Quantum spin-wave theory of ferromagnets with arbitrary single-ion anisotropy

V. V. Val'kov, T. A. Val'kova, and S. G. Ovchinnikov

Physics Institute, Siberian Division, USSR Academy of Sciences
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A diagram technique for Hubbard operators is used to develop a quantum theory of Heisenberg ferromagnets, with exact account taken of single-ion anisotropy of arbitrary symmetry. The Larkin equations and explicit expressions for the effective interactions were obtained. A dispersion equation that describes all the branches of the spin-wave excitation of an anisotropic ferromagnet is derived. The cases of easy-axis, easy-plane, and cubic anisotropy are considered. It is shown that the quantum spectrum of a ferromagnet with spin $S = 2$ and with cubic anisotropy consists in the low-temperature region of one longitudinal branch and two transverse ones. The correction to the ground state, necessitated by the zero-point quantum oscillations, as well as the low-temperature dependence of the magnetization, are calculated to first order in $1/r_0^3$.

The results of quantum-mechanical and phenomenological treatments of easy-axis ferromagnets do not agree, generally speaking, even in the simplest geometry, when the external magnetic field **H** is oriented along the anisotropy axis (a quantum description leads, for example, to new branches in the spectrum of the spin-wave excitations[1–5]). The difference increases for easy-plane ferromagnets, as well as at arbitrary direction of **H**, when the orientation of the equilibrium magnetization does not agree with that of the anisotropy axis.[6,7] In this geometry, a purely quantum "spin cancellation" effect manifests itself as a decrease of the magnetization of an anisotropic ferromagnet[8] (or of the magnetization of an antiferromagnet sublattice[9,10]). The effect increases with increasing anisotropy, so that an adequate description of strongly anisotropic magnets calls for a quantum treatment.

Quantum effects should manifest themselves to a greater degree in ferromagnets of higher symmetry (say, cubic), since the operators of the Zeeman energy and of the single-ion anisotropy do not commute at arbitrary orientation of H. At the same time, development of a quantum theory for $S \geqslant 2$ (spin values starting with which single-ion anisotropy of cubic symmetry is possible) is seriously hindered by the large number of single-ion states. In an approach formulated in Ref. 11, single-ion anisotropy of arbitrary symmetry is taken into account by including it in a single-site Hamiltonian, and the intersite exchange interaction is considered in the anisotropic molecular field approximation.

It has been found that the physical properties of anisotropic ferromagnets can be conveniently described by representing the spin operators in terms of Hubbard operators.[12–14] The diagram technique developed in Ref. 13 for Hubbard operators has made it possible to calculate, in the case of easy-axis ferromagnets with arbitrary spin S, the corrections $\propto r_0^{-3}$ to the spectrum and to the damping of magnons, as well as the contributions $\propto r_0^{-6}$ of the magnon-magnon interaction to the temperature dependence of the free energy and of the magnetization.[15] The theory of a uniaxial ferromagnet with collinear structure is thus constructed by the self-consistent field (SCF) method, in analogy with the theory of Vaks, Larkin, and Pikin for isotropic magnets.[16–18] The question of development of the theory of ferromagnets with arbitrary form of single-ion anisotropy has remained open.

We develop here a quantum spin-wave theory of ferromagnets with single-ion anisotropy of arbitrary symmetry, which is included in the Hamiltonian $\mathscr{H}_0(f)$. It is shown that the exchange-interaction matrix is represented as a sum of three matrices with separated dependences on the indices (see below). This circumstance was decisive in the derivation of the Larkin equations, from which a dispersion equation was obtained to describe the total spin-wave spectrum of anisotropic ferromagnets. The theory developed has made it possible to consider quantum spin-wave singularities of a cubic ferromagnet, and to determine its spectrum and the quantum spin cancellation. The temperature dependence of the magnetization and ground-state correction connected with the zero-point oscillations are calculated in first order in $1/r_0^3$.

1. HEISENBERG-FERROMAGNET HAMILTONIAN WITH SINGLE-ION ANISOTROPY (GENERAL CASE)

Consider a Heisenberg ferromagnet with arbitrary form of single-ion anisotropy (uniaxial, cubic etc.) and with a general direction of the external magnetic field **H**. The Hamiltonian of such a system can be written in the form

$$\mathscr{H} = \frac{1}{2} NI_0(\langle S^x\rangle^2+\langle S^y\rangle^2+\langle S^z\rangle^2) - \frac{1}{2}\sum_{fm} I_{fm}(\Delta \mathbf{S}_f)(\Delta \mathbf{S}_m) + \sum_f \mathscr{H}_0(\mathbf{S}_f; \overline{H}_x, \overline{H}_y, \overline{H}_z) \quad (\Delta \mathbf{S}_f = \mathbf{S}_f - \langle \mathbf{S}_f\rangle), \tag{1}$$

where the last term describes the influence of the single-ion anisotropy, of the external magnetic field, and of the self-consistent field $I_0\langle S\rangle$

$$\overline{H}_i = H_i + I_0\langle \mathbf{S}^i\rangle \quad (i = x, y, z). \tag{2}$$

The solution of the single-ion problem with Hamiltonian $\mathscr{H}_0(f)$ leads to diagonalization of the last term of (1) if the spin operators are represented in terms of Hubbard operators

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$$\sum_f \mathcal{H}_0(S_f; \overline{H}_x, \overline{H}_y, \overline{H}_z) \equiv \sum_f \mathcal{H}_0(f) = \sum_{fn} E_n h_{fn} = \mathcal{H}_0, \qquad (3)$$

where E_n is the self-energy of the Hamiltonian $\mathcal{H}_0(f)$ in the nth quantum state $|\psi_{fn}\rangle$ of site f, $h_{fn} = X_f^{n,n}$, $X_f^{nn'} = |\psi_{fn}\rangle\langle\psi_{fn'}|$ are Hubbard operators constructed on the complete basis of the states $|\psi_{fn}\rangle$.

