Eur. Phys. J. B **5**, 745–750 (1998)

THE EUROPEAN PHYSICAL JOURNAL B

Quadrupolar order in the S = 2 Heisenberg ferromagnet with the single-ion cubic anisotropy*

M. Dudziński and J. Sznajd

Institute for Low Temperature and Structure Research, Polish Academy of Sciences, P.O. Box 937, 50-950 Wroclaw 2, Poland

Received: 19 February 1998 / Accepted: 17 March 1998

Abstract. The ground state properties of $S = 2$ ferromagnets with isotropic Heisenberg exchange (J) and single-ion cubic anisotropy (D) are studied. The perturbation theory for $J/D \ll 1$ is used to find an effective Hamiltonian up to the fourth order for 1, 2 and 3 dimensions. It is shown that in opposition to the MFA prediction there is the quadrupolar long range order at $T = 0$ in the non-magnetic state of the system without a quadrupolar type of interaction. The effect is a consequence of the quantum nature of the model.

PACS. 75.10.Dg Crystal-field theory and spin Hamiltonians – 75.10.Jm Quantized spin models – 75.30.Gw Magnetic anisotropy

1 Introduction

In many real magnetic materials the magnetic ordering produced by an isotropic short-range exchange is affected by the crystal field of lower symmetry. For a lattice of cubic symmetry the lowest-term which describes the crystal field anisotropy can be written in the form

$$H_c = -D \sum_{i,\alpha} (S_i^\alpha)^4, \qquad \alpha = x, y, z \tag{1}$$

where $S \geq 2$ ($S = 2$ is the lowest value for which this term is non-trivial). The effect of a cubic anisotropy on the critical behavior has been studied since 1973 [1] to this day [2]. Much less attention has been devoted to the problem of the low temperature phase and ground state properties of the cubic quantum spin systems.

In a system with higher value of spin $S \geq 1$ in addition to the magnetic (dipolar) long range order one can observe also multipolar order *e.g.* quadrupolar one. It is obvious that a magnetic ground state will always have quadrupolar moment while for a non-magnetic (paramagnetic) state this may be not true. Of course one can expect the existence of quadrupolar ordering even without dipolar one if there is a quadrupolar type of interaction in the system under consideration (there are several mechanisms which give rise to the higher degree - multipolar pair interactions connected not only with electric multipole interactions but also with phonon coupling or multielectron exchange). The question is if there is a possibility to observe the multipolar ordering in paramagnetic phase of the system with only magnetic interaction between ions and single-ion anisotropy. It is clear that such a possibility does not exist for the classical spin systems. However, the quantum character of the spin can lead to qualitatively different physical properties.

In the present paper we show that as a consequence of the quantum character of spins the quadrupolar long range order can be observed in the paramagnetic ground state of the $S = 2$ system without a quadrupolar type of interaction. Taking into account the ground state properties the quantum nature of the systems with higher spin values can be more important than the quantum nature of the systems with so-called the most quantum spin $S = 1/2$ [3].

We consider the cubic model described by the Hamiltonian

$$H = -J \sum_{(ij)} (\boldsymbol{S_i} \boldsymbol{S_j}) - D \sum_i [(S_i^x)^4 + (S_i^y)^4 + (S_i^z)^4], \tag{2}$$

where spin operators for $S = 2$ are denoted by S_i^α. We consider the d-dimensional hypercubic lattice, later on we discuss the case of a linear chain ($d = 1$), square lattice ($d = 2$) and a simple cubic lattice ($d = 3$). The first term of the Hamiltonian (2), with the coupling constant $J > 0$, is the usual isotropic (ferromagnetic) exchange between nearest neighbors. The second term, the crystal field interaction, describes the influence of neighboring ions onto the magnetic ion carrying the magnetic moment. In terms of the effective spin model this is a single-site interaction. We consider the crystal-field anisotropy of cubic symmetry, and the main aim of our work is to investigate what happens with the long-range order (LRO) at $T = 0$ when this anisotropy is present.

