbitrary H_0 , and calculate on their basis the various vertex-block types.

In particular, the simplest paired vertex blocks shown in Fig. 2 correspond to the analytic expressions

$${}^{i}/{}_{2}(\sigma+\lambda) \left[K_{12}{}^{i}(\tau_{1}-\tau_{2})C_{+}{}^{\alpha\beta} + K_{21}{}^{i}(\tau_{2}-\tau_{1})C_{-}{}^{\alpha\beta} \right]$$

$$+ {}^{i}/{}_{2}(\sigma-\lambda) \left[K_{12}{}^{2}(\tau_{1}-\tau_{2})D_{+}{}^{\alpha\beta} + K_{21}{}^{2}(\tau_{2}-\tau_{1})D_{-}{}^{\alpha\beta} \right]$$

$$+ \sigma \left[K_{12}{}^{3}(\tau_{1}-\tau_{2})E_{+}{}^{\alpha\beta} + K_{21}{}^{3}(\tau_{2}-\tau_{1})E_{-}{}^{\alpha\beta} \right]$$

$$+ \sum_{l, \ l'=1,2} A_{ll'} \langle O_{l}{}^{0}(\mathbf{r}_{1})O_{l'}{}^{0}(\mathbf{r}_{2}) \rangle_{\widetilde{H}_{0}}, \qquad (26)$$

where

$$C_{+}{}^{\alpha\beta} = - (B_{-1}{}^{\alpha} - A_{-1}{}^{\alpha}) (B_{1}{}^{\beta} - A_{1}{}^{\beta}),$$

$$C_{-}{}^{\alpha\beta} = - (B_{1}{}^{\alpha} - A_{1}{}^{\alpha}) (B_{-1}{}^{\beta} - A_{-1}{}^{\beta}),$$

$$D_{+}{}^{\alpha\beta} = - (B_{-1}{}^{\alpha} + A_{-1}{}^{\alpha}) (B_{1}{}^{\beta} + A_{1}{}^{\beta}),$$

$$D_{-}{}^{\alpha\beta} = - (B_{1}{}^{\alpha} + A_{1}{}^{\alpha}) (B_{-1}{}^{\beta} + A_{-1}{}^{\beta}),$$

$$E_{+}{}^{\alpha\beta} = B_{-2}{}^{\alpha}B_{2}{}^{\beta}, \quad E_{-}{}^{\alpha\beta} = B_{2}{}^{\alpha}B_{-2}{}^{\beta},$$
(27)

 $\overrightarrow{a} \rightarrow - \cancel{B}$ FIG. 2.

$$\begin{array}{ll} A_{11}{}^{\alpha\beta} = A_0{}^{\alpha}A_0{}^{\beta}, & A_{12}{}^{\alpha\beta} = A_0{}^{\alpha}B_0{}^{\beta}, \\ A_{21}{}^{\alpha\beta} = B_0{}^{\alpha}A_0{}^{\beta}, & A_{22}{}^{\alpha\beta} = B_0{}^{\alpha}B_0{}^{\beta}. \end{array}$$

A characteristic feature of these blocks is that they differ from zero for an arbitrary pair of operators S^{α} and S^{β} , in contrast to the cases of an isotropic FM and an FM with uniaxial anisotropy in a longitudinal field, where only the blocks with $\alpha = -$, $\beta = +$ differ from zero. Analogously, the number of nonzero vertex blocks containing three and four vertices increases (compared with Fig. 1) because they can include arbitrary combinations of the operators S^{α} and S^{β} .

As already noted, we use the representation of the correlation functions in terms of a scattering matrix (Eq. 5.7) of Ref. 18). This representation generates expansions of these functions in powers of the interaction. The correction of nth order in n is shown in the form of individual blocks, pertaining to one site, which are joined by bare-interaction lines.

As a result of the foregoing features of vertex block, the nonzero paired correlation functions $G^{\alpha\beta}$ will be those of the spin operators S^{α} and S^{β} with arbitrary indices and α and β , and the equations for them will be coupled with correlation functions with other different indices, i.e., the series will be branched in terms of the indices. We can nevertheless proceed as usual,¹⁷ i.e., introduce the concept of a nonreducible part of the diagram which cannot be cut on the elementary line of the interaction if defined in matrix form $(\hat{\Sigma})$. The connection between the correlation function and the irreducible part is then given by the relation $\hat{G} = (\hat{I} - \hat{\Sigma}\hat{V})^{-1}\hat{\Sigma}$, which differs from Larkin's equation only in the matrix form (\hat{V}) is the bare-interaction matrix and \hat{I} is the unit matrix).