At arbitrary geometry of the problem, the representation of the spin operators S_f^+, S_f^- and S_f^z in terms of the Hubbard operators takes the form[19]

$$S_f^+ = \sum_\alpha \gamma_\perp(\alpha) X_f^\alpha + \sum_n \Gamma_\perp(n) h_{fn}, \quad S_f^- = (S_f^+)^+, \qquad (4)$$

$$S_f^z = \sum_n \Gamma_\parallel(n) h_{fn} + \sum_\alpha \gamma_\parallel(\alpha) X_f^\alpha. \qquad (5)$$

Here α is the root vector (13) whose definition, as in Ref. 20, follows from the relations

$$X_f^{n,m} \equiv X_f^{\alpha(n,m)} \equiv X_f^\alpha.$$

For a system with spin S, the total number of root vectors α is $2S(2S+1)$. Each root vector α has $2S+1$ components and at the present definition of α the qth component can be represented in the simple form

$$\alpha_q(n, m) = \delta_{qn} - \delta_{qm}. \qquad (6)$$

It must be noted that in Refs. 13, 21, and 22 zero-trace atomic operators are used to construct the diagram technique, whereas we use nonzero-trace Hubbard operators. One of the advantages of our approach, particularly convenient for the case of arbitrary spin, is the ease of calculating the root-vector components. On the whole, the physical results of both approaches are the same.

It follows from the general relations of quantum mechanics[23] that the coefficients $\Gamma_\perp(n)$ and $\Gamma_\parallel(n)$ of the transformations (4) and (5) are connected with the derivatives of the energies E_n with respect to the fields $\overline{H}_i$:

$$\frac{\partial E_n}{\partial \overline{H}_x} = -\frac{1}{2}[\Gamma_\perp(n) + \Gamma_\perp^*(n)],$$

$$\frac{\partial E_n}{\partial \overline{H}_y} = \frac{i}{2}[\Gamma_\perp(n) - \Gamma_\perp^*(n)], \qquad (7)$$

$$\frac{\partial E_n}{\partial \overline{H}_z} = -\Gamma_\parallel(n).$$

From the hermiticity of the spin operators and from the commutation rules for them[23] follow the relations

$$\gamma_\parallel(\alpha) = \gamma_\parallel^*(-\alpha), \quad \Gamma_\parallel(n) = \frac{1}{2}\sum_\alpha \alpha_n |\gamma_\perp(\alpha)|^2,$$

$$\Gamma_\perp(n) = \sum_\alpha \alpha_n \gamma_\parallel(\alpha)\gamma_\perp(-\alpha), \qquad (8)$$

$$\Gamma_\perp^*(n) = -\sum_\alpha \alpha_n \gamma_\parallel(\alpha)\gamma_\perp^*(\alpha).$$

Substituting (4) and (5) in the initial Hamiltonian (1), we get

$$\mathcal{H} = \frac{NI_0}{2}(\langle S^x\rangle^2 + \langle S^z\rangle^2 + \langle S^y\rangle^2) + \mathcal{H}_0 + \mathcal{H}_{int}, \qquad (9)$$

where

$$\mathcal{H}_{int} = -\frac{1}{2}\sum_{fm}\sum_{\alpha\beta} I_{fm}^{\alpha\beta} \Delta X_f^\alpha \Delta X_m^\beta - \sum_{fm}\sum_{\alpha n} I_{fm}^{\alpha n} \Delta X_f^\alpha \Delta h_{mn'} - \frac{1}{2}\sum_{fm}\sum_{nn'} I_{fm}^{nn'} \Delta h_{fn} \Delta h_{mn'}. \qquad (10)$$

The interaction-matrix components in (10) are given by

$$I_{fm}^{\alpha\beta} = I_{fm}\left\{\frac{1}{2}[\gamma_\perp(\alpha)\gamma_\perp^*(-\beta) + \gamma_\perp^*(-\alpha)\gamma_\perp(\beta)] + \gamma_\parallel(\alpha)\gamma_\parallel(\beta)\right\}, \qquad (11)$$

$$I_{fm}^{\alpha n} = I_{fm}\left\{\frac{1}{2}[\gamma_\perp(\alpha)\Gamma_\perp^*(n) + \gamma_\perp^*(-\alpha)\Gamma_\perp(n)] + \Gamma_\parallel(n)\gamma_\parallel(\alpha)\right\}, \qquad (12)$$

$$I_{fm}^{nn'} = I_{fm}\left\{\frac{1}{2}[\Gamma_\perp(n)\Gamma_\perp^*(n') + \Gamma_\perp^*(n)\Gamma_\perp(n')] + \Gamma_\parallel(n)\Gamma_\parallel(n')\right\}. \qquad (13)$$

It follows from the foregoing explicit dependence of the matrix components on the indices that we are dealing in fact with a sum of three matrices, in each of which the index has a split character. This enables us to find the explicit forms of Larkin's equations and of the expressions for the effective interactions.

2. LARKIN'S EQUATIONS FOR A FERROMAGNET WITH SINGLE-ION ANISOTROPY OF ARBITRARY SYMMETRY

The dynamic and thermodynamic properties of an anisotropic ferromagnet can be investigated with the aid of the Matsubara[24] Green's functions

$$D_{\lambda\lambda'}(f\tau; g\tau') = -\langle T_\tau \tilde{A}_f^\lambda(\tau) \tilde{B}_g^{\lambda'}(\tau')\rangle, \qquad (14)$$

where the indices λ and λ' take on values from the following ordered set of $(2S+1)^2$ elements:

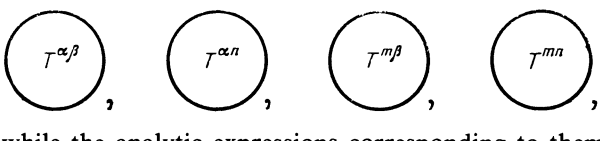
$$\alpha_1, \alpha_2, \ldots, \alpha_{2S(2S+1)}, \quad n_1, n_2, \ldots, n_{2S+1}. \qquad (15)$$

