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Ground-State Energy of Biquadratic Spin Systems ($S = 3/2$) in the $(1/z)^1$-Approximation

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Corrections to the molecular-field ground-state energies of the Heisenberg model with isotropic biquadratic interactions (spin $S = 3/2$) are calculated in the $(1/z)^1$-approximation using the diagrammatic technique based on the Wick reduction theorem (z is the number of spins interacting with any given spin). The present results for the antiferri- and antiferromagnetic phases complete the previously obtained data for the antiquadrupolar, ferri- and ferromagnetic phases. From among the boundaries between different ground states only that between the antiferri- and antiferromagnetic phases is shifted with respect to its molecular-field value.

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1. Introduction

Inclusion of biquadratic terms into the ordinary Heisenberg Hamiltonian made it possible to describe thermomagnetic properties of many real magnetic substances. The biquadratic interactions are especially important in transition-metal and rare-earth compounds with unquenched orbital momentum [1, 2]. In some cases, such as the Jahn–Teller effect [3] or the quadrupolar ordering of molecules in solid hydrogen, they prevail over the bilinear ones or may be even the only interactions (no bilinear coupling) [4, 5].

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The physical origins of the biquadratic interactions are diverse and range from the superexchange [6] and magnetoelastic effect [7, 8] to the virtual phonon exchange between ions [2] and the indirect exchange via the conduction electrons [9]. Also, the description of the lowest multiplet levels of magnetic ions in terms of the spin Hamiltonian may introduce biquadratic components [1]. Experiments on europium, gadolinium and uranium compounds revealed that the fourth-order (biquadratic, three- and four-spin) exchange interactions cause anomalous (non-Bloch) low-temperature variation of the spontaneous magnetization in ferromagnets and of the sublattice magnetization in antiferromagnets. In 3-dimensional magnetic systems the deviation of both quantities from their $T = 0$ value is given by the $T^{9/2}$ or T^2 law for integer or half-integer spin S, whereas in 2-dimensional systems the exponents are 2 or 3/2, respectively [10]. For a more extensive review and for the recent papers on the Heisenberg-biquadratic systems the reader is referred to our previous work [11] and the references cited therein.

The exact ground state of a spin system with isotropic bilinear (J) and biquadratic (K) nearest-neighbours interactions can be determined only in a few special cases, such as the ferromagnetic pure Heisenberg ($J > 0,\ K = 0$) model or the linear antiferromagnetic Heisenberg ($J < 0,\ K = 0$) chain of spins $S = 1/2$. Also, the spin-1 Schrödinger exchange model ($J = K$) with the Zeeman and single-ion anisotropy contributions is known to have the exact ferromagnetic or quadrupolar ground state [12]. In a general case it is possible to establish upper and lower bounds for the ground-state energy [13]. From the condition of coincidence of these bounds Nauciel-Bloch et al. [14] and Barma [15] obtained the sufficient conditions for the ferromagnetic state to be the exact ground state, namely

$$J > 0, \qquad -\frac{J}{2S^2} < K < \frac{J}{S} \qquad (S \geq 1).$$

For spin one, a more thorough inspection [16] yields the broader region

$$J > 0, \qquad K < J \qquad (S = 1).$$

When the exact ground state is unknown, an approximate one, given by the molecular-field approximation, is assumed as a starting point for calculations of elementary excitations. The order parameters at $T = 0$ and the ground-state energy are afterwards corrected by an appropriate perturbation procedure. The presence of such corrections is called the zero-point effect. The theory of spin waves in the Heisenberg antiferromagnet predicts the zero-point effects to be the more pronounced, the lower dimensionality, and the lower spin [17]. The zero-point spin reduction has been observed experimentally in quasi 2-dimensional antiferromagnets [18]. In the Heisenberg model with biquadratic interactions the zero-point effects come from magnons and quadrons (by quadrons we mean magnetic excitations with $\Delta S^z = 2$).