The next step is to classify the diagram in accord with some small parameter. In our problem we can choose it to be the reciprocal of the average relative radius of the exchange interaction, $1/r_0^3$ (Ref. 17). In the magnets of interest to us, in which the OA is comparable with the exchange interaction, the latter is mainly indirect, since it is long-range, so that the chosen parameter is quite good (of course, far from the fluctuation region).

The diagrams are classified in accord with this parameter with account taken of the fact that the result of a single summation over the intermediate momentum **q** is of the order of 1/r. Accordingly, the main contribution is made by those diagrams with not even one summation over **q**. These steplike diagrams for the correlation functions, in which the paired vertex blocks shown in Fig. 2 are connected by bareinteraction lines. Calculation of an infinite series of such diagrams for the correlation functions is equivalent to zerothapproximation calculation of the irreducible part when its components are determined by Fourier transforms of the vertex blocks (26), i.e., by formulas (26) in which $K^{l}(\tau_{i} - \tau_{j})$ are replaced by their Fourier transforms:

$$K^{i}(i\omega_{n}) = [\beta(\varepsilon_{l} - i\omega_{n})]^{-1}, \quad l=1, 2, 3.$$

$$\varepsilon_{1, 2} = \tilde{h} \mp \tilde{d}, \quad \varepsilon_{3} = 2\tilde{h}.$$
 (28)

4. GROUND STATE AND SPECTRUM OF COLLECTIVE EXCITATIONS

In the considered zeroth approximation the ground state is determined by one of the eigenvectors of the Hamil-

tonian H_0 . In the 8-dimensional-space coordinates, in which this Hamiltonian is diagonal, the ground state is an eigenstate of the operator \tilde{S}^z and is therefore characterized by the maximum, in absolute value, projection of the spin on the z axis, i.e.,

$$\sigma = \langle \tilde{S}^z \rangle_0 = 1, \quad \lambda = 3 \langle \tilde{O}^0 \rangle_0 = 1 \quad \text{or} \quad \sigma = -1, \quad \lambda = 1, \quad (29)$$

or by the minimum projection

$$\sigma=0, \quad \lambda=-2. \tag{30}$$

The foregoing solutions of Eqs. (11) of Ref. 9 are unitarily equivalent and correspond to a different choice of the 8-dimensional-space frame in which \tilde{H}_0 is diagonal.

What is the ground state in the initial space? Since S^z is a superposition of generators of an 8-dimensional space [Eq. (34)], in the general case the ground state is an eigenstate of an operator that is a certain linear combination of the spin and five quadrupole operators. Obviously, if there is no spinoperator contribution to this linear combination, the ground state will be nonmagnetic (all the magnetization projections are equal to zero, and the order will be determined by the mean values of the quadrupole operators O_2^m . Such a ground state can be naturally called a quadrupole-ordered phase.³⁾, Obviously, it cannot be realized in the absence of OA, when the complete aggregate of the operators that describe the state of the site includes only spin operators.

Another possibility, when the linear combination contains both spin and quadrupole operators, corresponds from the magnetization viewpoint to the ordinary ferromagnetic phase. It is important to note, however, that since the maximum, unity value is reached at T = 0 by the length of the vector in 8-dimensional space, and its components are both spin and quadrupole operators, the average magnetization does not reach saturation at T = 0, and can be arbitrary. The ferromagnetic phase with the most complicated structure is the canted phase, in which the magnetization vector makes an angle with the external-field direction, i.e., both magnetization projections $M^z = \langle S^z \rangle$ and $M^x = \langle S^x \rangle$ differ from zero (the third component M^{ν} vanishes identically if the external field has no y component); in addition, the mean values of the quadrupole operators differ from zero, $\langle O^0 \rangle \neq 0, \langle O^2 + O^{-2} \rangle \neq 0, \langle O^1 - O^{-1} \rangle \neq 0$. The closest to the usual ferromagnetic phase is a collinear ferromagnetic phase in which the magnetization is parallel to the external field.

We proceed now to a quantitative description of the ground state. We consider for the sake of argument an external field parallel to the z axis. With account taken of an OA of arbitrary symmetry in the initial Hamiltonian, followed by analysis of all three possible phases, this choice is general enough, since one can reduce to it any arbitrary case of rotation of the z axis in the xz plane through the angle between that plane and the external field. When uniaxial OA is considered this is not the case, since rotation in three-dimensional case leads to the appearance of one-ion terms of more complicated symmetry. On the other hand, this is precisely the preferred choice for the analysis of the Hamiltonian eigenvectors corresponding to collective excitations (see the end of the section), for in this case the z axis singled out by the

external field coincides with the singled-out quantization axis in the expression (9) for the quadrupole operators.