The operators $A_f^\lambda(\tau)$ and $B_g^{\lambda'}(\tau')$ are defined by the relations

$$\tilde{A}_f^\lambda(\tau) = \begin{cases} \tilde{X}_f^\alpha(\tau), & \lambda = \alpha \\ \tilde{h}_{fn}(\tau), & \lambda = n \end{cases}; \quad \tilde{B}_g^{\lambda'}(\tau') = \begin{cases} \tilde{X}_g^{-\alpha}(\tau'), & \lambda' = \alpha, \\ \tilde{h}_{gn}(\tau'), & \lambda' = n, \end{cases}$$

and are taken in the Heisenberg representation.[24]

It is convenient to calculate the functions (14) by the diagram method for Hubbard operators.[13] Just as in Refs. 16–18 we call a diagram irreducible if it cannot be cut along one interaction line into two unconnected parts (irreducibility after Larkin). The total aggregates of irreducible diagrams for the functions (14) are called the irreducible parts and are correspondingly represented as

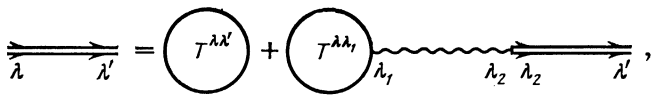


while the analytic expressions corresponding to them are designated by

$$T^{\alpha\beta}, T^{\alpha n}, T^{m\beta}, T^{mn}. \qquad (16)$$

In diagrammatic form, the system of equations for the

complete Green's functions can be written in the (k, ω_n) representation in the form



(17)

where the double line $\lambda\lambda'$ corresponds to $D_{\lambda\lambda'}(\mathbf{k}, \omega_n)$, and the wavy line corresponds to the bare interaction $I_k^{\lambda\lambda'}$ (11–13). The split character of expressions (11)–(13) allows us to reduce the solution of the system of linear equations (17) of order $(2S+1)^4$ to the solution of a third-order system and obtain the explicit connection between $D_{\lambda\lambda'}$ and $T^{\lambda\lambda'}$. Omitting the intermediate operations, we write down the Larkin equation in the form

$$D_{\lambda\lambda'}(\mathbf{k}, \omega_n) = -T^{\lambda\lambda'}(\mathbf{k}, \omega_n) + \frac{I_{\mathbf{k}}}{2\Delta(\mathbf{k}, \omega_n)} \sum_{i=1}^{3} (e_\lambda \hat{T}(\mathbf{k}, \omega_n) A_i) \Delta_i^{\lambda'}(\mathbf{k}, \omega_n), \tag{18}$$

where the matrix $\hat{T}(\mathbf{k}, \omega_n)$ has the following block structure:

$$\hat{T}(\mathbf{k}, \omega_n) = \begin{pmatrix} T^{(\alpha\beta)}(\mathbf{k}, \omega_n), & T^{(\alpha n)}(\mathbf{k}, \omega_n) \\ T^{(m,\beta)}(\mathbf{k}, \omega_n), & T^{(mn)}(\mathbf{k}, \omega_n) \end{pmatrix} \tag{19}$$

with matrices $T^{(\lambda\lambda')}(\mathbf{k}, \omega_n)$ whose elements are the irreducible parts of $T^{\lambda\lambda'}(\mathbf{k}, \omega_n)$ (16), and e_λ is a unit $(2S+1)^2$-dimensional vector whose only nonzero component is the one whose number is that of the element λ in (15). The $(2S+1)^2$-dimensional vectors A_i take in block notation the form

$$A_1 = 2\begin{pmatrix} \gamma_\parallel(-\beta) \\ \Gamma_\parallel(n) \end{pmatrix}, \quad A_2 = \begin{pmatrix} \gamma_\perp^*(\beta) \\ \Gamma_\perp^*(n) \end{pmatrix}, \quad A_3 = \begin{pmatrix} \gamma_\perp(-\beta) \\ \Gamma_\perp(n) \end{pmatrix}. \tag{20}$$

The third-order determinant $\Delta(\mathbf{k}, \omega_n)$ is equal to

$$\Delta(\mathbf{k}, \omega_n) = \det\| \delta_{ij} - {}^1/_2 I_{\mathbf{k}} B_i \hat{T}(\mathbf{k}, \omega_n) A_j \|, \tag{21}$$

where the vectors B_i have, in a notation analogous to (20), the form

$$B_1 = \begin{pmatrix} \gamma_\parallel(\beta) \\ \Gamma_\parallel(n) \end{pmatrix}, \quad B_2 = \begin{pmatrix} \gamma_\perp(\beta) \\ \Gamma_\perp(n) \end{pmatrix}, \quad B_3 = \begin{pmatrix} \gamma_\perp^*(-\alpha) \\ \Gamma_\perp^*(n) \end{pmatrix}. \tag{22}$$

The determinants $\Delta_i^\lambda(\mathbf{k}, \omega_n)$ are obtained from $\Delta(\mathbf{k}, \omega_n)$ by replacing in the latter the ith column by a column made up of the elements

$$-B_1 \hat{T}(\mathbf{k}, \omega_n) e_{\lambda'}, \quad -B_2 \hat{T}(\mathbf{k}, \omega_n) e_{\lambda'}, \quad -B_3 \hat{T}(\mathbf{k}, \omega_n) e_{\lambda'}. \tag{23}$$

A distinguishing feature of an anisotropic ferromagnet is, generally speaking, the connection of each Green's function with all the irreducible parts, each of which can be calculated by expansion in the reciprocal interaction radius.[16–18] This is due to the existence of the longitudinal-transverse interaction (12). By making the analytic continuation $i\omega_n \to \omega$ we find that the spectrum of the spin-wave excitations is determined from the equation

$$\Delta(\mathbf{k}, i\omega_n \to \omega) = 0. \tag{24}$$

The number of the collective branches of the spin-wave spectrum depends on the form of the anisotropy and on the concrete geometries of the problems, as is illustrated below by a number of examples [see (31)–(33) and (39)]. The maximum number of collective branches is equal to the number of the different intra-atomic transitions $2S(2S+1)$.