* Dedicated to J. Zittartz on the occasion of his 60th birthday

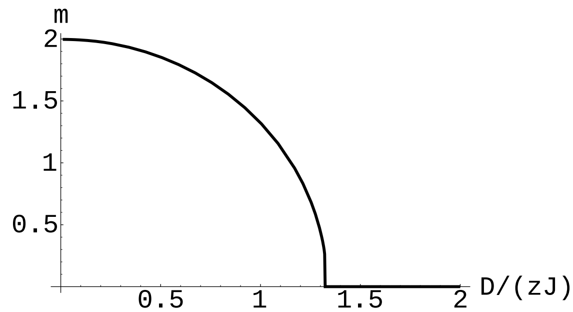


Fig. 1. Spontaneous magnetization $m = \langle S^z \rangle$ *versus* the reduced crystal field parameter $D/(zJ)$ at $T = 0$ (MFA approximation).

2 Preliminary considerations and MFA solution

To start with, we take one spin with the crystal field interaction

$$h = -D[(S^x)^4 + (S^y)^4 + (S^z)^4] \tag{3}$$

and diagonalize the Hamiltonian h. The five states of the $S = 2$ spin are split into a doublet and a triplet. The energies are

$$\begin{aligned} e_1 &= e_2 = -24D, \\ e_3 &= e_4 = e_5 = -18D, \end{aligned} \tag{4}$$

and the eigenstates of h are the following

$$\begin{aligned} |\psi_1\rangle &= \frac{1}{\sqrt{2}}(|2\rangle + |-2\rangle), \\ |\psi_2\rangle &= |0\rangle, \\ |\psi_3\rangle &= |1\rangle, \\ |\psi_4\rangle &= \frac{1}{\sqrt{2}}(|2\rangle - |-2\rangle), \\ |\psi_5\rangle &= -\,|-1\rangle, \end{aligned} \tag{5}$$

where by $|2\rangle, |1\rangle, \ldots, |-2\rangle$ we denoted eigenstates of S^z. The doublet $\{|\psi_1\rangle, |\psi_2\rangle\}$ has a property which is very important for our considerations. Within this doublet, every matrix element of a spin operator S^α ($\alpha = x, y, z$) vanishes:

$$\langle\psi_i \mid S^\alpha \mid \psi_j\rangle = 0, \tag{6}$$

for each $i = 1, 2$ and $j = 1, 2$. One can say this doublet is *nonmagnetic*. If the wave function of the whole system contained only products of $|\psi_1\rangle$ and $|\psi_2\rangle$ the system would never have a nonzero magnetization. In this paper we consider the case of $D \geq 0$ in (2), it means that the nonmagnetic states $|\psi_1\rangle$ and $|\psi_2\rangle$ are favored. For $D = 0$ we have the classical ground state and saturated magnetization. One may expect that the magnetization should decrease or even disappear when D increases.

The model (2) was investigated with the use of the mean-field approximation (MFA) by many authors [4–7]. These authors considered also phase transitions at finite temperature, whereas we concentrate on the LRO in the ground state. It was found by the MFA that the crystal field anisotropy in the model (2) defines easy axes of magnetic ordering; for $D > 0$ there may occur spontaneous ordering along the direction [100] or other equivalent directions. Within the MFA one obtains that the spontaneous magnetization disappears continuously for $D/(zJ) = 4/3$, where z denotes the number of nearest neighbors. The dependence of the spontaneous magnetization on the parameter D is shown in the Figure 1. The MFA always overestimates the tendency towards ordering because it neglects the disordering effect of fluctuations. Therefore, we believe that, indeed, the magnetic order vanishes at some D_c depending on the lattice.

The authors did not discuss the nature of the phase with no spontaneous magnetization. However, it is easy to see that in the mean-field approximation this phase is found to be disordered. Assuming there is no magnetic order we get the MFA Hamiltonian in the form

$$H_{MFA} = -D[(S^x)^4 + (S^y)^4 + (S^z)^4],$$

the exchange term of (2) is completely neglected in this approximation and the spins do not interact with one another. Thus, the MFA leaves a huge degeneracy in the ground state of the phase with no magnetic order. In this approximation each product of the states $|\psi_1\rangle$ and $|\psi_2\rangle$ is a ground state of the system and the total degeneracy is 2^N. In the next section we consider an effective Hamiltonian acting in this subspace of states.

As mentioned in the Introduction if there is no magnetic order in the system, it does not necessarily imply that the system is disordered, there may exist a *multipolar order* described by a correlation function containing the second or higher powers of spin operators. The quadrupolar order could not be obtained by the MFA because of the very nature of this method. It is easy to understand that only magnetization can act as a mean field when the exchange interactions contain only first powers of spin operators, as it is in the Hamiltonian (2).

