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Phase diagram of a spin-1 non-Heisenberg antiferromagnet

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The phase transitions in temperature of an anisotropic antiferromagnet with a biquadratic exchange interaction are investigated. The conditions for realization of a quadrupolar phase in the study are determined, and the critical temperatures of the transition to the paramagnetic phase are found. The phase diagram are constructed for different relations among the material constants. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1630718]

INTRODUCTION

The study of magnets with antiferromagnetic ordering has always attracted great interest from both theorists and experimentalists.[1–9] This is primarily because the technical implementation of the experimental studies on antiferromagnets is considerably simpler than for ferromagnets. As a rule, the Heisenberg model is used for describing antiferromagnetically ordered systems. However, there are a number of materials whose magnetic properties cannot be explained in the framework of that model. Such substances include, e.g., GdMg, EuS, and $CrBr_3$ (Refs. 10–13). In Refs. 10 and 12 it was conjectured that the magnetic properties of these compounds can be explained by taking the fourth-order (e.g., biquadratic) exchange interactions into account.

Interest in systems in which the influence of the biquadratic exchange interaction is substantial came up quite some time ago.[6–8] This interest arose, first, because non-Heisenberg ferromagnets can have a quadrupolar phase characterized by tensor order parameters. This is a clear example of the manifestation of the quantum properties of such systems. The non-Heisenberg character of the exchange interaction can be described by the following Hamiltonian:[7]

$$H=-\frac{1}{2}\sum_{n,m}\left[J\mathbf{S}_n\cdot\mathbf{S}_m+K(\mathbf{S}_n\cdot\mathbf{S}_m)^2\right], \tag{1}$$

where J and K are the bilinear and biquadratic exchange constants, respectively.

Such a Hamiltonian permits description of a wider class of phenomena than the Heisenberg model. In particular, it has been shown[7–9] that ferromagnetic, antiferromagnetic, and quadrupolar phase states can form, depending on the relationship of the exchange constants in (1).

For a 3D ferromagnet, when $J>0$ and $K>0$, a ferromagnetic phase ($J>K$) and a quadrupolar phase ($K>J$) can exist. In a ferromagnet with a negative biquadratic interaction constant ($K<0$) a ferromagnetic, quadrupolar, or quadrupolar–ferromagnetic phase can be realized.[7,14]

This raises the question of what phases can be realized in a non-Heisenberg antiferromagnet, i.e., when $J<0$ and $K\neq 0$. The goal of this study is to examine this question in the framework of spin-wave theory.

The nature of the magnetism in two-dimensional systems is of fundamental interest, since it differs from the magnetism of three-dimensional systems. It is known that there is no long-range magnetic order at any finite temperature in an isotropic two-dimensional magnet.[15] The question of stabilization of long-range magnetic order in non-Heisenberg ferromagnets was studied in Refs. 16–18. However, as far as we know, the question of the stabilization of long-range magnetic order in a two-dimensional non-Heisenberg antiferromagnet has not been investigated. This question is the subject of the present study.

ANTIFERROMAGNET WITH "EASY PLANE" ANISOTROPY

Let us consider the question of the existence of a quadrupolar (Q) phase in an antiferromagnet with single-ion anisotropy (SA) of the "easy plane" type for the case $K>0$. The antiferromagnet is two-sublattice, with equivalent sublattices, and the magnetic ion has spin $S=1$. We choose the coordinate system such that the XOZ plane coincides with the basal plane of the system. The Hamiltonian of such a system can be written in the form

$$H=-\frac{1}{2}\sum_{n,m}\left[J\mathbf{S}_n\cdot\mathbf{S}_m+K(\mathbf{S}_n\cdot\mathbf{S}_m)^2\right]+\frac{\beta}{2}\sum_n (S_n^y)^2, \tag{2}$$

where $\beta>0$ is the single-ion anisotropy constant. In Hamiltonian (2) each bond is counted twice, and the summation is over z nearest neighbors.