The qualitative classification of the possible phases corresponds to the following solutions of Eqs. (8) and (11) of Ref. 9 for the angles φ , K, and L and the mean values σ and λ at T = 0.

1) Quadrupole-ordered (QO) phase:

$$\sin \varphi = \sin K = 0, \quad \text{tg } 2L = -E/h, \quad \sigma = 0, \quad \lambda = -2, \quad (31)$$

 $M^z = M^x = 0, \quad 3\langle O^0 \rangle = 1, \quad \langle O^2 + O^{-2} \rangle = -1. \quad (31a)$

2) Collinear ferromagnetic (FM_z) phase:

$$\sin \varphi = \sin K = 0, \quad \sigma = 1, \quad \lambda = 1.$$
 (32)

The parameter 2L is determined by the fourth-degree equation

$$h \sin 2L + \frac{1}{2} J \sigma \sin 4L + E \cos 2L = 0.$$
 (32a)

The magnetizations are equal to

s

$$M^{x}=0, \quad M^{z}=\cos 2L\neq 0. \tag{32b}$$

3) Canted ferromagnetic phase (FM_{zx}) :⁴⁾

$$\cos 2K = \frac{d+E}{2J} - \frac{h^2}{2J(d-E)},$$

$$\sin \varphi = \frac{h}{E-d} \left\{ \frac{(d-E)(2J-d-E) + h^2}{(d-E)(2J+d+E) - h^2} \right\}^{1/2}$$
(33)

$$\operatorname{ctg} 2L = -H(\varphi, K)/D_{2}(\varphi, K), \quad \sigma = 0, \quad \lambda = -2,$$
$$M^{z} = -\sin \varphi \sin 2K, \quad M^{x} = \cos \varphi \sin 2K. \quad (33a)$$

The foregoing explicit values of the parameters φ , K, L, σ , λ in the different phases determine exhaustively all the equilibrium characteristics of the system at T = 0. For example, the average magnetizations and the quadrupole mean values are connected with these parameters by Eqs. (13) of Ref. 9, from which follow, in particular, the relations (31a), (32b), and (33a) of the present paper; from them, on the other hand, it is easy to obtain explicit formulas for the susceptibilities, etc. The frequencies of the collective excitations, obtained later in the present paper, are also determined explicitly by the values of φ , K, L, σ , and λ .

The same three phases can be realized at finite T. The corresponding values of the angles φ , K, and L and of the mean values σ and λ , just as at T = 0, are determined by the solution of Eqs. (8)–(11). In the general case, however, the solution cannot be obtained analytically at arbitrary T. Nonetheless, the classification of the solutions corresponding to different phases can be easily implemented also in this case. In particular, for the FM_z and QO phases, we have as before

$$\sin \varphi = \sin K = 0, \tag{34}$$

the angle L is determined by Eq. (32a) but with $\sigma \neq 1$, and the values σ and λ are determined by the transcendental equations (11) of Ref. 9.

An analysis of the transitions between the foregoing phases, with respect to field and temperature, is carried out in part in Ref. 9.

We proceed here to an investigation of the spectra of collective excitations in different phases. The sought fre-

quencies are determined by the poles of the correlation function or by the zeros of the dispersion equation

det $[\hat{I} - \hat{\Sigma}(\mathbf{k}, \omega) \hat{\mathcal{V}}_{\mathbf{k}}] = 0.$

In the considered lowest approximation, the irreducible part was determined in Sec. 3. Explicit calculations yield after a number of similarlity transformations of the matrix $\hat{\Sigma}\hat{V}$ a third-degree dispersion equation

$$\det \left[\omega^2 \hat{I} - \hat{D}_+(\mathbf{k}) \hat{D}_-(\mathbf{k}) \right] = 0.$$
(35)

The matrix elements of the three-dimensional matrices \hat{D}_{+} (k) are explicitly given by the formulas

$$[D_{\pm}(\mathbf{k})]_{pq} = d_{\pm}^{pq}(\mathbf{k}) [\frac{1}{2} \delta_{p1}(\sigma + \lambda) + \frac{1}{2} \delta_{p2}(\sigma - \lambda) + \delta_{p3}\sigma] + \delta_{pq}e_{p},$$

$$d_{\pm}^{11} = V_{1} \pm V_{2}, \quad d_{\pm}^{22} = V_{3} \pm V_{4}, \quad d_{\pm}^{33} = u_{1} \pm u_{2}, \quad (35a)$$

$$d_{\pm}^{12} = v_{1} \pm v_{2}, \quad d_{\pm}^{13} = W_{1} \pm w_{1}, \quad d_{\pm}^{23} = W_{2} \pm w_{2}.$$

