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## Biquadratic exchange and first-order ferromagnetic phase transitions

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We have calculated some properties of a ferromagnet using a Heisenberg Hamiltonian with a biquadratic exchange term added. The constant-coupling approximation was employed. If the strength of the biquadratic exchange relative to the bilinear is given by a dimensionless parameter  $\alpha$ , there is a critical value  $\alpha_c$  at which the phase transition changes character from second order for  $\alpha < \alpha_c$  to first order for  $\alpha > \alpha_c$ . The spontaneous magnetization, the exchange energy, and the spin-correlation function show discontinuous jumps at  $\alpha = \alpha_c$  and unstable behavior for  $\alpha > \alpha_c$ .

### INTRODUCTION

Some calculations are presented here on the properties of a ferromagnet with the two-particle Hamiltonian:

$$\mathcal{H} = -2J[\vec{S}_1 \cdot \vec{S}_2 + \alpha(\vec{S}_1 \cdot \vec{S}_2)^2] - \mu H(S_{1z} + S_{2z}). \quad (1)$$

This is the Heisenberg Hamiltonian including biquadratic exchange, the strength of which can be adjusted by the numerical parameter  $\alpha$ . For  $\alpha$  varying between 0 and 1, the results for the spin quantum number  $S_0 = 1$  have already been published.<sup>1</sup> We now extend these calculations to a wider range of  $\alpha$  and higher spin values employing the same technique as in Ref. 1, namely, the constant-coupling approximation.<sup>2</sup> It is found that the ferro-para phase transition changes from second order to first order for large enough  $\alpha$ .

The existence of a biquadratic term has been argued on several theoretical grounds (see Ref. 1, Refs. 2-9 therein), but these all lead to a quite small and sometimes negative value for  $\alpha$ . However, in the present calculation, we employ rather large values of  $\alpha$  whose origin must be explained. We offer the following as a possibility: The two-

particle Hamiltonian (1) is obtained, as explained in Ref. 2, by taking the trace of the  $N$ -particle density matrix  $\rho_N$   $N-2$  times to obtain  $\rho_2 \sim e^{-\beta \mathcal{H}}$ . That is, one does this in principle and then asks: On what can the effective two-particle Hamiltonian  $\mathcal{H}$  depend? The answer, for spin  $\frac{1}{2}$ , is given in Ref. 2 to be that  $\mathcal{H}$  can be a linear combination of  $\vec{S}_1 \cdot \vec{S}_2$ ,  $\sigma_{1x}\sigma_{2x}$ , and  $\sigma_{1x} + \sigma_{2x}$ . That is, allowable functions of the spin operators  $\vec{S}_1$  and  $\vec{S}_2$  can be reduced to a linear combination of these three terms. If the middle term is discarded on the reasonable assumption that the total exchange contribution remains spherically symmetric, and the coefficient of the first taken to be  $J$ , independent of  $T$ , then only the coefficient of the Zeeman term remains to be evaluated.

However, this argument does not apply if the spin is greater than  $\frac{1}{2}$  because functions of the spin operators do not then reduce to such simple linear combinations. The effective Hamiltonian for spin 1 could include, at the very least, a term in  $(\vec{S}_1 \cdot \vec{S}_2)^2$  with an arbitrary—i.e., not necessarily small—coefficient. This leads to our consideration of the Hamiltonian (1) with unrestricted values for  $\alpha$ .

The Hamiltonian (1) has eigenvalues  $E_{SM}$  easily found as functions of  $\vec{S} = \vec{S}_1 + \vec{S}_2$  and  $M = S_{1z} + S_{2z}$ . The partition function is then obtained by summing

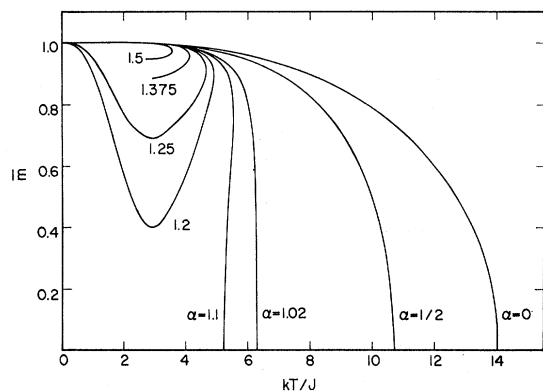


FIG. 1. Magnetization  $\bar{m}$  vs temperature  $kT/J$  for a face-centered-cubic lattice of spin-1 atoms for various values of the biquadratic exchange strength  $\alpha$ .