3. ZEROTH APPROXIMATION IN THE RECIPROCAL INTERACTION RADIUS

In this approximation we have

$$T^{\alpha m}(\mathbf{q}, \omega_n) = T^{m\beta}(\mathbf{q}, \omega_n) = 0, \quad T^{ml}(\mathbf{q}, \omega_n) = \frac{b_{ml}}{T} \delta_{\omega_n, 0},$$
$$T^{\alpha\beta}(\mathbf{q}, \omega_n) = -\delta_{\alpha\beta} D_\alpha(\omega_n) b(\alpha), \quad b(\alpha) = \sum_m \alpha_m N_m, \tag{25}$$

where T is the temperature,

$$b_{ml} = \delta_{ml} N_m - N_l N_m,$$
$$N_m = \exp(-E_m/T)/Z_0, \quad Z_0 = \sum_m \exp(-E_m/T),$$
$$D_\alpha(\omega_n) = (i\omega_n + \alpha E)^{-1}, \quad \omega_n = 2\pi n T. \tag{26}$$

The determinant $\Delta(\mathbf{k}, \omega_n)$ takes in this case the simpler form

$$\Delta_0 = \begin{vmatrix} 1+x_1 & x_2 & x_3 \\ y_1 & 1+y_2 & y_3 \\ z_1 & z_2 & 1+z_3 \end{vmatrix}, \tag{27}$$

where

$$x_1 = I_{\mathbf{k}}[c(\omega_n) - B_{22}(\omega_n)], \quad x_2 = {}^1/_2 I_{\mathbf{k}}[a(\omega_n) - B_{12}^*(\omega_n)], \quad x_3 = x_2^*,$$
$$y_1 = I_{\mathbf{k}}[a^*(-\omega_n) - B_{12}(\omega_n)], \quad y_2 = {}^1/_2 I_{\mathbf{k}}[d(\omega_n) - C_{11}(\omega_n)],$$
$$y_3 = {}^1/_2 I_{\mathbf{k}}[f(\omega_n) - B_{11}(\omega_n)], \quad z_1 = y_1^*, \tag{28}$$
$$z_2 = y_3^*, \quad z_3 = {}^1/_2 I_{\mathbf{k}}[d(-\omega_n) - C_{11}(\omega_n)],$$

and the functions that depend on ω_n are

$$B_{ij}(\omega_n) = \sum_{ml} \Gamma_i(m) \Gamma_j(l) b_{ml} \delta_{\omega_n, 0}/T,$$
$$i = 1, 2, \quad \Gamma_1(n) \equiv \Gamma_\perp(n), \tag{29}$$
$$\Gamma_2(n) \equiv \Gamma_\parallel(n), \quad C_{11}(\omega_n) = \sum_{lm} \Gamma_\perp(m) \Gamma_\perp^*(l) \delta_{\omega_n, 0} b_{ml}/T;$$

$$a(\omega_n) = \sum_\alpha \gamma_\parallel(\alpha) \gamma_\perp^*(\alpha) D_\alpha(\omega_n) b(\alpha),$$
$$c(\omega_n) = \sum_\alpha |\gamma_\parallel(\alpha)|^2 D_\alpha(\omega_n) b(\alpha),$$
$$d(\omega_n) = \sum_\alpha |\gamma_\perp(\alpha)|^2 D_\alpha(\omega_n) b(\alpha), \tag{30}$$
$$f(\omega_n) = \sum_\alpha \gamma_\perp(\alpha) \gamma_\perp(-\alpha) D_\alpha(\omega_n) b(\alpha).$$

For easy-axis ferromagnets with a magnetic field oriented along the anisotropy axis we have[15] $\gamma_\parallel(\alpha) = 0$,

$\gamma_\perp(\alpha)\gamma_\perp(-\alpha)=0$, and the dispersion equation (24) becomes

$$\left(1+\frac{1}{2}I_k\sum_\alpha\frac{\gamma_\perp^2(\alpha)b(\alpha)}{\omega+\alpha E}\right)$$

$$\times\left(1-\frac{1}{2}I_k\sum_\alpha\frac{\gamma_\perp^2(\alpha)b(\alpha)}{\omega-\alpha E}\right)=0 \tag{31}$$

and determines, as in Ref. 1, the $2S$ collective branches of the spin-wave spectrum. We note that allowance for biquadratic exchange or for another multipole interaction would lead to collectivization of new transitions (e.g., two-magnon ones).

For easy-plane ferromagnets with a magnetic field oriented perpendicular to the anisotropy axis, the following relations hold:

$$\gamma_\perp(\alpha)\gamma_\parallel(\alpha)=0, \quad \Gamma_\perp(n)=0 \quad \text{for all} \quad \alpha, n.$$

Writing (24) out fully, we find that the spectrum of the longitudinal oscillations is obtained from the equations

$$1+I_k\sum_\alpha\frac{\gamma_\parallel^2(\alpha)b(\alpha)}{\omega+\alpha E}=0, \tag{32}$$

whereas the transverse branches of the spin-wave spectrum are solutions of the equation

$$\left[1+\frac{1}{2}I_k\sum_\alpha\frac{\gamma_\perp^2(\alpha)b(\alpha)}{\omega+\alpha E}\right]\left[1+\frac{1}{2}I_k\sum_\alpha\frac{\gamma_\perp^2(\alpha)b(\alpha)}{-\omega+\alpha E}\right] \tag{33}$$

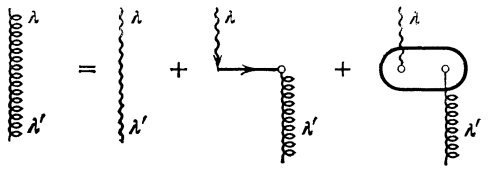
$$-\frac{1}{4}I_k^2\left[\sum_\alpha\frac{\gamma_\perp(\alpha)\gamma_\perp(-\alpha)b(\alpha)}{\omega+\alpha E}\right]^2=0.$$

In the particular case $S=1$ and $T\ll T_c$, Eqs. (32) and (33) yield the spectrum calculated in Ref. 13. For arbitrary S and T, Eqs. (32) and (33) were derived here for the first time.