The zero-point effects in the Heisenberg-biquadratic spin systems have already been investigated by the diagrammatic technique based on the Wick reduction theorem, in the $(1/z)^1$-approximation(z is the number of spins interacting with any given spin). Skrobiś and Westwański [19] determined, for $S = 1$, the zero-point deviation of the dipolar and quadrupolar order parameters as well as the corrections to the ground-state energy in the antiferromagnetic and ferriquadrupolar phases. Błaszczak and Westwański [20] considered the spin $S = 3/2$. For $J_0 \geq \frac{1}{2}K_0$, $K_0 < 0$ they calculated the zero-point effects in the antiquadrupolar and ferrimagnetic phases. The present paper complements the latter by taking into account the range of $J_0 < \frac{1}{2}K_0$, $K_0 < 0$, where the antiferrimagnetic or antiferromagnetic phases are the molecular-field ground states (J_0 and K_0 are Fourier transforms of the interaction parameters J_{fg} and K_{fg} in Eq. (1) for the wave vector $\boldsymbol{k} = \mathbf{0}$). For the sake of completeness we shall collect together the new results and the previous ones.

The layout of the paper is as follows. In Sec. 2 the two-sublattice Hamiltonian, expressed by the Racah operators, is split into the molecular-field part and the interaction. The matrix structure of the interaction and of the Green function is displayed. In Sec. 3 the molecular-field ground-state diagram is presented. Order parameters, energies, and phase boundaries are given for the five ground states (ferromagnetic, ferrimagnetic, antiquadrupolar, antiferrimagnetic, and antiferromagnetic), which occur for $K_0 < 0$. Negative branches of magnons and quadrons, which contribute to the zero-point shift of the ground-state energy, are indicated. In Sec. 4 corrections to the ground-state energy in the $(1/z)^1$-approximation are calculated and depicted. Discussion of the results and critical remarks, pointing out to some deficiencies of the applied approximation, conclude the paper.

2. The basic formalism

2.1. Hamiltonian and order parameters

We shall examine a two-sublattice regular array of $2N$ magnetic ions, interacting by one of the mechanisms mentioned in Sec. 1, with the effective spin-3/2 Hamiltonian

$$\begin{aligned} H = &-\sum_f \left[hS_f^z + D(S_f^z)^2\right] - \sum_g \left[hS_g^z + D(S_g^z)^2\right] \\ &-\sum_{f,g} \left[J_{fg}(\boldsymbol{S}_f \cdot \boldsymbol{S}_g) + K_{fg}(\boldsymbol{S}_f \cdot \boldsymbol{S}_g)^2\right], \end{aligned} \tag{1}$$

where h and D are the external magnetic field and the single-ion anisotropy, whereas J_{fg} and K_{fg} are the isotropic Heisenberg and biquadratic couplings. The site indices f and g run over the sublattice A and B, respectively (each spin from sublattice A interacts only with its nearest neighbours belonging to B, and vice versa). A similar (Ising-like) spin-3/2 Hamiltonian has been found to describe qualitatively two successive phase transitions in DyVO$_4$ [21].

It is convenient, for diagrammatic calculations [22, 23], to express the Hamiltonian (1) in terms of the non-normalized Racah tensor operators I_l^m ($l = 1, 2$; $m = -l, \ldots, l$). For $S = 3/2$, they read

$$I_1^0 = S^z, \quad I_1^{\pm 1} = S^{\pm},$$

$$I_2^0 = (S^z)^2 - 5/4, \quad I_2^{\pm 1} = S^z S^{\pm} + S^{\pm} S^z, \quad I_2^{\pm 2} = (S^{\pm})^2. \tag{2}$$

Then, we apply to the operators (2) the transformation

$$\widehat{A} = A - \langle A \rangle, \tag{3}$$

where the symbol $\langle \ldots \rangle$ denotes the thermal average. Since the Hamiltonian (1) is invariant with respect to the rotations around the z-axis, only four averages in (3) do not vanish, namely

$$m_1^j = \langle I_{1j}^0 \rangle, \quad m_2^j = \langle I_{2j}^0 \rangle, \quad (j = A, B). \tag{4}$$