Here δ_{pq} is the Kronecker delta, $p, q = 1,2,3; d_{\pm}^{qp} = d_{\pm}^{qp}$, and the dependence of the coefficients on the wave vector **k** is determined by the formulas

$$V_{1,3} = J(\mathbf{k}) [\frac{1}{2} \sin^2 2K (1 \pm \sin 2L) - 1],$$

$$V_{2,4} = \pm J(\mathbf{k}) \cos 2K (\sin 2L \cos^2 K \mp \sin^2 K),$$

$$v_1 = -J(\mathbf{k}) \cos^2 K \cos 2K \cos 2L,$$

$$v_2 = -2J(\mathbf{k}) \cos^2 K \sin^2 K \cos 2L,$$

$$W_{1,2} = \frac{J(\mathbf{k})}{\sqrt{2}} \sin 2K \cos^2 K \cos 2L (\cos L \pm \sin L),$$
$$w_{1,2} = -\frac{J(\mathbf{k})}{\sqrt{2}} \sin 2K (\sin^2 K \mp \cos^2 K \sin 2L) (\cos L \mp \sin L),$$

$$u_{1} = -J(\mathbf{k}) (1 - \cos^{4} K \cos^{2} 2L),$$

$$u_{2} = -J(\mathbf{k}) (\cos 2K - \cos^{4} K \cos^{2} 2L),$$
(36)

where $J(\mathbf{k})$ is the Fourier transform of the exchange integral.

The roots of the secular equation, (35) and (35a), yield the general solution of the problem of determining, in the approximation considered, the frequencies of the three branches of the spectrum of the collective excitations for the case of arbitrary OA symmetry and arbitrary external-field direction in any of the three possible phases. The dependences on the OA constants, on the field, and on the temperature are determined by the dependences, on these quantities, of the mean values σ and λ and of the unitary-transformation parameters σ , K, and L; these dependences, we recall are given by Eqs. (8) and (11) of Ref. 9.

We now study some particular cases. Consider first the collinear ferromagnetic and nonmagnetic phases, for which according to (34) $\sin \varphi = \sin K = 0$, so that the secular equation factors out and the spectrum decays into independent parts

$$\omega_{1,2}^{2}(\mathbf{k}) = (\varepsilon_{1}^{2} + \varepsilon_{2}^{2})/2 + V_{1}[(\sigma + \lambda)\varepsilon_{1} + (\sigma - \lambda)\varepsilon_{2}]/2 + v_{1}^{2}\sigma^{2}/2$$

$$\pm \{ [\varepsilon_{1}^{2} - \varepsilon_{2}^{2} + V_{1}[(\sigma + \lambda)\varepsilon_{1} - (\sigma - \lambda)\varepsilon_{2}] + v_{1}^{2}\sigma^{2}]^{2}/4$$

$$+ v_{1}^{2}\sigma^{2}\varepsilon_{2}[\varepsilon_{2} + V_{1}(\sigma - \lambda)] - v_{1}^{2}[\varepsilon_{1}(\sigma - \lambda) - \varepsilon_{2}(\sigma + \lambda)]^{2}/4 \}^{V_{1}},$$
(37a)

 $\omega_{s}^{2}(\mathbf{k}) = \varepsilon_{s} [\varepsilon_{s} - 2\sigma J(\mathbf{k}) \sin^{2} 2L]. \qquad (37b)$

The constants in (37a) and (37b), which determine the disper-

sion of the collective excitations, and the frequencies of the local transitions, are respectively equal to

$$V_{1}=V_{3}=-J(\mathbf{k}), \quad V_{2}=-V_{4}=J(\mathbf{k}) \sin 2L,$$

$$v_{1}=-J(\mathbf{k}) \cos 2L, \qquad (38)$$

$$u_{1}=u_{2}=-J(\mathbf{k}) \sin^{2} 2L,$$

$$\varepsilon_{1,2}=h \cos 2L+J\sigma \cos^{2} 2L-E \sin 2L\mp d,$$

$$\varepsilon_{3}=2(h \cos 2L+J\sigma \cos^{2} 2L-E \sin 2L).$$

$$(39)$$

They contain, besides the constants of the initial Hamiltonian, the parameter sin 2L defined by Eq. (32a) (in which $\sigma \neq 1$ unlike at T = 0) and the mean values σ and λ defined by the transcendental equations (11) of Ref. 9.