3 Perturbation theory for $J/D \ll 1$

We apply the perturbation theory for $J/D \ll 1$, treating the crystal field interaction as a main term

$$H_0 = -D\sum_i[(S_i^x)^4 + (S_i^y)^4 + (S_i^z)^4],$$

and the exchange term as a perturbation

$$V = -J\sum_{(ij)}(\boldsymbol{S_i S_j}).$$

Since the Hamiltonian H_0 consists of single-site terms, it can be easily diagonalized. For $D > 0$ we can have either the state $|\psi_1\rangle$ or $|\psi_2\rangle$ on each site in the ground state of H_0, and the degeneracy is 2^N. This is exactly the subspace of states which was obtained by the MFA in the phase with no spontaneous magnetization. We are going to find some effective interaction which could lift this degeneracy.

We follow the approach used in the investigation of the Hubbard model in the limit of strong Coulomb repulsion [8–11]. We base our calculations on the work of Takahashi [11], here we only sketch the method referring the reader to [11] for details.

We denote the subspace of the ground states of H_0 by U_0, the operator of projection onto U_0 is called P_0 and E_0 is the unperturbed energy of the ground states belonging to U_0. The subspace of perturbed states (after the addition of V) is denoted by U and the projector onto it by P. The series expansion for P was derived by Kato [12]. Takahashi used this expansion to construct the operator Γ which maps states from U_0 to U. The operator Γ, such that $\Gamma^\dagger\Gamma = 1$, enables to reduce the eigenvalue problem for H to the subspace U_0. For the Hamiltonian H and the physical quantity A we find the mappings

$$H_{\mathit{eff}} = \Gamma^\dagger H \Gamma, \tag{7}$$

$$a = \Gamma^\dagger A \Gamma. \tag{8}$$

Working with H_{eff} and a in U_0 is equivalent to considering H and A in U. In order to calculate the average of an operator A in the ground state of H one must first perform the above mapping, then find the ground state of H_{eff} and compute the average of a.

The operator P_0 projects onto the subspace of products of the nonmagnetic states $|\psi_1\rangle$ and $|\psi_2\rangle$. From the property (6) of these states it follows that $P_0VP_0 = 0$. In such a case, we obtain from (7) and perturbational expansion of [11] the effective Hamiltonian in the form

$$H_{\mathit{eff}} = H_{\mathit{eff}}^{(2)} + H_{\mathit{eff}}^{(3)} + H_{\mathit{eff}}^{(4)} + \cdots, \tag{9}$$

where

$$\begin{aligned} H_{\mathit{eff}}^{(2)} &= P_0VRVP_0, \\ H_{\mathit{eff}}^{(3)} &= P_0VRVRVP_0, \\ H_{\mathit{eff}}^{(4)} &= P_0VRVRVRVP_0 \\ &\quad -\frac{1}{2}(P_0VR^2VP_0VRVP_0 + h.c.), \end{aligned}$$

and

$$R = \frac{1-P_0}{E_0 - H_0}\cdot$$

In the above expansion we omitted the zeroth-order term $P_0H_0P_0$, which is a constant and only shifts the energy scale. The property (6) makes the first order of the expansion vanish. Since there are two possible states per site in U_0, the effective Hamiltonian H_{eff} will have a form of a spin $S = 1/2$ Hamiltonian. We will write it down in terms of Pauli matrices $\{\sigma^x, \sigma^y, \sigma^z\}$ with the identification

$$|\psi_1\rangle \to |+\rangle,$$
$$|\psi_2\rangle \to |-\rangle,$$

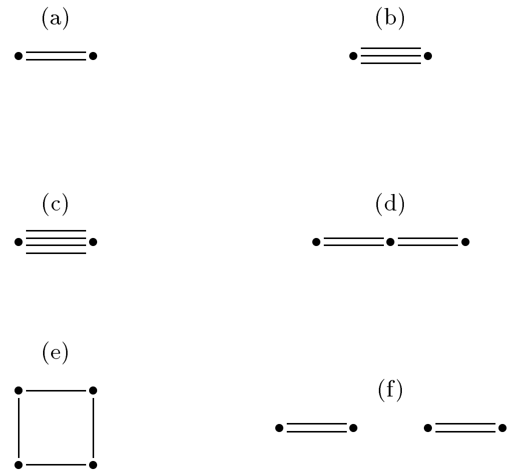


Fig. 2. Diagrams appearing in the perturbational calculation of the effective Hamiltonian: second order diagram (a), third order (b), fourth order (c), (d), (e) and (f).

where $|+\rangle$ and $|-\rangle$ stand for the states of the effective "spin" $S = 1/2$ (quotation marks remind that this two-state object is not a real spin; operators σ^α are, in fact, operators of the quadrupolar moment).