After the transformations (3) the Hamiltonian (1) takes the form

$$H = NC + H_0 - V, \tag{5a}$$

where

$$C = -\frac{5}{2}D - \frac{25}{8}\varphi_0 + \delta_0 m_1^A m_1^B + \varphi_0 m_2^A m_2^B, \tag{5b}$$

$$H_0 = \sum_f H_f + \sum_g H_g, \tag{5c}$$

$$H_f = -h^A I_{1f}^0 - D^A I_{2f}^0, \quad H_g = -h^B I_{1g}^0 - D^B I_{2g}^0,$$

$$h^{A,B} = h + \delta_0 m_1^{B,A} \equiv \beta^{-1} y_1^{A,B}, \quad (\beta = 1/kT), \tag{5d}$$

$$D^{A,B} = D + \varphi_0 m_2^{B,A} \equiv \beta^{-1} y_2^{A,B}, \tag{5e}$$

$$V = \sum_{f,g} \left[\frac{1}{2}\delta_{fg}(\widehat{I}_{1f}^0 \widehat{I}_{1g}^0 + I_{1f}^{\bar{1}} I_{1g}^1) + \frac{1}{6}\varphi_{fg}(I_{2f}^{\bar{1}} I_{2g}^1 + I_{2f}^{\bar{2}} I_{2g}^2) + \frac{1}{2}\varphi_{fg}\widehat{I}_{2f}^0 \widehat{I}_{2g}^0 \right.$$
$$\left. + \text{ the same terms with } \quad f \leftrightarrow g \right]. \tag{5f}$$

Here δ_0 and φ_0 are the Fourier transforms, $\delta_{\boldsymbol{k}}$ and $\varphi_{\boldsymbol{k}}$, of $\delta_{fg} = J_{fg} - \frac{1}{2}K_{fg}$ and $\varphi_{fg} = \frac{3}{2}K_{fg}$, respectively, taken for the wave vector $\boldsymbol{k} = \boldsymbol{0}$. We also define

$$x = -\frac{\delta_0}{\varphi_0}, \qquad \gamma_{\boldsymbol{k}} = \frac{\delta_{\boldsymbol{k}}}{\delta_0} = \frac{\varphi_{\boldsymbol{k}}}{\varphi_0}. \tag{6}$$

2.2. Interactions and Green functions

The interaction term, V, given by (5f), may be rewritten as

$$V = \sum_{f,g} \left[\sum_{l=1}^{2} \sum_{m=0}^{l} \left(V_{lf,lg}^{m\overline{m}} \widehat{I_{lf}^{\overline{m}}} \widehat{I}_{lg}^{m} + V_{lg,lf}^{m\overline{m}} \widehat{I_{lg}^{\overline{m}}} \widehat{I}_{lf}^{m} \right) \right],$$

where the coefficients $V^{m\overline{m}}_{li,l'i'}$ ($m = 0,1,2$; $l,l' = 1,2$; $i,i' = f,g$) form a 10×10 symmetric matrix, which is the simple sum of 4×4, 4×4, and 2×2 matrices, corresponding to $m = 0$, 1, and 2, respectively. The non-vanishing elements of its Fourier transform

$$\boldsymbol{V}(\boldsymbol{k}) = \boldsymbol{V}^{00}(\boldsymbol{k}) \oplus \boldsymbol{V}^{1\overline{1}}(\boldsymbol{k}) \oplus \boldsymbol{V}^{2\overline{2}}(\boldsymbol{k})$$

are as follows:

$$V^{00}_{1A,1B}(\boldsymbol{k}) = V^{00}_{1B,1A}(\boldsymbol{k}) = V^{1\overline{1}}_{1A,1B}(\boldsymbol{k}) = V^{1\overline{1}}_{1B,1A}(\boldsymbol{k}) = \frac{1}{2}\delta_0\gamma_{\boldsymbol{k}},$$

$$V^{00}_{2A,2B}(\boldsymbol{k}) = V^{00}_{2B,2A}(\boldsymbol{k}) = \frac{1}{2}\varphi_0\gamma_{\boldsymbol{k}},$$

$$V^{1\overline{1}}_{2A,2B}(\boldsymbol{k}) = V^{1\overline{1}}_{2B,2A}(\boldsymbol{k}) = V^{2\overline{2}}_{2A,2B}(\boldsymbol{k}) = V^{2\overline{2}}_{2B,2A}(\boldsymbol{k}) = \frac{1}{6}\varphi_0\gamma_{\boldsymbol{k}}.$$