We shall consider cubic ferromagnets separately, after calculating the effective interactions.

4. EFFECTIVE INTERACTIONS OF AN ANISOTROPIC FERROMAGNET

The existence of a bare longitudinal-transverse interaction (12) causes the longitudinal interaction (13) to contribute to the effective transverse interaction. A coupled system of equations is thus obtained for the effective interactions, in the graphic form



(34)

(if the index λ corresponds to a root vector, an arrow is implied at the end of the interaction line). Such a system of equations of order $(2S+1)^4$ can also be solved explicitly in view of the already noted split form of the bare interactions (11)–(13). After a number of transformations we find that the solution of (34) can be written in the form

$$\tilde{I}^{\lambda\lambda'}(\mathbf{q},\ \omega_n)=I_{\mathbf{q}}(\mathbf{c}_\lambda\hat{v}\mathbf{c}_{\lambda'})-I_{\mathbf{q}}(\mathbf{c}_\lambda\hat{v}\mathbf{R}_{\lambda'}(\mathbf{q},\ \omega_n)), \tag{35}$$

where the three-dimensional vectors $\mathbf{c}_\lambda$ are defined by the expressions

$$\mathbf{c}_\lambda=\begin{cases}[\gamma_\parallel(\alpha),\ \gamma_\perp(\alpha),\ \gamma_\perp^*(-\alpha)], & \lambda=\alpha,\\ [\Gamma_\parallel(n),\ \Gamma_\perp(n),\ \Gamma_\perp^*(n)], & \lambda=n,\end{cases} \tag{36}$$

and the matrix $\hat{v}$ is equal to

$$\hat{v}=\begin{pmatrix}1&0&0\\0&0&1/2\\0&1/2&0\end{pmatrix}. \tag{37}$$

The component numbered i ($i=1, 2, 3$) of the vector $\mathbf{R}_\lambda(\mathbf{q}, \omega_n)$ is written in the form

$$\mathbf{R}_\lambda^i(\mathbf{q},\ \omega_m)=\Delta_\lambda^i(\mathbf{q},\ \omega_m)/\Delta_0(\mathbf{q},\ \omega_m), \tag{38}$$

where the determinants $\Delta_\lambda^i(\mathbf{q}, \omega_m)$ are obtained from $\Delta_0(\mathbf{q}, \omega_m)$ defined by (27) by replacing in it the ith column by the column $(\mathbf{c}_\lambda\mathbf{x}, \mathbf{c}_\lambda\mathbf{y}, \mathbf{c}_\lambda\mathbf{z})$, in which the three-dimensional vectors $\mathbf{x}$, $\mathbf{y}$, and $\mathbf{z}$ are defined by their components (28). Knowing the explicit expressions for the effective interactions and for the Green's functions in the zeroth approximation in $1/r_0^3$ of the SCF method, we can deduce an actual analytic expression with each diagram and develop by the same token a regular procedure for calculating the physical quantities in the form of expansions in powers of the small quantities T/T_c or $1/r_0^3$.

5. FERROMAGNETS WITH SINGLE-ION ANISOTROPY OF CUBIC SYMMETRY

We confine ourselves to the case when the magnetic field is directed along the C_4 axis, which is simultaneously the easy-magnetization axis.[25,26] In this case (see Ref. 10) $\gamma_\perp(\alpha)\gamma_\parallel(\alpha)=0$, $\gamma_\perp(\alpha)\gamma_\perp(-\alpha)=0$ and from (31) we find that the equation that determines the spectrum of the longitudinal oscillations has the same form as (32), and the branches of the transverse collective excitations satisfy the equation

$$\left[1+\frac{1}{2}I_k\sum_\alpha\frac{\gamma_\perp^2(\alpha)b(\alpha)}{\omega+\alpha E}\right]$$

$$\times\left[1+\frac{1}{2}I_k\sum_\alpha\frac{\gamma_\perp^2(\alpha)b(\alpha)}{-\omega+\alpha E}\right]=0. \tag{39}$$

The number of branches of the longitudinal and transverse excitations is determined by the number of nonzero parameters of the representation $\gamma_\parallel(\alpha)$ and $\gamma_\perp(\alpha)$. At $T\ll T_c$ the degrees of Eqs. (32) and (39) are lower because of the exponential smallness of the terminal factors $b(\alpha)$ for those $\alpha(n, m)$ in which neither n nor m is an index of the ground state.

Let us consider in greater detail a cubic ferromagnet with $S=2$, inasmuch as at $S<2$ the energy of single-ion anisotropy of cubic symmetry becomes constant[27] and magnetic-anisotropy effects are possible only through anisotropic interaction between the atoms. From the solution obtained in Ref. 19 for the single-ion problem it follows that in the region $T\ll T_c$ the following parameters are of importance:

$$\gamma_\parallel(1,5)=\gamma_\parallel(5,1)=-2\sin 2\theta(\gamma(n,\ m)\equiv\gamma[\alpha(n,\ m)]),$$
$$\gamma_\perp(4,1)=2\sin\theta,\ \gamma_\perp(1,2)=2\cos\theta, \tag{40}$$

where

$$\cos\theta=\frac{U}{(K^2+U^2)^{1/2}},\quad \sin\theta=\frac{K}{(K^2+U^2)^{1/2}},$$
$$U=2\bar{H}+(4\bar{H}^2+K^2)^{1/2},\quad \bar{H}=H+I_0\langle S^z\rangle, \tag{41}$$

and K is the cubic-anisotropy constant.[25,26] Substituting expressions (40) in (32), we find that the dispersion law of the longitudinal branch of the collective excitations is of the form

$$\omega_{\parallel}(\mathbf{q})=(\varepsilon_{51}^2-8\sin^2 2\theta\varepsilon_{51}I_{\mathbf{q}})^{1/2},\quad \varepsilon_{51}=2(4\bar{H}^2+K^2)^{1/2}. \tag{42}$$