The calculation of H_{eff} in the second order (the lowest nonvanishing order) is quite simple, there is only one diagram, which is shown in Figure 2a. The effective Hamiltonian have the same form in $d = 1$, 2 and 3

$$H_{\mathit{eff}}^{(2)} = -\frac{J^2}{2D}\sum_{(ij)}(\sigma_i^x\sigma_j^x + \sigma_i^z\sigma_j^z), \tag{10}$$

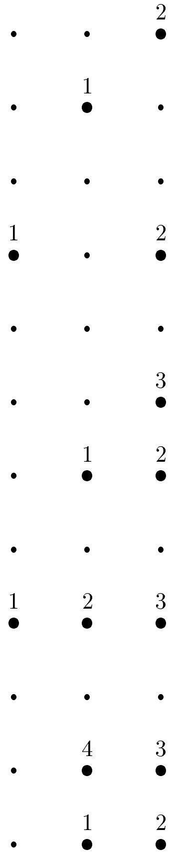
the summation goes over pairs of nearest neighbors. Thus, we have obtained an effective interaction between the nonmagnetic states. This is a hint that the MFA prediction about the disordered state for big D may be not correct. The model described by $H_{\mathit{eff}}^{(2)}$, the XY-type planar model, is known to exhibit the long-range order in $d = 2$ and 3 [13], and to be critical in $d = 1$.

The Hamiltonian $H_{\mathit{eff}}^{(2)}$ has a continuous rotational symmetry in its XZ plane, whereas the original model possesses a discrete cubic symmetry. This difference may only be a result of the approximation; obviously the value of J/D does not change the symmetry. We should find the symmetry breaking terms in higher orders of H_{eff}.

We do not find them in the third order contribution to H_{eff}, which is obtained from the diagram (b) of Figure 2:

$$\begin{aligned} H_{\mathit{eff}}^{(3)} = &\frac{J^3}{12D^2}\sum_{(ij)}(\sigma_i^x\sigma_j^x + \sigma_i^z\sigma_j^z) \\ &-\frac{J^3}{8D^2}\sum_{(ij)}(\sigma_i^y\sigma_j^y). \end{aligned} \tag{11}$$

Although the new coupling between components σ^y appeared, the Hamiltonian still has the rotational symmetry in the plane XZ.



$$I^{(1)} = \sigma_1^x \sigma_2^x + \sigma_1^z \sigma_2^z$$

$$I^{(2)} = \sigma_1^x \sigma_2^x + \sigma_1^z \sigma_2^z$$

$$I^{(3)} = \sigma_1^z \sigma_2^z \sigma_3^z - \sigma_1^x \sigma_2^x \sigma_3^z - \sigma_1^x \sigma_2^z \sigma_3^x - \sigma_1^z \sigma_2^x \sigma_3^x$$

$$I^{(4)} = \sigma_1^z \sigma_2^z \sigma_3^z - \sigma_1^x \sigma_2^x \sigma_3^z - \sigma_1^x \sigma_2^z \sigma_3^x - \sigma_1^z \sigma_2^x \sigma_3^x$$

$$\begin{aligned} I^{(5)} &= \sigma_1^x \sigma_2^x \sigma_3^x \sigma_4^x + \sigma_1^z \sigma_2^z \sigma_3^z \sigma_4^z \\ &+ \tfrac{1}{3}(\sigma_1^x \sigma_2^x \sigma_3^z \sigma_4^z + \sigma_1^x \sigma_2^z \sigma_3^x \sigma_4^z + \sigma_1^x \sigma_2^z \sigma_3^z \sigma_4^x \\ &+ \sigma_1^z \sigma_2^x \sigma_3^x \sigma_4^z + \sigma_1^z \sigma_2^x \sigma_3^z \sigma_4^x + \sigma_1^z \sigma_2^z \sigma_3^x \sigma_4^x) \end{aligned}$$

Fig. 3. Types of interactions obtained in the fourth order of the expansion for the effective Hamiltonian. A piece of a square lattice is shown, interactions $I^{(n)}$ are between "spins" denoted by bold points. Interactions $I^{(1)}$, $I^{(3)}$ and $I^{(5)}$ are present only in $d = 2$ and 3.