We introduce the matrix Green function, $\boldsymbol{G}(\boldsymbol{k}, \mathrm{i}\omega_n)$, depending upon two external vertices [23]

$$\boldsymbol{G}(\boldsymbol{k}, \mathrm{i}\omega_n) = \boldsymbol{G}^{00}(\boldsymbol{k}, \mathrm{i}\omega_n) \oplus \boldsymbol{G}^{\overline{1}1}(\boldsymbol{k}, \mathrm{i}\omega_n) \oplus \boldsymbol{G}^{\overline{2}2}(\boldsymbol{k}, \mathrm{i}\omega_n) \tag{7}$$

($\mathrm{i}\omega_n = \mathrm{i}2\pi n/\beta$ is the imaginary Bose frequency). It satisfies the diagrammatic Larkin equation [24]

$$\boldsymbol{G} = \boldsymbol{\Sigma} + \boldsymbol{\Sigma}\boldsymbol{V}\boldsymbol{G},$$

where $\boldsymbol{\Sigma}$, the full irreducible part of the Green function, has the same structure (7) as $\boldsymbol{G}$. The expressions for $\boldsymbol{\Sigma}^{\overline{m}m}(\boldsymbol{k}, \mathrm{i}\omega_n)$ ($m = 0$, 1, 2), in the zeroth-order approximation with respect to $(1/z)$, are given by Eqs. (16), (17), and (18) in Ref. [25].

3. Molecular-field approximation

3.1. Ground state

Omitting the interaction term, V, in Eq. (5a), we derive the following expression for the free energy per ion in the molecular-field approximation (MFA):

$$\overline{F}_0 = C - \beta^{-1}\ln(Z_A Z_B), \tag{8}$$

where

$$Z_j = 2\left[\exp(y_2^j)\cosh\left(\frac{3}{2}y_1^j\right) + \exp(-y_2^j)\cosh\left(\frac{1}{2}y_1^j\right)\right], \quad \beta = 1/kT,$$

and y_l^j are defined by Eqs. (5d,e) ($j = A, B$).

The minimum condition for $\overline{F}_0$ yields the self-consistent equations for the order parameters (4):

$$m_l^j = b_l^j(y_1^i, y_2^j) \equiv Z_j^{-1}\frac{\partial Z_j}{\partial y_l^j}, \quad l = 1,2; \quad j = A,\ B. \tag{9}$$

The ground-state energy per ion in the MFA is

$$\overline{E}_0 = C + \frac{1}{N}\langle H_0 \rangle_0 .$$

Inserting (5b,c) and putting $h = D = 0$ (this assumption will be kept throughout the remainder of the present paper) we get

$$E_0 \equiv \frac{\overline{E}_0}{|K_0|} = \frac{3}{2}\left(\frac{25}{8} - x b_1^A b_1^B + b_2^A b_2^B\right). \tag{10}$$

Here the order parameters b_l^j $(l = 1,2;\ j = A, B)$ assume their zero-temperature values. Below we write down the values of the order parameters, b_l^j, $(l = 1,2;$ $j = A, B)$, the expressions for the ground-state energy, E_0, and the region of x, where a given phase is the ground state.

(i) Ferromagnetic (FE) phase:

$$b_1^A = b_1^B = \frac{3}{2}, \quad b_2^A = b_2^B = 1; \quad E_0 = \frac{9}{8}\left(\frac{11}{2} - 3x\right); \quad x \geq \frac{4}{3}. \tag{11}$$

(ii) Ferrimagnetic (FI) phase:

$$b_1^A = \frac{3}{2}, \quad b_1^B = \frac{1}{2}; \quad b_2^A = -b_2^B = 1;$$

$$E_0 = \frac{3}{8}\left(\frac{17}{2} - 3x\right); \quad 0 < x \leq \frac{4}{3}. \tag{12}$$

(iii) Antiquadrupolar (AQ) phase:

$$b_1^A = b_1^B = 0, \quad b_2^A = -b_2^B = 1; \quad E_0 = \frac{51}{16}; \quad x = 0. \tag{13}$$

(iv) Antiferrimagnetic (AFI) phase:

$$b_1^A = \frac{3}{2}, \quad b_1^B = -\frac{1}{2}, \quad b_2^A = -b_2^B = 1;$$

$$E_0 = \frac{3}{8}\left(\frac{17}{2} + 3x\right); \quad -\frac{4}{3} \leq x < 0. \tag{14}$$