In the exchange part of Hamiltonian (2) we separate the mean field and the additional fields B_2^p ($p=0,2$) due to the quadrupole moments; we then obtain the one-site Hamiltonian $H_0(n)$:

$$H_0(n)=-\overline{H}S_n^z-B_2^0O_{2n}^0-B_2^2O_{2n}^2+\frac{\beta}{2}(S_n^y)^2, \tag{3}$$

where

$$\overline{H}=\left(J+\frac{K}{2}\right)\langle S_n^z\rangle;\quad B_2^0=\frac{K}{6}q_2^0;\quad B_2^2=\frac{K}{2}q_2^2;$$

$$O_{2n}^0=3(S_n^z)^2-S(S+1);\quad O_{2n}^2=\frac{1}{2}\left[(S_n^+)^2+(S_n^-)^2\right];$$

$q_2^0=\langle O_{2n}^0\rangle$, $q_2^2=\langle O_{2n}^2\rangle$ are the quadrupolar order parameters. Here and below we use the notation $J=Jz$, $K=Kz$, where z is the number of nearest neighbors.

In separating the mean field and the additional quadrupole fields, we assume, as in Ref. 19, that in the first sublattice $S_n^x=1/2(S_n^+ + S_n^-)$, $S_n^y=1/2i\,(S_n^+ - S_n^-)$, S_n^z, and in the second sublattice one can make the following substitution: $S_m^x\to S_m^x$, $S_m^y\to -S_m^y$, $S_m^z\to -S_m^z$.

Solving the one-site problem with Hamiltonian (3), we determine the energy levels of the magnetic ion,

$$E_1=\frac{\beta}{4}-B_2^0-\chi,\quad E_0=\frac{\beta}{2}+2B_2^0,\quad E_{-1}=\frac{\beta}{4}-B_2^0+\chi \tag{4}$$

and the eigenfunctions of the one-site Hamiltonian

$$\Psi(1)=\cos\varphi|1\rangle+\sin\varphi|-1\rangle,\quad \Psi(0)=|0\rangle,$$
$$\Psi(-1)=-\sin\varphi|1\rangle+\cos\varphi|-1\rangle, \tag{5}$$

where $\chi^2=\overline{H}^2+(\beta/4+B_2^2)^2$, $|i\rangle$ are the eigenfunctions of the operator S^z $(i=1,0,-1)$,

$$\cos\psi=\sqrt{\frac{\chi+\overline{H}}{2\chi}},\quad \sin\varphi=\sqrt{\frac{\chi-\overline{H}}{2\chi}}.$$

Using the basis of eigenfunctions (5) of the magnetic ion, we construct the Hubbard operators $X_n^{MM'}=X_n^{\alpha(M'M)}\equiv|\Psi_n(M)\rangle\langle\Psi_n(M')|$ which describe the transition of the magnetic ion from the state M' to the state M. Here $\alpha(M'M)$ are the root vectors determined by the algebra of the Hubbard operators.[20] The spin operators are related to the Hubbard operators as

$$S_n^+=\sqrt{2}[\cos\varphi(X_n^{10}+X_n^{0-1})+\sin\varphi(X_n^{01}-X_n^{-10})],$$
$$S_n^z=\cos 2\varphi(X_n^{11}-X_n^{-1-1})-\sin 2\varphi(X_n^{1-1}+X_n^{-11}),$$
$$S_n^-=(S_n^+)^+. \tag{6}$$

In studying the phase states of the system we use the method of bosonization of the Hubbard operators.[21] The basic idea of the method is to construct the Bose analog of Hamiltonian (2). The first step consists in the diagonalization of the one-site Hamiltonian and representation of the spin operators in terms of Hubbard operators. Then the Hubbard operators X_n^α are associated to pseudo-Hubbard operators $\tilde{X}_n^\alpha$, which are related to the Bose creation and annihilation operators as follows:[22,23]

$$\tilde{X}_n^{11}=1-a_n^+a_n-b_n^+b_n;\quad \tilde{X}_n^{00}=a_n^+a_n;$$
$$\tilde{X}_n^{-1-1}=b_n^+b_n;\quad \tilde{X}_n^{10}=(1-a_n^+a_n-b_n^+b_n)a_n;$$
$$\tilde{X}_n^{01}=a_n^+;\quad \tilde{X}_n^{1-1}=(1-a_n^+a_n-b_n^+b_n)b_n;$$
$$\tilde{X}_n^{-11}=b_n^+;\quad \tilde{X}_n^{0-1}=a_n^+b_n;\quad \tilde{X}_n^{-10}=b_n^+a_n. \tag{7}$$