The desired terms are found in the fourth order of the expansion for H_{eff}. The diagrams (c), (d) and (e) from Figure 2 need to be considered, the unlinked diagram (f) does not give any contribution. The variety of new interactions appearing in the fourth order is shown in Figure 3. The interactions $I^{(3)}$ and $I^{(4)}$ are crucial for our considerations, because they reduce the symmetry of H_{eff} from the full rotational symmetry in the XZ plane to the symmetry of discrete rotations through the angle $\pm(2\pi)/3$. All other interactions present, including the four-"spin" term $I^{(5)}$ have the continuous symmetry in the XZ plane.

We introduce the notation $I^{(n)}_{chain}$, $I^{(n)}_{square}$, $I^{(n)}_{cubic}$ for the sums of all the terms $I^{(n)}$ on a chain, square lattice and a cubic lattice, respectively. Now we also use such abbreviations for the sums of nearest-neighbors couplings: the first sum in (11) will be called $I^{(0)xz}$ and for the second sum of (11) the name $I^{(0)y}$ will be used; subscripts will indicate a type of the lattice. Using this notation we write the fourth order contribution to H_{eff} on the chain in the form

$$\begin{aligned} H^{(4)}_{eff}(chain) = &-\frac{J^4}{72D^3} I^{(0)xz}_{chain} - \frac{J^4}{32D^3} I^{(0)y}_{chain} \\ &-\frac{J^4}{18D^3} I^{(2)}_{chain} - \frac{J^4}{24D^3} I^{(4)}_{chain}. \end{aligned} \quad (12)$$

In the case of the square lattice we have

$$\begin{aligned} H^{(4)}_{eff}(square) = &-\frac{53J^4}{72D^3} I^{(0)xz}_{square} - \frac{J^4}{32D^3} I^{(0)y}_{square} \\ &-\frac{7J^4}{18D^3} I^{(1)}_{square} - \frac{J^4}{18D^3} I^{(2)}_{square} \\ &-\frac{13J^4}{72D^3} I^{(3)}_{square} - \frac{J^4}{24D^3} I^{(4)}_{square} \\ &-\frac{5J^4}{24D^3} I^{(5)}_{square}, \end{aligned} \quad (13)$$

and in the case of the simple cubic lattice we get

$$\begin{aligned} H^{(4)}_{eff}(cubic) = &-\frac{35J^4}{24D^3} I^{(0)xz}_{cubic} - \frac{J^4}{32D^3} I^{(0)y}_{cubic} \\ &-\frac{7J^4}{18D^3} I^{(1)}_{cubic} - \frac{J^4}{18D^3} I^{(2)}_{cubic} \\ &-\frac{13J^4}{72D^3} I^{(3)}_{cubic} - \frac{J^4}{24D^3} I^{(4)}_{cubic} \\ &-\frac{5J^4}{24D^3} I^{(5)}_{cubic}. \end{aligned} \quad (14)$$

In order to understand the role of the symmetry breaking terms $I^{(3)}$ and $I^{(4)}$ we apply a simple approximation. The main part of H_{eff} is the rotationally invariant XY-type Hamiltonian $H^{(2)}_{eff}$. It is known to have a spontaneous magnetization in its XZ plane, in $d = 2$ and 3. We take a classical approximation for the ground state of $H^{(2)}_{eff}$ in $d = 2$ and 3. For one "spin" we define the state

$$|\phi\rangle = C(\phi)[(1 + \sin\phi)\ |+\rangle + \cos\phi\ |-\rangle], \quad (15)$$

where $C(\phi)$ is a normalization factor such that $\langle\phi\,|\phi\rangle = 1$. This state satisfies the equation

$$(\sigma^x \cos\phi + \sigma^z \sin\phi)\ |\phi\rangle = 1\ |\phi\rangle.$$

The function $|\phi\rangle$ describes the "spin" lying in the XZ plane along the direction given by the angle ϕ, which is measured with respect to the axis X. This function has the following properties

$$\begin{aligned} \langle\phi\mid\sigma^x\mid\phi\rangle &= \cos\phi, \\ \langle\phi\mid\sigma^y\mid\phi\rangle &= 0, \\ \langle\phi\mid\sigma^z\mid\phi\rangle &= \sin\phi. \end{aligned}$$