(v) Antiferromagnetic (AFE) phase:

$$b_1^A = -b_1^B = \frac{3}{2}, \quad b_2^A = b_2^B = 1; \quad E_0 = \frac{9}{8}\left(\frac{11}{2} + 3x\right); \quad x \leq -\frac{4}{3}. \tag{15}$$

For $h = D = 0$ the molecular-field equations (8–9) are equivalent to those obtained by Sivardière and Blume [21] for the $S = 3/2$ Ising-like spin system with dipolar and quadrupolar interactions. Our interaction parameters δ_0 and φ_0 are related to J and K in [21] as follows:

$$\frac{\delta_0}{\varphi_0} = \frac{J}{K} \quad (= -x).$$

The ground-state diagram, determined by Eqs. (11–15), corresponds to that in Fig. 7 in Ref. [21], with the replacements FE → AFE and FI → AFI for $x < 0$.

3.2. Negative branches of the spectrum at $T = 0$

The spectrum of elementary excitations is determined by the poles of the analytical continuation $\mathrm{i}\omega_n \longrightarrow -\omega - \mathrm{i}\varepsilon$ ($\varepsilon \longrightarrow +0$) of the Green functions $\boldsymbol{G}^{\overline{m}m}(\boldsymbol{k}, \mathrm{i}\omega_n)$ (m = 1, 2). The spectrum in the $(1/z)^0$-approximation has been calculated in Refs. [25] and [11] for the FE, FI, AQ, and AFI, AFE phases, respectively. Here we shall only quote the formulae for the negative energy branches, since only they contribute to the free energy corrections in the $(1/z)^1$-approximation at $T = 0$.

In the ferromagnetic phase magnon and quadron spectra are positive. On the other hand, in the antiferromagnetic phase there exists one negative magnon branch

$$\Omega_{2\boldsymbol{k}} = 3\left(\frac{3}{4}x + 1\right)(1 - \gamma_{\boldsymbol{k}}^2)^{1/2}. \tag{16}$$

as well as one negative quadron branch

$$\Omega_{4\boldsymbol{k}} = -3\left[\left(\frac{3}{2}x + 1\right)^2 - \gamma_{\boldsymbol{k}}^2\right]^{1/2} \tag{17}$$

(we follow the notation of Eqs. (27) and (28a) in Ref. [11]).

Both in the ferrimagnetic and antiferrimagnetic phases there is one negative magnon branch. Their energies, ω, are obtained from the equation

$$(-\omega)^3 + b(-\omega)^2 + c(-\omega) + d = 0, \tag{18a}$$

where (FI)

$$b = \frac{21}{4}x,$$

$$c = 9\left[\frac{3}{16}x^2\left(5 - \gamma_{\boldsymbol{k}}^2\right) + \frac{1}{2}x(1 - 3\gamma_{\boldsymbol{k}}^2) - (1 - \gamma_{\boldsymbol{k}}^2)\right],$$

$$d = \frac{81}{4}x(1 - \gamma_{\boldsymbol{k}}^2)\left(\frac{3}{16}x^2 + \frac{1}{2}x - 1\right), \tag{18b}$$

or (AFI)

$$b = 6\left(\frac{5}{8}x + 1\right),$$

$$c = 9\left[\frac{3}{16}x^2(1 + \gamma_{\boldsymbol{k}}^2) + \frac{1}{2}x(4 - 3\gamma_{\boldsymbol{k}}^2) + (1 - \gamma_{\boldsymbol{k}}^2)\right],$$

$$d = -\frac{81}{4}x(1 - \gamma_{\boldsymbol{k}}^2)\left(\frac{3}{16}x^2 - \frac{1}{2}x - 1\right). \tag{18c}$$

As concerns quadrons, there is no negative branch in the ferrimagnetic phase, while in the antiferrimagnetic phase the one negative branch is given by (once again we use the notation of Ref. [11])

$$\omega_{5\boldsymbol{k}} = \frac{3}{2}\left\{x - 2[(x-1)^2 - \gamma_{\boldsymbol{k}}^2]^{1/2}\right\}. \tag{19}$$

In the antiquadrupolar phase the magnon spectrum coincides with that of quadrons. The two negative branches are given by (Eq. (21a) in Ref. [25])

$$\varepsilon_{1,2\boldsymbol{k}} = -3(1 \mp |\gamma_{\boldsymbol{k}}|)^{1/2}. \tag{20}$$

The presence of negative elementary excitations in most of the phases considered (except of FE) indicates that the MFA ground state is not exact and needs to be improved by the zero-point corrections.