Let us analyze the nature of the collective excitations in the phases considered. Whereas in an isotropic FM the only collective-excitation branch is connected with the oscillations of the transverse components of the spin, i.e., is a spin wave, in the presence of OA the picture of the collective oscillations becomes complicated even in the considered simplest phases. Namely, each of the two branches with frequencies $\omega_1(\mathbf{k})$ and $\omega_2(\mathbf{k})$ is connected with a superposition of oscillations of the transverse components of the spin and the components O_{-k}^{1} , O_{k}^{-1} of the quadrupole moment, while the branch with frequency $\omega_3(\mathbf{k})$ is connected with a superposition of the longitudinal component of the spin and the components O_{-k}^2 , O_{k}^{-2} , of the quadrupole moment, i.e., all three branches are mixed spin-quadrupole waves. The foregoing follows from the fact, which can be easily verified by direct calculations, that the branch $\omega_3(\mathbf{k})$ is connected with a subspace specified by the operators S_{k}^{z} , O_{k}^{-2} , O_{-k}^{2} , since $S^{\mathbf{z}}(\tau) = \alpha_2 O^2 + \alpha_0 S^{\mathbf{z}} + \alpha_{-2} O^{-2}$, while the branches $\omega_1(\mathbf{k})$ and $\omega_2(\mathbf{k})$ are connected with a subspace specified by the operators $S_{\mathbf{k}}^{-}$, $S_{-\mathbf{k}}^{+}$, $O_{\mathbf{k}}^{-1}$, $O_{-\mathbf{k}}^{1}$, since

$$S^{-}(\tau) = \alpha_1 S^{+} + \alpha_{-1} S^{-} + \beta_1 O^{1} + \beta_{-1} O^{-1}.$$

The equations (37a) and (37b) for the frequencies are formally alike for both considered phases, but for the ferromagnetic and quadrupole phases the solutions for the parameters σ , λ , sin 2L are different, in analogy with the difference between the solutions (31a) and (32a) at T = 0. This leads to a different character of the spectrum in the considered phases. For example, for the QO phase the mode that becomes separated, with frequency $\omega_3(\mathbf{k})$, is thermal, i.e., it is excluded from the collective-excitation spectrum at T = 0. It corresponds to local transitions from one excited level to another. For the FM_z phase, the same mode has dispersion at T = 0.

We note that in the uniaxial case [when sin 2L = 0 according to (32a)], the formulas for the FM_z -phase spectrum simplify and coincide with the results of Refs. 1 and 2.

As for the most complicated canted ferromagnetic phase, at finite T the dispersion equation is not factored out in this case and the frequencies of the three branches of the collective excitations, each of which has dispersion, are given by the three solutions of the cubic equation (35), which are too elaborate to write down here. The nature of the collective excitation is also more complicated in this phase—each of the three branches corresponds to a superposition of the oscillations of all eight generators of the SU(3) algebra, i.e., the three spin components and the five components of the quadrupole moment, since it is connected with the complete eight-dimensional space. At T = 0 [where $\sigma = 0$ according to (33)] one of the branches drops out of the spectrum of the collective excitations and the solutions of the remaining quadratic dispersion equations are

$$\omega_{1,2}^{2}(\mathbf{k}) = (\varepsilon_{1} - V_{1} + V_{2}) (\varepsilon_{1} - V_{1} - V_{2})/2 + (\varepsilon_{2} + V_{3} + V_{4}) (\varepsilon_{2} + V_{3} - V_{4})/2 + v_{2}^{2} - v_{1}^{2} \pm \{ [(\varepsilon_{1} - V_{1} + V_{2}) (\varepsilon_{1} - V_{1} - V_{2})/2 - (\varepsilon_{2} + V_{3} + V_{4}) (\varepsilon_{2} + V_{3} - V_{4})/2]^{2} + [(\varepsilon_{1} - V_{1} - V_{2}) (v_{1} - v_{2}) + (\varepsilon_{2} + V_{3} - V_{4}) (v_{1} + v_{2})] + [(\varepsilon_{1} - V_{1} + V_{2}) (v_{1} + v_{2}) + (\varepsilon_{2} + V_{3} + V_{4}) (v_{1} - v_{2})] \}^{V_{2}}.$$
(40)

It can be easily shown that at T = 0, just as at finite temperatures, each of the two branches is a superposition of the oscillations of three components of the spin magnetic moment and five components of the quadrupole moment.