Our approximation for the ground state of $H^{(2)}_{eff}$ in $d = 2$ and 3 is

$$|\Phi\rangle = \prod_{i=1}^{N} |\phi_i\rangle, \quad (16)$$

where the product is taken over all lattice sites. Now we treat $|\Phi\rangle$ as a trial wave function and compute

$$E_\phi = \langle \Phi \,|\, H_{eff} \,|\Phi\rangle$$

to check which directions of ordering are preferred by the symmetry breaking terms $I^{(3)}$ and $I^{(4)}$. In the case of the square lattice we obtain

$$E_\phi^{(d=2)}/N = \frac{29}{36}\frac{J^4}{D^3}\sin(3\phi) + const.,$$

and for the cubic lattice

$$E_\phi^{(d=3)}/N = \frac{55}{24}\frac{J^4}{D^3}\sin(3\phi) + const.$$

Finding the minima in the above functions, we conclude that the H_{eff} with the fourth order symmetry breaking terms possesses three equivalent directions of magnetic ordering: $\phi = \pi/2$, $(7/6)\pi$, $-(\pi/6)$. The function $|\Phi\rangle$ for these values of ϕ will be our approximation for the ground state of H_{eff} in $d = 2$ and 3.

In the case of the chain the main term of the effective Hamiltonian has no long-range order. However, the model described by $H^{(2)}_{eff}$ is critical, and the small symmetry breaking perturbations from $H^{(4)}_{eff}$ are added to it. We expect, that these perturbations will stabilize the magnetic LRO in the effective $S = 1/2$ model in $d = 1$, although in this case our argument is weaker than for $d = 2$ and 3.

After establishing the ground-state properties of H_{eff} we must find the relation between the original $S = 2$ spin operators S_i^α and the operators σ_i^α of the effective $S = 1/2$ model. This is done according to the relation (8). We consider the following operators

$$S_i^\alpha, \qquad (S_i^\alpha)^2 - 2, \qquad S_i^\alpha S_i^\beta + S_i^\beta S_i^\alpha \tag{17}$$

where $\alpha, \beta = x, y, z$ and $\alpha \neq \beta$. The perturbational calculation of the mapping (8) in a given order of expansion is more tedious than the calculation of H_{eff} in the same order. The effective Hamiltonian was found up to the fourth order because the crucial symmetry breaking terms appeared only in the fourth order. We have made a rather crude approximation for finding the ground state of H_{eff}, therefore we will be satisfied with lower orders of expansion in the calculation of mappings of the operators (17). Up to the third order we find

$$\Gamma^\dagger S_i^\alpha \Gamma = 0,$$
$$\Gamma^\dagger (S_i^\alpha S_i^\beta + S_i^\beta S_i^\alpha)\Gamma = 0.$$

In this order of the perturbation theory there is no possibility of obtaining a magnetic order in our model (2), it agrees with the MFA result.

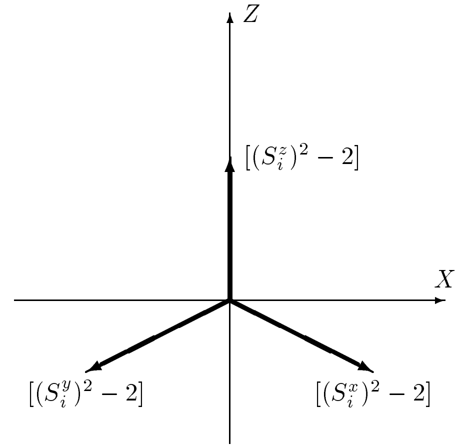


Fig. 4. Correspondence between quadrupolar operators $[(S_i^\alpha)^2 - 2]$ and directions in the plane XZ of the effective model.