4. Ground state of the system

4.1. Free energy in the $(1/z)^1$-approximation

The Green functions (7) in the $(1/z)^0$-approximation being known, we may calculate the first-order corrections to the free energy per ion (in units of $|K_0|$):

$$F = F_0 + \Delta F_0 + \Delta F_1 + \Delta F_2, \tag{21}$$

where $F_0 = \overline{F}_0/|K_0|$ is the MFA value of the free energy, Eq. (8), and

$$\Delta F_m = \frac{1}{N\beta}\sum_{\boldsymbol{k},n} \ln Z_m(\boldsymbol{k}, \mathrm{i}\omega_n), \tag{22}$$

$$Z_m(\boldsymbol{k}, \mathrm{i}\omega_n) = \det\left[1 - \overset{0}{\boldsymbol{\Sigma}}{}^{\overline{m}m}(\boldsymbol{k}, \mathrm{i}\omega_n)\, \boldsymbol{V}^{m\overline{m}}(\boldsymbol{k})\right], \quad m = 1,\ 2. \tag{23}$$

The correction ΔF_0 in (21), coming from the diagonal part of the Green function (7), vanishes at $T = 0$ and therefore does not contribute to the ground-state energy. The two further corrections, ΔF_1 and ΔF_2, correspond to magnons and quadrons, respectively. Then (23) can be expressed by the collective excitation spectrum $\varepsilon_{\boldsymbol{k}}^{(r)}$ and by the corresponding distances between the molecular-field energy levels $H^{(r)}$ as follows:

$$Z_m(\boldsymbol{k}, \mathrm{i}\omega_n) = \prod_r \frac{\mathrm{i}\omega_n + \varepsilon_{\boldsymbol{k}}^{(r)}(m)}{\mathrm{i}\omega_n + H^{(r)}(m)}, \quad m = 1,\ 2. \tag{24}$$

Here $m = 1$ for magnons and $m = 2$ for quadrons, whereas the index r numerates different branches of the excitation spectrum in a given phase (e.g. $r = 1,\ 2,\ 3$ for magnons in the FI phase). The energy distances $H^{(r)}$ are equal to the corresponding spectra $\varepsilon_{\boldsymbol{k}}^{(r)}$ for the interaction $V = 0$ in Eq. (5) and may be obtained by formally putting $\gamma_{\boldsymbol{k}} = 0$ in Eqs. (16–20). Their explicit form is given by Eqs. (13) and (14) in Ref. [11].

Inserting (24) into (22) and performing the summation over n we obtain

$$\Delta F_m = \frac{1}{N\beta}\sum_{\boldsymbol{k}} \ln \prod_r \frac{1 - \exp[-\beta\varepsilon_{\boldsymbol{k}}^{(r)}(m)]}{1 - \exp[-\beta H^{(r)}(m)]}, \quad m = 1,\ 2. \tag{25}$$

4.2. Corrections to the ground-state energy in the $(1/z)^1$-approximation

The ground-state energy (in units of $|K_0|$) with accuracy up to $(1/z)^1$ reads

$$E = E_0 + \Delta E_1 + \Delta E_2, \tag{26}$$

where E_0 is the molecular-field value (10). The first-order corrections, ΔE_1 and ΔE_2, coming from magnons and quadrons, respectively, are obtained by taking the limit $T \rightarrow 0$ in (25)

$$\Delta E_m = \frac{1}{N}\sum_{\boldsymbol{k}}\sum_{r}\left[H^{(r)}(m) - \varepsilon_{\boldsymbol{k}}^{(r)}(m)\right], \quad m = 1,\ 2. \tag{27}$$

Now r runs over the negative branches of the spectrum only. We present underneath the corrections for the five phases in detail.