At $H=0$, the gap in the spectrum of this branch is equal to

$$\Delta_{\parallel}(H=0)=4I_0S_0,\quad S_0=2(1-(K/4I_0)^2)^{1/2} \tag{43}$$

and at $K=4I_0$ it vanishes. This means that a restructuring of the ground state will occur in the vicinity of $K=4I_0$. We therefore confine ourselves hereafter to the region $K<4I_0$. It follows from the self-consistency equation for $\langle S_z\rangle$ that at $H\ll 0$ the average magnetization, accurate to terms linear in H/I_0, is equal to

$$\langle S^z\rangle=2\cos 2\theta=S_0+(H/I_0)(K/4I_0)^2(2/S_0)^2. \tag{44}$$

We find then from (42) and (41) that at $H\ll I_0$

$$\omega_{\parallel}(\mathbf{q})=(\Delta_{\parallel}^2+(4K^2/I_0^2)(I_0-H/S_0)(I_0-I_{\mathbf{q}}))^{1/2}, \tag{45}$$

where

$$\Delta_{\parallel}=4I_0S_0+4H+6H(K/4I_0)^2(2/S_0)^2.$$

From (39) we find that the spectrum of the transverse oscillations consists of two branches

$$\omega_{1,2}=[((\varepsilon_{41}+\varepsilon_{21})/2-I_{\mathbf{q}})^2-I_{\mathbf{q}}^2\sin^2 2\theta]^{1/2}$$
$$\mp(\varepsilon_{41}-\varepsilon_{21}+2I_{\mathbf{q}}\cos 2\theta)/2, \tag{46}$$

where

$$\varepsilon_{41}=\bar{H}+K+(4\bar{H}^2+K^2)^{1/2},\quad \varepsilon_{21}=-\bar{H}+K+(4\bar{H}^2+K^2)^{1/2}. \tag{47}$$

The first term in (46) in the radicand is always substantially larger than $I_{\mathbf{q}}^2\sin^2 2\theta$, therefore the spectrum of the transverse spin-wave excitations can be represented in the simpler form

$$\omega_1(q)=\varepsilon_{21}-I_{\mathbf{q}}(1+\cos 2\theta)-I_{\mathbf{q}}^2\sin^2 2\theta/(\varepsilon_{41}+\varepsilon_{21}-2I_{\mathbf{q}}), \tag{48}$$

$$\omega_2(q)=\varepsilon_{41}-I_{\mathbf{q}}(1-\cos 2\theta)-I_{\mathbf{q}}^2\sin^2 2\theta/(\varepsilon_{41}+\varepsilon_{21}-2I_{\mathbf{q}}). \tag{49}$$

At $H\ll I_0$ we obtain from (47)

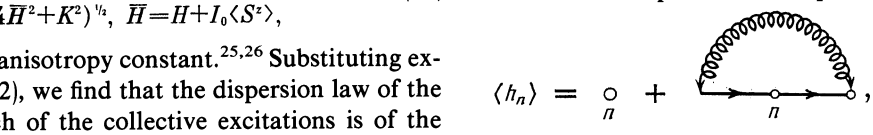
$$\varepsilon_{21}=I_0(4-S_0)+K+4H(1-1/S_0)/S_0,$$
$$\varepsilon_{41}=I_0(4+S_0)+K+4H(1+1/S_0)/S_0, \tag{50}$$

from which it follows that at small K the gap in the spectrum of the first branch of the collective excitations is equal to $K+H$, whereas for the second branch $\Delta_2=6I_0+K+3H$.

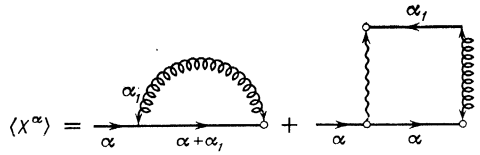
Let us calculate in first order in $1/r_0^3$ of the SCF method the low-temperature magnetization and the correction to the ground state for the zero-point quantum oscillations. It follows from (5) that

$$\langle S^z\rangle=\sum_n\Gamma_{\parallel}(n)\langle h_n\rangle+\sum_{\alpha}\gamma_{\parallel}(\alpha)\langle X^{\alpha}\rangle, \tag{51}$$

and the calculation of $\langle S_z\rangle$ reduces to calculation of the mean values of Hubbard operators. At $T\ll T_c$ and $K<4I_0$, in the approximation considered here, the mean values of diagonal Hubbard operators are represented by the diagrams

$\langle h_n\rangle =$  (52)

whereas the diagrams for $\langle X^{\alpha}\rangle$ are of the form

$\langle X^{\alpha}\rangle =$  (53)

The analytic expressions for the effective interactions $\tilde{I}^{\alpha\beta}(\mathbf{q}, \omega_n)$ in the diagrams (52) and (53) can be easily calculated from Eqs. (35)–(38). For our case we obtain

$$\tilde{I}^{\alpha\beta}(\mathbf{q},\omega_n)=\gamma_{\parallel}(\alpha)\gamma_{\parallel}(\beta)I_{\mathbf{q}}\frac{(i\omega_n)^2-\varepsilon_{51}^2}{(i\omega_n)^2-\omega_{\parallel}^2}$$
$$+\frac{1}{2}I_{\mathbf{q}}\Big\{\gamma_{\perp}(-\alpha)\gamma_{\perp}(\beta)\frac{(i\omega_n+\varepsilon_{41})(i\omega_n-\varepsilon_{21})}{(i\omega_n+\omega_2)(i\omega_n-\omega_1)}$$
$$+\gamma_{\perp}(\alpha)\gamma_{\perp}(-\beta)\frac{(i\omega_n-\varepsilon_{41})(i\omega_n+\varepsilon_{21})}{(i\omega_n-\omega_2)(i\omega_n+\omega_1)}\Big\}. \tag{54}$$