Nonvanishing expressions are obtained for the mappings of operators $[(S_i^\alpha)^2 - 2]$

$$\Gamma^\dagger[(S_i^x)^2 - 2]\Gamma = \sqrt{3}\sigma_i^x - \sigma_i^z + O\left(\frac{J^2}{D^2}\right),$$
$$\Gamma^\dagger[(S_i^y)^2 - 2]\Gamma = -\sqrt{3}\sigma_i^x - \sigma_i^z + O\left(\frac{J^2}{D^2}\right),$$
$$\Gamma^\dagger[(S_i^z)^2 - 2]\Gamma = 2\sigma_i^z + O\left(\frac{J^2}{D^2}\right)\cdot \tag{18}$$

One can see that *magnetic order* in the effective model with $S = 1/2$ implies *quadrupolar order* in the initial model (2). The three operators $[(S_i^\alpha)^2 - 2]$ correspond to three directions in the XZ plane of the effective model. According to (18), the operator $[(S_i^z)^2 - 2]$ is represented by the operator $2\sigma_i^z$, which projects the effective "spin" on the direction $(0, 1)$ in the XZ plane. In the same way the operators $[(S_i^x)^2 - 2]$ and $[(S_i^y)^2 - 2]$ correspond to the projections of the effective "spin" on the directions $(\sqrt{3}, -1)$ and $(-\sqrt{3}, -1)$, respectively (see Fig. 4). These three directions coincide with the previously found directions of spontaneous ordering of the effective model.

Now, in the case of $d = 2$ and $d = 3$, we can use the approximate wave function $|\Phi\rangle$ of the effective model to calculate the ground-state averages of the operators $[(S_i^\alpha)^2 - 2]$. The zeroth order approximations of $\langle (S_i^\alpha)^2 - 2\rangle$, obtained according to (18), will be denoted by $q_\alpha^{(0)}$. For the three possible directions ϕ of ordering in the $S = 1/2$ model we have three sets of $q_\alpha^{(0)}$

$$\{q_x^{(0)}, q_y^{(0)}, q_z^{(0)}\} = \{2, -1, -1\} \quad \text{for} \quad \phi = -\frac{\pi}{6},$$
$$\{q_x^{(0)}, q_y^{(0)}, q_z^{(0)}\} = \{-1, 2, -1\} \quad \text{for} \quad \phi = \frac{7}{6}\pi,$$
$$\{q_x^{(0)}, q_y^{(0)}, q_z^{(0)}\} = \{-1, -1, 2\} \quad \text{for} \quad \phi = \frac{\pi}{2}. \tag{19}$$

In classical terms, one can say that the obtained quadrupols are of prolate shape, they are stretched along a chosen axis.

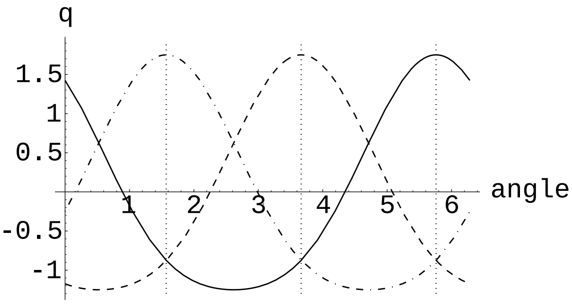


Fig. 5. Second-order averages $q_x^{(2)}$ (solid line), $q_y^{(2)}$ (dashed line) and $q_z^{(2)}$ (dashed-dotted line) *versus* the angle ϕ. The values $(J/D) = 1$ and $z = 6$ were chosen to make the second-order corrections visible. Vertical dotted lines show directions of ordering in the effective model (angle $\phi = (\pi/2), (7/6)\pi, (11/6)\pi$).

The second order corrections to (18) are a bit more complicated: in this order the representation of $[(S_i^\alpha)^2 - 2]$ involves not only operators σ_i^α but also operators from neighboring sites. For $[(S_i^z)^2 - 2]$ up to the second order we get

$$\Gamma^\dagger[(S_i^z)^2 - 2]\Gamma = 2\sigma_i^z + \left(\frac{J}{D}\right)^2 \left[-\frac{z}{12}\sigma_i^z - \frac{1}{24}\sigma_i^x \sum_j \sigma_j^x + \frac{1}{24}\sigma_i^z \sum_j \sigma_j^z\right] \quad (20)$$

where the sums are over nearest neighbors of the site i. More lengthy formulas are obtained for $[(S_i^x)^2 - 2]$ and $[(S_i^y)^2 - 2]$, one can calculate these expressions by applying rotations through the angle $\pm(2\pi)/3$ to (20).