(i) Ferromagnetic (FE) phase:
No negative magnon or quadron branch exists for the FE phase, hence the correction to the MFA value of the ground-state energy (11) vanishes.

(ii) Ferrimagnetic (FI) phase:
The magnon correction ΔE_1 is calculated by (27), with $H^{(r)}(1) = 3(\frac{3}{4}x - 1)$ and $\varepsilon_{\boldsymbol{k}}^{(r)}(1) = \varepsilon_{4\boldsymbol{k}}$ (the notation follows that of Ref. [25]); $\varepsilon_{4\boldsymbol{k}}$ is the negative magnon branch determined by Eq. (18a) with the coefficients (18b). The correction from quadrons vanishes.

(iii) Antiquadrupolar (AQ) phase:
Inserting (20) into (27) we get

$$\Delta E_1 = \Delta E_2 = -\frac{3}{N}\sum_{\boldsymbol{k}}\left[2 - (1 - |\gamma_{\boldsymbol{k}}|)^{1/2} - (1 + |\gamma_{\boldsymbol{k}}|)^{1/2}\right]. \tag{28}$$

(iv) Antiferrimagnetic (AFI) phase:
The magnon correction ΔE_1 is obtained from (27) by putting $H^{(r)}(1) = \frac{9}{4}x$ and $\varepsilon_{\boldsymbol{k}}^{(r)}(1) = \omega_{2\boldsymbol{k}}$ (the notation of Ref. [11]); $\omega_{2\boldsymbol{k}}$ is the negative magnon branch determined by Eq. (18a) with the coefficients (18c). To get the quadron correction we put $H^{(r)}(2) = 3(\frac{3}{2}x - 1)$ and $\varepsilon_{\boldsymbol{k}}^{(r)}(2) = \omega_{5\boldsymbol{k}}$, see Eq. (19), wherefrom

$$\Delta E_2 = 3(x-1) + \frac{3}{N}\sum_{\boldsymbol{k}}[(x-1)^2 - \gamma_{\boldsymbol{k}}^2]^{1/2}. \tag{29}$$

(v) Antiferromagnetic (AFE) phase:
Similar calculations, applied to negative magnon and quadron branches (Eqs. (16) and (17), respectively), yield the following expressions for the corrections to the ground-state energy:

$$\Delta E_1 = 3\left(\frac{3}{4}x + 1\right)\left[1 - \frac{1}{N}\sum_{\boldsymbol{k}}(1 - \gamma_{\boldsymbol{k}}^2)^{1/2}\right]. \tag{30}$$

$$\Delta E_2 = 3\left(\frac{3}{2}x+1\right) + \frac{3}{N}\sum_{\boldsymbol{k}}\left[\left(\frac{3}{2}x+1\right)^2 - \gamma_{\boldsymbol{k}}^2\right]^{1/2}. \tag{31}$$

The numerical calculations have been performed for the simple cubic (s.c.) and body-centred cubic (b.c.c.) lattices. Figure 1 shows, in the case of the s.c. lattice, the magnon and quadron corrections separately for the phases corresponding to different values of x (a similar behaviour holds for the b.c.c. lattice).

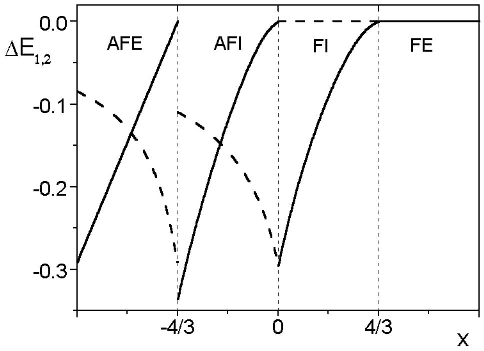

Fig. 1. Corrections (27) to the ground-state energy for the s.c. lattice. The solid and dashed lines correspond to magnon (ΔE_1) and quadron (ΔE_2) contributions, respectively. The interaction parameter x is given by Eq. (6) and the energy is in reduced units (see Eqs. (10) and (26)).

