Spin-1 exchange-interaction model of ferromagnetism

H. A. Brown

Physics Department, University of Missouri–Rolla,
Rolla, Missouri 65401

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The spin-exchange operator for spin $S = 1$ and a constant-coupling-like approximation are used to evaluate the thermodynamic properties of a system described by a two-spin Hamiltonian that includes dipole and quadrupole interactions. The dipole and quadrupole moments and the exchange energy are found as functions of temperature. There is a critical temperature, T_c, which depends on the coordination number of the lattice at which the system undergoes a second-order phase transition. Below T_c, both magnetic moments, simultaneously, take on nonzero values and have infinite derivatives there while the derivative of the exchange energy is discontinuous. Comparison is made with other results where possible and reasonable agreement found.

A brief history of the theoretical problem considered in this paper follows. In 1928, Heisenberg[1] proposed, as a possible source of ferromagnetism, the exchange interaction. This could be expressed as an operator involving the scalar product of the spins of two neighboring atoms, $\mathbf{S}_1 \cdot \mathbf{S}_2$. Dirac[2] showed that the permutation operator for the spins of two electrons has the form

$$P_{12} = \tfrac{1}{2}(1 + \boldsymbol{\sigma}_1 \cdot \boldsymbol{\sigma}_2) \ . \tag{1}$$

In 1941, Schrödinger[3] developed an expression for the spin-exchange operator for two identical particles of arbitrary spin. It included terms $(\mathbf{S}_1 \cdot \mathbf{S}_2)^\nu$ for $\nu = 0, 1, \ldots, 2S$. An application of this operator to the problem of ferromagnetism was made, using the high-temperature expansion (HTE) method, by Allan and Betts[4] in 1967 for a spin-1 system, for which

$$P_{12} = (\mathbf{S}_1 \cdot \mathbf{S}_2)^2 + \mathbf{S}_1 \cdot \mathbf{S}_2 - 1 \ . \tag{2}$$

Since then, numerous papers have appeared in which the original Heisenberg bilinear operator was modified by the addition of a biquadratic term, $(\mathbf{S}_1 \cdot \mathbf{S}_2)^2$, of arbitrary, adjustable magnitude. Much less numerous have been applications of the Schrödinger spin-exchange operator for $S = 1$ or greater. The present work is an addition to the latter group, using a pair of neighboring spins and a constant-coupling-like approximation.

We begin with the assumption that the effective two-spin Hamiltonian should be

$$\mathcal{H} = -JP_{12} - \mu H(S_{1z} + S_{2z}) - \nu K(S_{1z}^2 + S_{2z}^2) \ , \tag{3}$$

where J is the Heisenberg exchange integral and P_{12} is the spin-exchange operator for $S = 1$ [Eq. (2)]. The second term is the usual Zeeman energy, and the last term includes the quadrupole interactions. The parameter J fixes the temperature scale, while H and K are the dipolar and quadrupolar internal fields representing the interactions of the two spins with the rest of their neighbors, and are to be determined as functions of temperature. The magnetic dipole and quadrupole moments of the spins are given by μ and ν.

The eigenvalues of the Hamiltonian are found straightforwardly and the resulting partition function turns out to be

$$Z = e^p[1 + 2e^{2\kappa}\cosh(2\lambda)] + 2\cosh(p)[(e^{2\kappa} + 2e^\kappa \cosh(\lambda)] \ , \tag{4}$$

where

$$p = \beta J \ ,$$

$$\lambda = \beta\mu H \ ,$$

$$\kappa = \beta\nu K \ , \tag{5}$$

$$\beta = 1/kT \ .$$

The method used to evaluate the internal fields H and K has been shown,[5] in a similar calculation, to be equivalent to the constant-coupling approximation (CCA). Explicitly, the values of the dipole and quadrupole parameters

$$m = \frac{1}{2}\frac{\partial \ln Z}{\partial \lambda} \tag{6}$$

and

$$q = \frac{1}{2}\frac{\partial \ln Z}{\partial \kappa} \tag{7}$$

are evaluated and equated to those obtained from a one-spin (molecular field) Hamiltonian

$$\mathcal{H}' = -\mu H' S_z - \nu K' S_z^2 \ , \tag{8}$$

by

$$Z' = 1 + 2e^{\kappa'}\cosh\lambda' \ , \tag{9}$$

and

$$m' = \frac{\partial \ln Z'}{\partial \lambda'} \ , \tag{10}$$

$$q' = \frac{\partial \ln Z'}{\partial \kappa'} \ , \tag{11}$$

using the assumption that the internal fields are additive

$$\frac{H'}{H} = \frac{z}{z-1} = \frac{K'}{K} \ , \tag{12}$$

where z is the lattice coordination number.