The second-order corrections essentially do not change the picture obtained from (18). Applying the wave function $|\Phi\rangle$ with three possible values of ϕ we obtain three sets of $q_\alpha^{(2)}$ (second-order averages of $[(S_i^\alpha)^2 - 2]$). For $\phi = (\pi/2)$ we get

$$q_z^{(2)} = 2 - \frac{z}{24}\left(\frac{J}{D}\right)^2,$$

$$q_x^{(2)} = q_y^{(2)} = -1 + \frac{z}{48}\left(\frac{J}{D}\right)^2,$$

and identical relations, but with x,y and z interchanged, hold for $\phi = -(\pi/6)$ and $\phi = (7/6)\pi$. The second-order corrections reduce the averages of quadrupolar operators $[(S_i^\alpha)^2 - 2]$ from the saturation values, as it should be, since the two terms in (2) do not commute. The dependence of $q_\alpha^{(2)}$ on the angle ϕ is plotted in Figure 5. To conclude, it follows from the properties of the effective Hamiltonian H_{eff}, which has three directions of magnetic ordering, that the initial model (2) possesses the quadrupolar order in the case of $J/D \ll 1$. The above conclusion was reached with the help of the approximation for the ground state of H_{eff}, which is justified for $d = 2$ and 3.

As far as the chain is concerned, if we assume that the symmetry breaking perturbations from $H_{\mathit{eff}}^{(4)}$ stabilize the magnetic LRO in the effective model, then we have quadrupolar order in the initial model (2). If this is the case, the quadrupolar order should disappear asymptotically with $(D/J) \to \infty$, because then the symmetry breaking term $H_{\mathit{eff}}^{(4)}$ becomes smaller in comparison to the main term of the effective Hamiltonian ($H_{\mathit{eff}}^{(2)}$), and the critical XY-type model is approached.

4 Conclusions

The perturbation theory has been used to study the ground state properties of the $S = 2$ ferromagnets with cubic single-ion anisotropy in 1, 2 and 3 dimensions. The outcome of the paper is that in opposition to the MFA prediction the quadrupolar order can exist at $T = 0$ in the non-magnetic state of the system without quadrupolar interactions. This is a pure quantum effect which could not be observed in the case of classical spins. The result contradicts the common notion that the MFA predictions should be qualitatively correct in 3 dimensions.

The calculations presented in this paper allow us to assume that the quadrupolar order should exist, in the model under consideration, not only at the ground state but also at finite temperature, at least for $d = 3$. It follows from the fact, that, at the second order of approximation, our system is described by the effective Hamiltonian of the XY type, which for $d = 3$ exhibits the long range order also at finite temperature.

As mentioned in the previous section for $d = 1$ at $T = 0$ the existence of a long range order could be caused by the higher order corrections breaking the continuous symmetry of the effective Hamiltonian. When D/J goes to infinity one can expect a decrease of the quadrupolar order and an increase of the correlation length as a critical state is approached.

It is our pleasure to thank Gábor Fáth for interesting discussions. M.D. would also like to thank Romek Lemański for sharing his experience in perturbational calculations.

References

1. A. Aharony, Phys. Rev. B **8**, 4270 (1973).
2. H. Kleinert, S. Thoms, V. Schulte-Frohlinde, Phys. Rev. B **56**, 14 428 (1997).
3. J. Zittartz, private communication.
4. J. Sznajd, J. Magn. Magn. Mater. **42**, 269 (1984).
5. V.V. Valkov, T.A. Valkova, Teor. Mat. Phys. **59**, 269 (1984).
6. A.I. Mitsek, K.Y. Guslienko, S.V. Pavlovskii, Phys. Stat. Sol. (b) **135**, 173 (1986).
7. Z. Domański, J. Sznajd, Phys. Stat. Sol. (b) **129**, 135 (1985).
8. P.W. Anderson, Phys. Rev. **115**, 2 (1959).
9. L.N. Bulaevskii, Zh. Exp. Teor. Fiz. **51**, 230 (1966).
10. L.N. Bulaevskii, D.I. Khomskii, Fiz. Tverd. Tela **14**, 3594 (1972).
11. M. Takahashi, J. Phys. C, **10**, 1289 (1977).
12. T. Kato, Prog. Theor. Phys. **4**, 514 (1949).
13. T. Kennedy, E.H. Lieb, B. Sriram Shastry, Phys. Rev. Lett. **61**, 2582 (1988).