At T = 0, the equations for the spectra of the FM_z and QO phases are also simplified. For the FM_z phase, taking (32) into account, we have for the two modes with dispersion

$$\omega_1^2(\mathbf{k}) = \varepsilon_1^2 - 2J(\mathbf{k})\varepsilon_1 + (J(\mathbf{k})\cos 2L)^2, \ \varepsilon_1 = -d - E/\sin 2L,$$

$$\omega_3^2(\mathbf{k}) = \varepsilon_3(\varepsilon_3 - 2J(k)\sin^2 2L), \ \varepsilon_3 = -2E/\sin 2L.$$
(41)

The parameter sin 2L is determined by Eq. (32a) at $\sigma = 1$. For the QO phase the frequencies of the branches with dispersion take, with allowance for (31), the form

$$\omega_{1,2}^{2}(\mathbf{k}) = d^{2} + h^{2} + E^{2}$$

-2J(\mbox{k}) d\pm 2 { (E^{2} + h^{2}) (J(\mbox{k}) - d)^{2} - (J(\mbox{k}) h)^{2} }^{1/n}. (42)

We note also that at T = 0 the Hamiltonian eigenvectors corresponding to frequencies (41) and (42) change form. In particular, to the branch $\omega_1(\mathbf{k})$ for the FM_z phase and to each of the branches $\omega_1(\mathbf{k})$ and $\omega_2(\mathbf{k})$ for the QO phase there corresponds at T = 0 a superposition of oscillations of only the transverse components of the spin magnetic moment, i.e., a spin wave. This can be easily verified in the following manner. The eigenvectors of the zeroth Hamiltonian $H_0(i)$ for these phases are [Eq. (17) of Ref. 9 at $\varphi = K = 0$]

$$\mathbf{e}_{1} = \begin{vmatrix} \cos L \\ 0 \\ \sin L \end{vmatrix}, \quad \mathbf{e}_{2} = \begin{vmatrix} 0 \\ 1 \\ 0 \end{vmatrix}, \quad \mathbf{e}_{3} = \begin{vmatrix} -\sin L \\ 0 \\ \cos L \end{vmatrix}.$$
(43)

One of them, \mathbf{e}_1 for the FM_z phase and \mathbf{e}_2 for the QO phase, determines in the zeroth approximation the vacuum state. (This follows from the fact that the values (31) of the parameters σ and λ for the QO phase corresponds to an arrangement $E_0 < E_1 < E_{-1}$ of the levels of the Hamiltonian $H_0(i)$, while the values (32) of the parameters σ and λ for the FM_z phase correspond to the arrangement $E_1 < E_0 < E_{-1}$ so that the eigenvectors corresponding to the lower levels are

for the QO phase
$$\tilde{\mathbf{e}}_0 = \begin{vmatrix} 0 \\ 1 \\ 0 \end{vmatrix}$$
 and for the FM_z phase $\tilde{\mathbf{e}}_1 = \begin{vmatrix} 1 \\ 0 \\ 0 \end{vmatrix}$.

The transformation to the eigenvectors in the initial space is trivial.) At T = 0, when only transitions from the ground state are of importance from among the two types of excitations generated at a site by the operators S^{-} and O^{-1} , the action of the indicated operators is equivalent. (This can be easily verified by using the matrix representations of the operators S^{-} and O^{-1} [Eqs. (3) of Ref. 14] and our Eqs. (43).) Therefore the waves of the excitations generated at a site by the operators S_{i}^{-} , S_{i}^{+} , O_{i}^{-1} , O_{i}^{1} at finite T reduce at low values of T to pure spin waves generated by the operators S_{i}^{-} and S_{i}^{+} .

In the case of uniaxial OA, the spectrum of the collective excitations in the (only possible) FM_z phase reduces at T = 0 to a single branch of oscillations generated by the operators S_k^- and corresponding to the usual spin-wave theory.

5. CONCLUSION

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Let us summarize the features of the microscopic description of a magnet with OA, bearing in mind that they are determined by the presence of a wider (compared with the three spin operators) basis of operators that describe the state of an individual ion.

Generally speaking the algebra of these operators is specified only by the quantity S and reduces to the Lie SU(2S + 1) algebra, but the significance of the difference between this algebra and the ordinary SU(2) and of the influence of this difference on the physical characteristics of the system are determined by the form of the Hamiltonian. If the latter includes only operators of the SU(2) subalgebra (the case of absence of OA), all the eigenstates and hence the equilibrium characteristics of the Hamiltonian are described within the framework of this subalgebra. If, however, the Hamiltonian contains certain operators that take it out of SU(2) (the case when OA is present), its eigenstates are determined by the complete Lie algebra that is peculiar to the given value of the spin.