Here the a's are Bose operators corresponding to the transition of an ion from state 1 to state 0 and vice versa, and the operators b correspond to the transition from state 1 to state -1 and vice versa.

As we have said, for a certain relationship of the material constants in (2) an antiferromagnetic or quadrupolar phase is realized in the system. From the expressions (6) relating the spin operators and Hubbard operators one can obtain equations for the order parameters for $T\to 0$:

$$\langle S_n^z\rangle\approx\cos 2\varphi,\quad q_2^0\approx 1,\quad q_2^2\approx\sin 2\varphi. \tag{8}$$

The system of equations (8) has two solutions: {1}. $\langle S_N^z\rangle=\sqrt{1-(\beta/4J)^2}$, $q_2^0=1$, $q_2^2=\beta/4J$, which corresponds to an antiferromagnetic phase; {2}. $\langle S^z\rangle=0$, $q_2^0=1$, $q_2^2=1$, corresponding to a quadrupolar phase $(\beta\geqslant 4J)$. As can be seen from the solutions of system (8), the realization of a Q phase in an antiferromagnet does not depend on the values of the biquadratic exchange interaction constants but is determined solely by the single-ion anisotropy constant and the bilinear exchange interaction constant. It is easy to see that $\langle S^z\rangle<1$.

Let us assume that the relationship between the SA and Heisenberg exchange constants is such that an antiferromagnetic phase, corresponding to solution {1} is realized in the system. Using expressions (7), we rewrite (6) in terms of the Bose operators and write Hamiltonian (2) in the form

$$\begin{aligned}H^{(2)}=&\sum_k (E_0-E_1)a_k^+a_k+\sum_k (E_{-1}-E_1)b_k^+b_k\\&+\frac{1}{2}\sum_k\left(J+\frac{K}{2}\right)\gamma_k(a_k^+a_{-k}^++a_ka_{-k})\\&+\frac{1}{4}\sum_k K\gamma_k(a_k^+a_{-k}^++a_ka_{-k}-b_k^+b_{-k}^+-b_kb_{-k}\\&+2b_k^+b_k),\end{aligned} \tag{9}$$

where $\gamma_k=(\cos k_x+\cos k_y+\cos k_z)/3$ is the structure factor of a 3D antiferromagnet or $\gamma_k=(\cos k_x+\cos k_z)/2$ is the structure factor for a 2D system.

In Eq. (9) we have kept terms only to second-order in the creation and annihilation operators. Diagonalizing this Hamiltonian by means of a $u-v$ transformation, we get

$$H^{(2)}=\sum_k \varepsilon_\alpha(k)\alpha_k^+\alpha_k+\sum_k \varepsilon_\beta(k)\beta_k^+\beta_k, \tag{10}$$

where $\varepsilon_\alpha(k)$ and $\varepsilon_\beta(k)$ are, respectively, the spectra of the low-frequency and high-frequency magnons, which have the form

$$\varepsilon_\alpha(k)=\sqrt{\left[\frac{\beta}{4}+(J+K)(1-\gamma_k)\right]\left[\frac{\beta}{4}+(J+K)(1+\gamma_k)\right]},$$
$$\varepsilon_\beta(k)=\sqrt{[2J+K(1-\gamma_k)][2J+K(1+\gamma_k)]}. \tag{11}$$