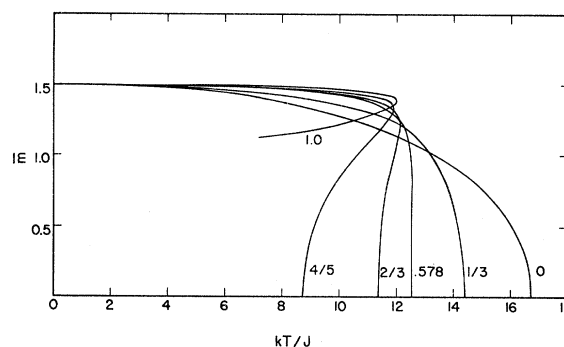


FIG. 2. Same as Fig. 1 but for a body-centered-cubic lattice of spin- $\frac{3}{2}$  atoms.

TABLE I. Critical values of the biquadratic exchange constant  $\alpha$ .

$n$	8	12	8	12	8	12
$S_0$	1	1	$\frac{3}{2}$	$\frac{3}{2}$	2	2
$\alpha_c$	1.02	1.02	0.578	0.535	0.335	0.305

$e^{-\beta E_{SM}}$  over  $M$  from  $-S$  to  $S$  and  $S$  from 0 to  $2S_0$ . The following properties have been calculated following the procedure in Ref. 1: the spontaneous magnetization per atom (in units of  $\mu_B \bar{m}$ ; the exchange energy per atom (in units of  $J$ )  $\bar{E}$ , and the spin correlation function

$$\bar{r} = \langle \vec{S}_1 \cdot \vec{S}_2 \rangle.$$

In Figs. 1 and 2 are shown the magnetization-temperature curves for  $S_0 = 1, \frac{3}{2}$ , and  $n = 12, 8$ , respectively, for various values of  $\alpha$  ( $n$  is the lattice coordination number). Both sets of curves are qualitatively similar as are those for  $S_0 = 1$  (not shown). As  $\alpha$  is increased, the Curie-temperature decreases and the magnetization changes more abruptly near  $T_c$ . For a certain critical value  $\alpha = \alpha_c$ , the curves show a vertical drop; the magnetization falls from  $\frac{1}{3}$  or  $\frac{1}{2}$  of saturation to zero for a temperature change of less than one part in  $10^3$ . Above  $\alpha_c$ , the magnetization becomes double valued in a region of temperature near the maximum. The curves reach a maximum in temperature and then bend back toward lower  $T$ , forming a "nose." At the tip of this "nose" where the slope becomes infinite, the system undergoes a first-order phase transition and the magnetization drops abruptly to zero. We call this temperature  $T_1$ .

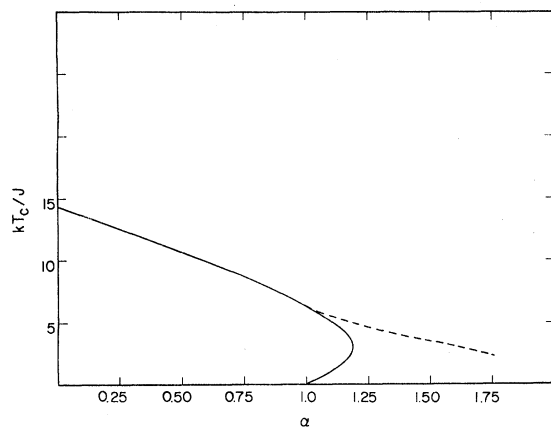


FIG. 3. Critical temperature vs the biquadratic exchange strength  $\alpha$  for a face-centered-cubic lattice of spin-1 atoms. The solid line gives the values of the temperature  $T_0$  at which the magnetization vanishes. The dashed portion gives the values of  $T_1$ , the temperature at the "nose" of the magnetization curve where  $d\bar{m}/dT$  becomes infinite.