For arbitrary SU(n) Lie algebra, the Wick theorem holds for those operators of this algebra which generate excited states of the zeroth Hamiltonian H_0 . In the uniaxial case, when the eigenstates of H_0 are characterized by the projection of the spin magnetic moment on the z axis, such operators are the Hubbard operators X^{pq} (p, q = -S,...S). In all other cases these operators are rather complicated superpositions of various Hubbard operators or, in another language, of spin, quadrupole, ... 2S-pole operators.

The situation described is a reflection of the inner symmetry of the systems, a symmetry that cannot be neglected by considering only, for example, spin operators by Lie splitting of the higher operators or by some other approximate method. Since, however, the physical characteristics are determined as a rule just by the spin-magnetic moment operators, it is possible to project accurately the complete $[(2S + 1)^2 - 1]$ -dimensional space into a subspace of spin-

moment operators and reformulate for them the diagram technique of Refs. 17 and 18, with account taken of the properties of the complete Lie algebra, as was done in Secs. 2 and 3.

We note, however, that this is only a convenient mathematical device, and that the excited states of the Hamiltonian H_0 are generated in the general case, as before, by the spin-magnetic, quadrupole, ... 2S-pole moments. Allowance for H^{int} makes these excitations collective, but does not change their nature. In Sec. 4 it is shown (with S = 1 as the example, that in the general case each of the three branches of the collective excitations is a superposition of oscillations of the three components of the spin-magnetic-moment operator and five components of the quadrupole-moment operator. In various particular cases (for the collinear ferromagnetic phase, for the quadrupole phase), the basis of the operators may be narrower.

We have considered here isotropic exchange interaction. Generalization to the case of anisotropic exchange entails no difficulty. All the basic equations and assumptions of the paper remain unchanged.

In addition, in the calculation of the spectrum we have confined ourselves to a spin S = 1. Generalization to arbitrary spin is not difficult in principle, but at large S the calculations become laborious, owing to the increased number of basis operators of the corresponding Lie algebra. At large S it is therefore more advantageous to use theories that contain the small parameter 1/S. In particular, it is possible in this case to use the Holstein-Primakoff transformation²² which, we recall, is effective for magnets with sufficiently strong OA only under this condition.²³

In conclusion, I wish to thank V. M. Adamyan for constant support and interest in the work, as well as A. A. Abrikosov and I. E. Dzyaloshinskiĭ for helpful discussions.

APPENDIX

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We present here the values of the constants in expression (23):

$$A_{\pm i}^{0} = \pm \frac{1}{\sqrt{2}} (\cos \varphi \sin K \sin L + \sin \varphi \cos 2K \cos L),$$

$$A_0^{\circ} = \cos K (\cos \varphi \cos 2L + \sin \varphi \sin K \sin 2L),$$

$$B_{\pm 2}^{0} = \cos K(-\sin 2L \cos \varphi + \sin \varphi \sin K \cos 2L),$$

$$B_{\pm i}^{0} = \pm \frac{1}{\sqrt{2}} (\cos \varphi \sin K \cos L - \sin \varphi \cos 2K \sin L),$$

$$B_0^{0} = 3\sin\varphi\sin K\cos K,$$

$$A_{\pm i}^{\ i} = \frac{1}{2} [\pm (-\sin\varphi\sin K\sin L + \cos\varphi\cos 2K\cos L)]$$

$$+\cos K\cos L$$
],

$$A_0^{4} = \frac{1}{\sqrt{2}} \cos K (-\sin \varphi \cos 2L + \sin K \cos \varphi \sin 2L),$$

 $B_{\pm 2}{}^{i} = \frac{1}{\sqrt{2}} [\cos K (\sin \varphi \sin 2L + \sin K \cos \varphi \cos 2L) \pm \sin K],$

$$B_{\pm 1}^{4} = \frac{1}{2} [\mp (\sin \varphi \sin K \cos L + \cos \varphi \cos 2K \sin L) + \cos K \sin L],$$
$$B_{0}^{4} = \frac{3}{\sqrt{2}} \cos K \sin K \cos \varphi.$$

¹⁾The solutions given for it in a number of papers, e.g., in Ref. 16, where rhombic OA is considered, in Ref. 4, where a uniaxial ferromagnet is investigated in the presence of a transverse field, and elsewhere, cannot be regarded as satisfactory, since the treatment of nontrivial cases in these papers is restricted to a single trivial phase, the collinear ferromagnetic one, whose properties are closest to those of a ferromagnet without OA.