We now assume that the relation between the SA and exchange interaction constants is such that a quadrupolar phase is realized in the system, i.e., $\langle S_n^z\rangle=0$, $q_2^0=1$, $q_2^2=1$; in that case a degeneracy of the excited energy levels occurs $(E_0=E_{-1})$. In this phase the Hamiltonian (2), written in terms of the magnon creation and annihilation operators, has the form

$$H^{(2)}=\sum_k (E_0-E_1)a_k^+a_k+\sum_k (E_{-1}-E_1)b_k^+b_k$$
$$+\frac{1}{2}\sum_k \left(J+\frac{K}{2}\right)\gamma_k(a_k^+a_{-k}^++a_ka_{-k}+2a_k^+a_k$$
$$+b_k^+b_{-k}^++b_kb_{-k}+2b_k^+b_k)+\frac{1}{4}\sum_k K\gamma_k(a_k^+a_{-k}^+$$
$$+a_ka_{-k}-2a_k^+a_k+b_k^+b_{-k}^++b_kb_{-k}-2b_k^+b_k). \tag{12}$$

Diagonalizing Hamiltonian (12) by a $u-v$ transformation, we obtain the magnon spectra in the Q phase. Here we must take into account that the energy levels of the magnetic ion are degenerate in the Q phase. Because of this, the spectra of the a and b magnons are the same:

$$\varepsilon_\alpha(k)=\varepsilon_\beta(k)=\sqrt{\left[\frac{\beta}{2}+K(1-\gamma_k)\right]\left[\frac{\beta}{2}+K(1+\gamma_k)+2J\gamma_k\right]}. \tag{13}$$

As was shown in Refs. 7, 9, and 24, in an isotropic non-Heisenberg ferromagnet a Q phase can be realized for $|J|<|K|$. Analysis of expression (13) shows that the existence of a Q phase is also possible in an anisotropic non-Heisenberg antiferromagnet for $\beta\geqslant 4J$.

However, it was shown in Ref. 9 that if the exchange interaction constants are related as $J=-J\cos\theta$, $K=-J\sin\theta$, $\theta\in[-\pi/2;0]$, then the Q phase is not realized in an isotropic non-Heisenberg antiferromagnet.

Let us consider an isotropic non-Heisenberg antiferromagnet described by Hamiltonian (1), assuming that $J<0$ and $K>0$. In this case we do not impose such rigid restrictions on the exchange interaction constants as in Ref. 9.

In this case the system of equations (8) has two solutions: {1}. $\langle S_n^z\rangle=1$, $q_2^0=1$, $q_2^2=0$, which corresponds to an antiferromagnetic phase; {2}. $\langle S^z\rangle=0$, $q_2^0=1$, $q_2^2=1$, which corresponds to a Q phase.

Let us assume that the system is found in the antiferromagnetic phase. The spectra of the low- and high-frequency magnons in this case are written in the form

$$\varepsilon_\alpha(k)=(J+K)\sqrt{1-\gamma_k^2},$$
$$\varepsilon_\beta(k)=\sqrt{(2J+K)^2-K^2\gamma_k^2}. \tag{14}$$

We consider the solution corresponding to the Q phase. The magnon spectra obtained from Eq. (13) for $\beta=0$ have the form

$$\varepsilon_\alpha(k)=\varepsilon_\beta(k)=\sqrt{K(1-\gamma_k)[K(1+\gamma_k)+2J\gamma_k]}. \tag{15}$$

Spectrum (15) becomes nonphysical (imaginary) if $\gamma_k<-K/(2J+K)$. This indicates the impossibility of realizing a quadrupolar phase in an isotropic non-Heisenberg antiferromagnet and confirms the numerical results of Ref. 9, since the magnon dispersion relation (15) does not hold for arbitrary values of the wave vector **k**. This allows us to state that the Q phase does not exist in an isotropic non-Heisenberg antiferromagnet for any relationship of the exchange constants.