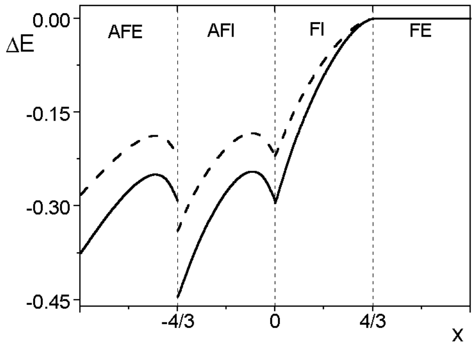

Fig. 2. The total correction to the ground-state energy, $\Delta E = \Delta E_1 + \Delta E_2$, given by Eqs. (27–31), calculated for the s.c. (solid line) and b.c.c. (dashed line) lattices. The interaction parameter x is given by Eq. (6) and the energy is in reduced units (see Eqs. (10) and (26)).

The total correction, $\Delta E = \Delta E_1 + \Delta E_2$, calculated for the two lattices, is presented in Fig. 2. Figure 3 compares, for the s.c. lattice, the full ground-state energy E in the $(1/z)^1$-approximation, Eq. (26), with its MFA value E_0, Eq. (10).

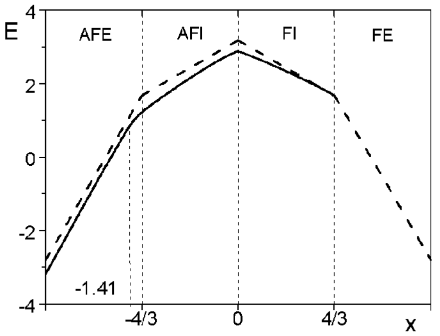

Fig. 3. The ground-state energy E versus the interaction parameter x for the s.c. lattice. Dashed line: molecular-field theory, Eqs. (10–15); solid line: $(1/z)^1$-approximation, Eqs. (26–31). The point $x' = -1.41$ is the new border between the AFI and AFE ground states (see Sec. 4.3 for details).

Figure 3 in this paper is an extension of Fig. 1 in Ref. [20] onto the region of negative x.

4.3. Remarks

As should be expected, the first-order corrections to the ground-state energy are small (a few percent of the MFA value E_0) and negative (only in the FE phase they vanish). Thus the corrected ground state is of lower energy than the MFA one. Moreover, the departure from the MFA is greater for the s.c. lattice ($z = 6$) than for the b.c.c. lattice ($z = 8$). These results legitimize the use of the high-density perturbation scheme, with $(1/z)$ being the perturbation parameter.

In Fig. 2 we see that the total correction, ΔE, is continuous at $x = 4/3$ and at $x = 0$ (also the antiquadrupolar correction, Eq. (28), agrees numerically with the corrections for the FI and AFI ground states at $x = 0$). Therefore, the MFA border between the FE and FI phases ($x = 4/3$) as well as that between the FI, AQ, and AFI phases ($x = 0$) persists in the $(1/z)^1$-approximation. This is not the case for $x = x_0 \equiv -4/3 = -1.33$ (MFA border between AFI and AFE). The condition of equal energies of the AFI and AFE ground states in the $(1/z)^1$-approximation leads to the new borderline value x' of the interaction parameter x: x' assumes the value of -1.41 and -1.39 for the s.c. and b.c.c. lattices, respectively (notably in both cases $|x'| > |x_0|$). However, for $x \in [x', x_0)$, the antiferrimagnetic magnon spectrum, given by Eqs. (18a) and (18c), becomes complex for small wave vectors $\boldsymbol{k}$; specifically, this occurs for $1 \le \gamma_{\boldsymbol{k}} < 0.94$ (s.c. lattice) or $1 \le \gamma_{\boldsymbol{k}} < 0.96$ (b.c.c. lattice). No such problem occurs for quadrons, determined by Eq. (19). Bearing this in mind, the correction ΔE_1 for $x \in [x', x_0)$ has been calculated by rejecting the above-mentioned values of $\boldsymbol{k}$ out of the region of integration in (27).

The small interval $x \in [x', x_0)$, in which the spectrum of non-interacting spin-waves is complex, is the region of instability of the AFI phase. This circumstance is to be considered as a defect of the applied approximation. Presumably taking into account non-collinear spin states in the phase diagram would alleviate this drawback. This approach is presently under our consideration.

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