²⁾In the present paper, just as in Refs. 14 and 15, $S \pm = \mp (1/\sqrt{2}) (S^{*} \pm iS^{*})$, so that $(S^{-})^{+} = -S^{+}$. Similarly, $(O^{-1})^{+} = -O^{1}, (O^{2})^{+} = O^{-2}$ [see Ref. 15, Eqs. (2)-(6)].

³⁾Many properties of this phase are similar to those of a quadrupole-ordered phase with biquadratic exchange (e.g., Refs. 20 and 21). The essential difference is that the quadrupole order in the latter is cooperative, and in the presence of OA it is induced by an "external quadrupole field"—the OA field.

⁴⁾The coefficients $H(\varphi, K)$, $D_2(\varphi, K)$ in (33) are defined by Eqs. (7) of Ref. 9.

- ³M. Tanaka and J. Kondo, Progr. Theor. Phys. 48, 1815 (1972).
- ⁴R. O. Zaĭtsev, Zh. Eksp. Teor. Fiz. **68**, 207 (1975) [Sov. Phys. JETP **41**, 100 (1975)].
- ⁵A. A. Kazakov, Teor. Mat. Fiz. 23, 281 (1975).
- ⁶V. Val'kov and S. G. Ovchinnikov, Teor. Mat. Fiz. 50, 466 (1982).
- ⁷M. P. Kashchenko, N. F. Balakhonov, and L. V. Kurbatov. Zh. Eksp. Teor. Fiz. **64**, 391 (1973) [Sov. Phys. JETP **37**, 201 (1973)
- ⁸S. N. Martynov and A. F. Sadreev, *ibid.* **81**, 46 (1981) [**54**, 130 (1981)].
- ⁹F. P. Onufrieva, Fiz. Tverd. Tela (Leningrad) 23, 2664 (1981) [Sov. Phys. Solid State 23, 1562 (1981)].
- ¹⁰I. G. Gochev and V. M. Tsukernik, *ibid.* 15, 1963 (1973) [15, 1315 (1973)].
- ¹¹L. V. Filatova and V. M. Tsukernik, Fiz. Nizk. Temp. 7, 1423 (1981) [Sov. J. Low Temp. Phys. 7, 601 (1981)].
- ¹²A. V. Chubukov, Zh. Eksp. Teor. Fiz. 85, 1319 (1983) [Sov. Phys. JETP 58, 765 (1983)].
- ¹³L. M. Noskova, Fiz. Tverd. Tela (Leningrad 18, 1669 (1976) [Sov. Phys. Solid State 18, 969 (1976)].
- ¹⁴F. P. Onufrieva, Zh. Eksp. Teor. Fiz. **80**, 2372 (1981) [Sov. Phys. JETP **53**, 1241 (1981)].
- ¹⁵F. P. Onufrieva, Teor. Mat. Fiz. 54, 299 (1983).
- ¹⁶V. N. Kitaev, M. P. Kashchenko *et al.* Preprint, Physicotech. Inst. Low Temp. Ukr. Acad. Sci., Kharkov, June 1973. V. N. Kitaev, M. P. Kashchenko, and L. V. Kurbatov. Fiz. Tverd. Tela (Leningrad) **15**, 2292 (1973) [Sov. Phys. Solid State **15**, (1974)].
- ¹⁷V. G. Vaks, A. I. Larkin, and S. A. Pikin, Zh. Eksp. Teor. Fiz. 53, 1089, 381 (1967) [Sov. Phys. JETP 26, 1530 (1968)].
- ¹⁸Yu. A. Izyumov, F. A. Gassan-Ogly, and Yu. N. Skryabin, Polevye metody v teorii ferromagnetizma [Field Theories in Ferromagnetism Theory), Nauka, 1974.
- ¹⁹B. R. Judd, Operator Techniques in Atomic Spectroscopy, McGraw-Hill, 1963.
- ²⁰F. P. Onufrieva, Fiz. Nizk. Temp. **3**, 1052 (1977) [Sov. J. Low Temp. Phys. **3**, 512 (1977)].
- ²¹E. L. Nagaev, Usp. Fiz. Nauk **136**, 61 (1982) [Sov. Phys. Usp. **25**, 31 (1982)].
- ²²T. Holstein and H. Primakoff, Phys. Rev. 58, 1098 (1940).
- ²³E. A. Turov, Physical Properties of Magnetically Ordered Crystals, Academic, 1965.

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¹S. L. Ginzburg, Fiz. Tverd. Tela (Leningrad) **12**, 1805 (1970) [Sov. Phys. Solid State **12**, 1429 (1970)].

²N. A. Potapkov, Teor, Mat. Fiz. 8, 381 (1971). L. M. Noskov, Fiz. Met. Metalloved. 33, 698 (1972).