PHASE TRANSITIONS IN TEMPERATURE

To determine the temperature T_N of the transition from the antiferromagnetic to the paramagnetic phase, we consider the order parameter $\langle S_n^z\rangle$:

$$\langle S_n^z\rangle=\frac{1}{N}\sum_n \langle 1-a_n^+a_n\rangle. \tag{16}$$

Expression (16) can be rewritten as

$$\langle S_n^z\rangle=1-\frac{1}{(2\pi)^d}\int_{-\pi}^{\pi}\frac{(u_k^2+v_k^2)d^dk}{\exp\left(\frac{\varepsilon_\alpha(k)}{T}\right)-1}-S(0), \tag{17}$$

where d is the dimensionality of the system; u_k and v_k are the parameters of the Bogolyubov transformation, $u_k^2+v_k^2=(E_0-E_1)/\varepsilon_\alpha(k)$; $S(0)$ are the zero-point vibrations, which are determined by the expression

$$S(0)=\frac{1}{(2\pi)^d}\int_{-\pi}^{\pi}v_k^2d^dk. \tag{18}$$

In the case of an isotropic antiferromagnet the zero-point vibrations are independent of the value of the exchange interactions, and $S(0)=0.078$. In the case of a 2D anisotropic antiferromagnet the zero-point vibrations are dependent on the value of the exchange interaction (a change in K from 0 to $1.5J$ will cause the value of $S(0)$ to increase from 0.167 to 0.177). In a 3D antiferromagnet the zero-point vibrations are almost a factor of 2 smaller than in a 2D antiferromagnet (a change in K from 0 to 1.5 will cause the value of $S(0)$ to increase from 0.075 to 0.077).

The antiferromagnetic–paramagnetic phase diagrams of the two-dimensional and three-dimensional anisotropic antiferromagnets in the variables $K-T$ are presented in Fig. 1.

To determine the temperature T_Q of the transition in an anisotropic antiferromagnet from the quadrupolar to the paramagnetic phase, we consider the order parameter q_2^2:

$$q_2^2=\frac{1}{N}\sum_n \langle 1-3a_n^+a_n\rangle. \tag{19}$$

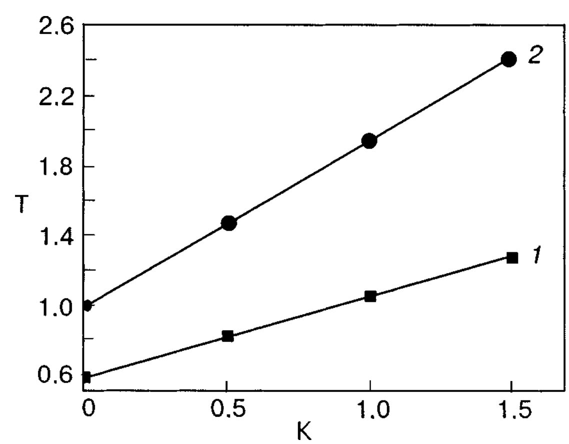

FIG. 1. Antiferromagnetic-paramagnetic phase diagram of an anisotropic non-Heisenberg antiferromagnet in the variables $K-T$ (K is the biquadratic exchange constant and T is the temperature (in units of J) ($\beta/J=0.02$): *1*—the line of the antiferromagnetic-paramagnetic phase transition for a 2D system; *2*—the line of the antiferromagnetic-paramagnetic phase transition for a 3D system.

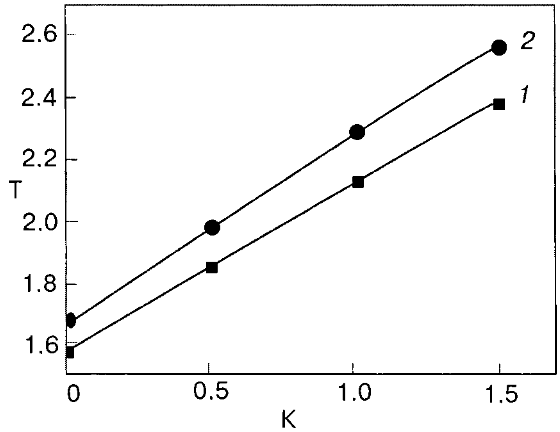

FIG. 2. Quadrupolar-paramagnetic phase diagram of an anisotropic non-Heisenberg antiferromagnet in the variables $K-T$ (K is the biquadratic exchange constant and T is the temperature (in units of J) ($\beta/J=5$): *1*—the line of the quadrupolar-paramagnetic phase transition for a 2D system; *2*—the line of the quadrupolar-paramagnetic phase transition for a 3D system.