Setting (52) and (53) in correspondence with the analytic expression, we find from (51)–(53) that

$$\langle S^z\rangle=2\cos 2\theta-\frac{2}{N}\sum_{\mathbf{q}}I_{\mathbf{q}}\Big\{\frac{(2\cos 2\theta-1)\cos^2\theta(\varepsilon_{41}-\omega_2)}{(\omega_2+\varepsilon_{21})(\omega_1+\omega_2)}$$
$$+\frac{4\sin 2\theta\sin 4\theta}{\omega_{\parallel}}+\frac{(2\cos 2\theta+1)\sin^2\theta(\varepsilon_{21}-\omega_1)}{(\omega_1+\varepsilon_{41})(\omega_1+\omega_2)}$$
$$+\frac{4\cos 2\theta\sin^2\theta(\varepsilon_{51}-\omega_{\parallel})}{\omega_{\parallel}(\omega_{\parallel}+\varepsilon_{51})}-\frac{2\sin^2 2\theta}{\varepsilon_{51}}\frac{\omega_1-\omega_2+\varepsilon_{42}}{\omega_1+\omega_2}\Big\}$$
$$-\frac{2}{N}\sum_{\mathbf{q}}\Big\{\frac{(2\cos 2\theta+1)\sin^2\theta(\varepsilon_{21}-\omega_1)}{(\omega_1+\varepsilon_{41})(\omega_1+\omega_2)}-\frac{\sin^2 2\theta}{\varepsilon_{51}}\frac{2\omega_1+\varepsilon_{42}}{\omega_1+\omega_2}$$
$$+\frac{(2\cos 2\theta-1)\cos^2\theta(\omega_1+\varepsilon_{41})}{(\varepsilon_{21}-\omega_1)(\omega_1+\omega_2)}\Big\}I_{\mathbf{q}}f_B\Big(\frac{\omega_1}{T}\Big), \tag{55}$$

where $f_B(x)$ is the Bose-Einstein function. The first term of (55) describes the behavior of $\langle S_z\rangle$ with change of the system parameters in the zeroth approximation in $1/r_0^3$ of the SCF method. The temperature-independent sum over q describes the first-order correction in $1/r_0^3$ to the ground state of the system, necessitated by the presence of zero-point quantum oscillations. The last sum over $\mathbf{q}$ determines, in the approximation considered, the temperature dependence of the magnetization. We have left out of expression (55) for $\langle S_z\rangle$ the terms containing Bose-Einstein functions of the form

$$f_B(\omega_2/T),\ f_B(\omega_{\parallel}/T),\ f_B(\varepsilon_{21}/T),\ f_B(\varepsilon_{41}/T),\ f_B(\varepsilon_{51}/T),$$

since they all lead to a temperature dependence with an activation energy substantially higher than that in the ω_1 spectrum if the anisotropy parameter satisfies the condition

$$0\leqslant K\lesssim 3I_0. \tag{56}$$

To simplify the final expression, we can use in this range of K

the small parameter

$$\varepsilon = I_0 \sin^2 2\theta / (\varepsilon_{41} + \varepsilon_{21} - 2I_0), \quad (57)$$

where the functions that depend on $\langle S_z \rangle$ are taken at the magnetization value determined by the zeroth approximation of the SCF method [see Eq. (44)].

Calculating the integrals with respect to $\mathbf{q}$ in (55), we get

$$\langle S^z \rangle = 2\cos 2\theta + \delta S^z(0) + \delta S^z(T), \quad (58)$$

where the correction $\delta S_z(0)$ takes, accurate to terms linear in ε, the form

$$\delta S^z(0) \approx -(\cos 2\theta / r_0^3)\,((4+W)/W\cos^2\theta + 4W\sin^2\theta)\,\varepsilon, \quad (59)$$

where

$$W = (\varepsilon_{41} + \varepsilon_{21})/I_0 - 2.$$

The temperature dependence of the magnetization ($T \ll T_c$) is given by

$$\delta S^z(T) = -\frac{\alpha}{r_0^3}\left(\frac{3T}{4\pi I_0}\right)^{3/2} Z_{3/2}\left(\frac{\Delta}{T}\right) + O\left(T^{5/2} Z_{5/2}\left(\frac{\Delta}{T}\right)\right), \quad (60)$$

where α was calculated in the same approximation linear in ε:

$$\alpha = (2\cos 2\theta - 1 + a\varepsilon)/\cos^3\theta\cos^2 2\theta,$$

$$a = \cos 2\theta\,\frac{W+4}{W} - \frac{3(\cos 2\theta - 1)}{2\cos^2\theta} - \frac{1}{2}W, \quad (61)$$

$$\Delta = \varepsilon_{21} - 2I_0\cos^2\theta - I_0\varepsilon.$$

Expresion (60) shows that the presence of cubic anisotropy affects the temperature dependence of the magnetization not only through the appearance of the function

$$Z_\alpha(x) = \sum_{n=1}^{\infty} e^{-nx}/n^\alpha,$$

as in easy-axis ferromagnets,[25] but also through the appearance of an additional factor α that depends on the anisotropy, with $\alpha \to 1$ as $K \to 0$. This factor, as well as the quantum cancellation of the spin, appears because the Zeeman-energy and the single-ion anisotropy energy do not commute. It follows from (60) that at $T \ll \Delta$, when $Z_{3/2}(\Delta/T) \approx \exp(-\Delta/T)$, the temperature dependence of the magnetization contains, as is usual in anisotropic ferromagnets, an activation factor besides the factor $T^{3/2}$. In the region $\Delta \ll T \ll T_c$, however, when $Z_{3/2}(\Delta/T) = \zeta(3/2) - 2(\pi\Delta/T)^{1/2} + \ldots$, we find that

$$\delta S^z(T) = -\frac{\alpha}{r_0^3}\zeta(3/2)\left(\frac{3T}{4\pi I_0}\right)^{3/2} + \frac{\alpha}{r_0^3}\left(\frac{3\Delta}{I_0}\right)^{1/2}\left(\frac{3T}{4\pi I_0}\right) - \ldots,$$

i.e., that a term linear in T is present besides the Bloch term. These singularities should manifest themselves primarily in ferromagnets with large anisotropy $K \sim I_0$, so that the singularities predicted in this paper might be observed, e.g., in rare-earth compounds, in most of which the anisotropy plays the principal role.[28]

6. CONCLUSION

Many workers have attempted to take exact account of the single-ion energy when developing a theory for anisotropic magnets, and have succeeded in particular cases.[8-11] Moreover, there exist a general formalism, making use of unitary symmetry, which permits exact construction of a single-ion states at arbitrary geometry of the system, as well as a realization of this formalism for $S = 1$ (Ref. 29). Up to now, however, there was no consistent method of taking into account the interatomic interactions between single-ion states with arbitrary nonequidistant spectrum. The use of a diagram technique for Hubbard's atomic operators, as noted in Ref. 30, provides such a method, and such a program was implemented in the present paper for Heisenberg ferromagnets with arbitrary form of single-ion anisotropy. Anisotropic multisublattice magnets can be considered similarly.