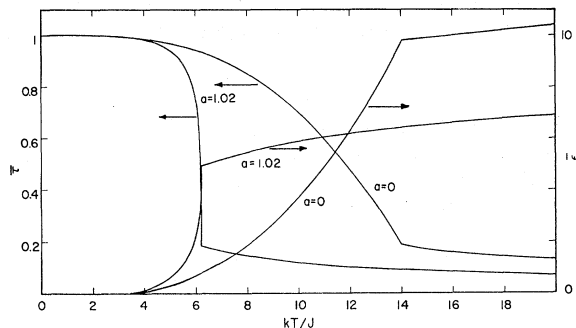


FIG. 4. Average exchange energy per atom (in units of  $J$ ) and the spin correlation function  $\bar{r} = \langle \vec{S}_1 \cdot \vec{S}_2 \rangle$  vs temperature for a face-centered-cubic lattice of spin-1 atoms for two values of  $\alpha$ , 0 and 1.02.

The values of  $\alpha_c$  are shown in Table I. (In the current literature, it is customary to call these points, at which the first-order transition takes over, tricritical points.)

The dependence of the transition temperature on  $\alpha$  is shown in Fig. 3 corresponding to the magnetization curves shown in Fig. 1. The solid lines represent the values of  $T_0$ , the temperatures at which the magnetization goes to zero. As  $\alpha$  increases,  $T_0$  shows a nearly linear decrease. For  $\alpha > 1/S_0$ , the curve is nearly parabolic and there are two roots. For a somewhat higher value of  $\alpha$  (1.18), the magnetization curves no longer go to zero and there is no  $T_0$ . Also shown in these figures, by the dashed lines, are the values of  $kT_1/J$ , the temperatures at which the magnetization curves have a vertical slope—the tip of the nose. This curve begins at  $\alpha_c$  and extends to  $\alpha \approx 1.77$ . Above this value of  $\alpha$  the equation giving the internal field (and therefore the magnetization) as a function of temperature has no nontrivial real roots (the zero-field solution always exists). The curves for other spins and lattices are qualitatively the same.

Somewhat similar results are also obtained by Chen and Levy<sup>3</sup> using both the molecular-field theory and high-temperature series expansions of the susceptibility. Our  $T_c$  vs  $\alpha$  curve falls between their two results for dipolar ordering. Chen and Levy also investigate quadrupolar transitions which have not been considered here. To do so would have required the introduction of another arbitrary parameter into the Hamiltonian to represent the internal quadrupolar field. [It is to be noted that the presence of such a term in the Hamiltonian would have no effect on the result for  $T_c$  vs  $\alpha$  in Fig. 3, contrary to the statement made by Chen and Levy<sup>3</sup> (Ref. 5 therein).] This was done by Westwanski<sup>4</sup> using a Green's-function technique to calculate the dipole and quadrupole correlation functions. Unfortunately, this does not lead to explicit numerical results.

The temperature dependence of the spin-correlation function and the average exchange energy are

shown in Fig. 4 for  $\alpha=0$  and  $\alpha_c$ . The change to a first order transition is evident.

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<sup>1</sup>H. A. Brown, Phys. Rev. B 4, 115 (1971).

<sup>2</sup>P. W. Kasteleijn and J. van Kranendonk, Physica 22, 317 (1956).

<sup>3</sup>H. H. Chen and P. M. Levy, Phys. Rev. B 7, 4267

(1973; 7, 4284 (1973)).

<sup>4</sup>B. Westwanski, Communications of the Joint Institute for Nuclear Research, Dubna, 1973 (unpublished).