In Eq. (19) we have taken into account that $\langle a_n^+ a_n\rangle = \langle b_n^+ b_n\rangle$ in the Q phase because of the degeneracy of the energy levels ($E_0=E_{-1}$).

Expression (19) can be rewritten as

$$q_2^2=1-\frac{3}{(2\pi)^d}\int_{-\pi}^{\pi}\frac{(u_k^2+v_k^2)d^dk}{\exp\left(\frac{\varepsilon_\alpha(k)}{T}\right)-1}-q(0), \tag{20}$$

where

$$q(0)=\frac{1}{(2\pi)^d}\int_{-\pi}^{\pi}v_k^2 d^dk$$

are the zero-point vibrations in the Q phase.

The zero-point vibrations in the Q phase are smaller than in the antiferromagnetic phase: for a 2D system a change in K from 0 to $1.5J$ will increase the value of $q(0)$ from 0.016 to 0.037; for a 3D system the zero-point vibrations, as in the antiferromagnetic phase, are almost a factor of 2 smaller than in the 2D case; increasing K from 0 to $1.5J$ increases $q(0)$ from 0.009 to 0.022.

The quadrupolar–paramagnetic phase diagrams of 2D and 3D anisotropic antiferromagnets in the variables $K-T$ are shown in Fig. 2.

CONCLUSION

The goal of this study was to explore the possibility of realization of a quadrupolar phase in a non-Heisenberg antiferromagnet with spin 1. The studies showed that in an anisotropic non-Heisenberg antiferromagnet with "easy plane" anisotropy the appearance of a quadrupolar phase is possible for $\beta \geqslant 4J$, independently of the value of the biquadratic exchange interaction. In isotropic 2D and 3D non-Heisenberg antiferromagnets the quadrupolar phase is not realized under any conditions. Analogous results were obtained by a numerical simulation in Ref. 9. It should be noted, however, that in the present study the value of the exchange interaction constant was not so rigidly restricted as in Ref. 9.

Using expressions (17) and (20), we constructed phase diagrams of 2D and 3D anisotropic non-Heisenberg antiferromagnets for different values of the biquadratic exchange interaction constant. In comparing the phase diagrams of the 2D and 3D antiferromagnets (Fig. 1), one notices that the temperature of the transition from the antiferromagnetic to the paramagnetic phase for a 2D antiferromagnet is almost a factor of 2 lower than for the 3D system. This is due to the fact that fluctuations in the 3D systems are significantly smaller than in 2D systems, and that affects the value of the Néel temperature. The temperature of the transition from the quadrupolar to the paramagnetic phase (see Fig. 2) is practically the same for 2D and 3D antiferromagnets. Such a weak dependence of the transition temperature on the dimensionality of the system can be explained, we believe, by the circumstance that the quadrupolar phase is "less ordered" than the antiferromagnetic phase. This "disorder" is due to the fact that the magnetizations of the sublattices equal zero and to the specific structure of the ground-state wave functions, which in the quadrupolar phase are a superposition of the states $|1\rangle$ and $|-1\rangle$ of the operator S^z (see expression (5), with $\cos\varphi=\sin\varphi=1/\sqrt{2}$). The small difference in the phase diagrams in the quadrupolar phase is due to the fact that the zero-point vibrations are larger for 2D systems than for 3D systems.

Thus the dimensionality of the system has a substantial influence on the temperature of the transition to the paramagnetic phase only in the antiferromagnetic phase.

The phase diagram of a 3D isotropic non-Heisenberg antiferromagnet in the antiferromagnetic phase is similar to that of a 3D anisotropic non-Heisenberg antiferromagnet. The small differences in the phase transition temperatures are due to the differences in the zero-point vibrations and the value of the single-ion anisotropy.

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