A diagram technique for arbitrary single-ion anisotropy and for an arbitrary geometry was developed recently.[31] For $S = 1$ and for the easy-axis and easy-plane cases considered above, the spectrum of the excitations obtained in Ref. 31 coincides with our results and with earlier results by others. At the same time, as noted in Ref. 31 by the author himself, his method is difficult to apply to cases with $S > 1$, but in our approach there are no spin restrictions.

Note added in proof (26 December 1984). A diagram technique for the Hubbard operators was used in the theory of uniaxial magnetic systems with arbitrary S by D. A. Garanin and V. S. Lutovinov [Teor. Mat. Fiz. **55**, 106 (1983); Sov. Phys. JETP. **58**, 1194 (1983); Sov. Phys. Sol. State **26**, 1706 (1984)].

[1] S. L. Ginzburg, Fiz. Tverd. Tela (Leningrad) **12**, 1805 (1970) [Sov. Phys. Solid State **12**, 1429 (1971).
[2] N. A. Potapkov, Teor. Mat. Fiz. **8**, 381 (1971).
[3] L. M. Noskova, Fiz. Met. Metallov. **33**, 698 (1972).
[4] T. K. Devlin, Phys. Rev. **B4**, 136 (1971).
[5] M. Tanako and Y. Kondo, Progr. Theor. Phys. **48**, 1815 (1972).
[6] I. Gochev and V. Tsukernik, Fiz. Tverd. Tela (Leningrad) **15**, 1963 (1973) [Sov. Phys. Solid State **15**, 1315 (1974)].
[7] L. M. Noskova, *ibid.* **18**, 1669 (1976) [**18**, 969 (1976)].
[8] V. M. Loktev and V. S. Ostrovskiĭ, Ukr. Fiz. Zh. **23**, 1708 (1978).
[9] V. S. Ostrovskiĭ, Phys. Stat. Sol. **74b**, K157 (1976).
[10] V. S. Ostrovskiĭ, Preprint No. 6. Phys. Inst. Ukr. Acad. Sci., 1978.
[11] V. V. Druzhinin, S. P. Zapasskiĭ, and V. M. Povyshev, Fiz. Tverd. Tela (Leningrad) **17**, 23 (1975) [Sov. Phys. Solid State **17**, 12 (1975)].
[12] Yu. B. Gaĭdideĭ and V. M. Loktev, Fiz. Nizk. Temp. **3**, 507 (1977) [Sov. J. Low Temp. Phys. **3**, 246 (1977)].
[13] R. O. Zaĭtsev, Zh. Eskp. Teor. Fiz. **68**, 207 (1975) [Sov. Phys. JETP **41**, 100 (1975)].
[14] V. V. Val'kov and S. G. Ovchinnikov, Teor. Mat. Fiz. **50**, 466 (1982).
[15] V. V. Val'kov and S. G. Ovchinnikov, Zh. Eksp. Teor. Fiz. **85**, 1666 (1983) [Sov. Phys. JETP **58**, 970 (1983)].
[16] V. G. Vaks, A. I. Larkin, and S. A. Pikin, *ibid.* **53**, 281 (1967) [**26**, 188 (1968)].
[17] V. G. Vaks, A. I. Larkin, and S. A. Pikin, *ibid.* **53**, 1089 (1967) [**26**, 647 (1968)].
[18] Yu. A. Izyumov, F. A. Kassan-Ogly, and Yu. N. Skryabin, Field Methods in Ferromagnetism Theory [in Russian], Nauka, 1974.
[19] V. V. Val'kov and T. A. Val'kova, Preprint No. 247F, Phys. Inst., Siberian Div. USSR Acad. Sci., 1983.
[20] I. S. Sandalov and A. N. Podmarkov, Preprint No. 156F, Phys. Inst., Siberian Div. USSR Acad. Sci., 1980.
[21] A. V. Vedyaev and V. A. Ivanov, Teor. Mat. Fiz. **47**, 425 (1981).
[22] A. V. Vedyaev and V. A. Ivanov, *ibid.* **50**, 415 (1982).
[23] L. D. Landau and E. M. Lifshitz, Quantum Mechanics, Nonrelativistic Theory, Pergamon 1978 [p. 237 of Russ. Orig.].
[24] A. A. Abrikosov, L. P. Gor'kov, and I. E. Dzyaloshinskii, Quantum Field Theoretical Methods in Statistical Methods, Pergamon, 1965 [p. 137 of Russ. Orig.].
[25] S. V. Vonsovskii, Magnetism, Halsted, 1975.

[26]A. G. Gurevich, Magnetic Resonance in Ferrites and in Antiferromagnets [in Russian], Nauka, 1973.
[27]A. Abragam and B. Bleaney, Electron Paramagnetic Resonance of Transition Ions, Oxford, 1970 [Russ. transl., Vol. 2, p. 349].
[28]K. P. Belov, Rare Earth Magnets and Their Use [in Russian], Nauka, 1980, p. 127.
[29]F. P. Onufrieva, Zh. Eksp. Teor. Fiz. **80**, 2372 (1981) [Sov. Phys. **53**, 1241 (1981)].
[30]R. O. Zaĭtsev, Atomic Energy Inst. Preprint IAE No. 2378, 1974.
[31]F. P. Onufrieva, Zh. Eksp. Teor. Fiz. **86**, 1691 (1984 [Sov. Phys. JETP **59**, 986 (1984)].

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