The equations

$$m = m' \ , \qquad q = q' \ , \tag{13}$$

can both be solved numerically. Thus, the internal fields and m and q are found as functions of temperature. Also
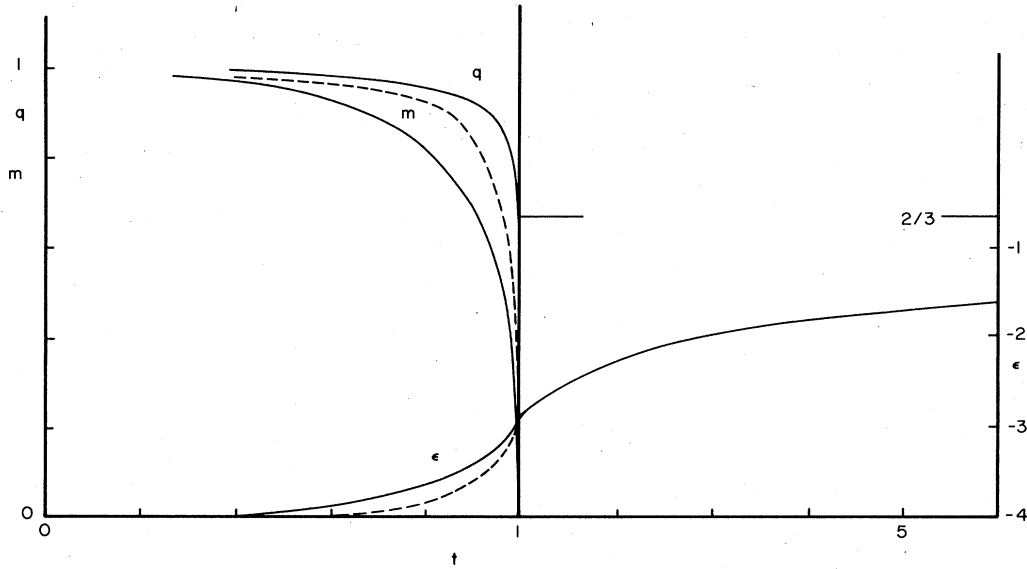
FIG. 1. The magnetization parameters $m = \langle S_z \rangle$ and $q = \langle S_z^2 \rangle$ and the exchange energy per atom $\epsilon = E_{\text{ex}}/J = 4\langle P \rangle$ for a body-centered cubic lattice $(z = 8)$ as functions of the reduced temperature $t = T/T_c$. Note the change in scale for $t > 1$. The dashed lines are the results without the quadrupole terms in (3).

obtainable is the average exchange energy per atom (in units of J):

$$\frac{E_{\text{ex}}}{J} = \epsilon = -\frac{z}{2} \frac{\partial \ln Z}{\partial p} \ . \tag{14}$$

An important result of the solutions for m and q is that there is just one critical temperature T_c and a very simple phase diagram. The system is magnetically disordered above T_c; below T_c both dipole and quadrupole moments are nonzero.

The algebraic solutions of Eqs. (13) show that the value of T_c would be unchanged if either K or H were assumed to be zero. The thermodynamic properties will be different, but not T_c. More specific conditions are as follows.

(a) If $K = 0$, T_c is found from the low-field limit, $H \rightarrow 0$. Then $q = q' = \frac{2}{3}$ is satisfied and $m = m'$ gives the critical temperature for dipole ordering as

$$p_c = \frac{J}{kTc} = \frac{1}{2} \ln \left| \frac{z+1}{z-5} \right| \ . \tag{15}$$

(b) If $H = 0$, $m \equiv 0 \equiv m'$, and with $K \rightarrow 0$, $q = q'$ gives the same critical temperature, but now the ordering is quadrupolar.

(c) If neither H nor $K = 0$, then the order in which the double limit H, $K \rightarrow 0$ is taken is not important, the only difference is which of Eqs. (13) defines T_c. Some other results show a similar phase diagram. Using the spin-exchange operator, Chen and Joseph[6] found that all the multipolar susceptibilities, X_ν, $\nu = 1, \ldots, 2S$, diverge at the same temperature. However, the Hamiltonian used to calculate X_ν included only one field term, proportional to S_z^ν, and so is not the same as (3).

For further comparisons we must go to the literature on biquadratic exchange. Using the molecular-field approximation (MFA), Chen and Levy[7] find that there is only one critical temperature for any α, but the symmetry of the or-

dered state is in doubt. The same authors evaluated the dipole and quadrupole susceptibilities, X_1 and X_2, using the HTE method[8] up to terms in T^{-4}. The coefficients in these series are used to find the singularities at, say, T_1 and T_2, the higher of the two taken to indicate the type of order below that temperature. In both calculations, the same ambiguity exists. For $\alpha < 1$, the transition is to ordered dipoles, for $\alpha > 1$, the transition is to quadrupoles. At $\alpha = 1$ (and $T_1 = T_2$) the situation is uncertain. Another MFA result[9] predicts that the purely quadrupolar phase will not exist for any α. A Green's function (GF) calculation[10] agrees. Another calculation using the CCA (Ref. 11) employs a Hamiltonian that is almost the Ising model equivalent of (3). There are differences that prevent any direct comparison, however, and in any event, the region near $\alpha = 1$ was not considered, only $\alpha \geqslant 2$. Note that usual-

TABLE I. Calculated values of kT_c/J. Numerical values of the reduced critical temperature vs the coordination number z from various calculations with the method of approximation employed (see text for abbreviations). The numbers in parentheses were obtained by the same authors in some of their earlier work.

z				Approximate
6	8	12	Reference	method
		3.09	4,6	HTE
2.5	3	4	14	GF
	2.4		15	GF
	(4.55)			
2.52–2.76			13	GF
(3.48)				
3.6		7.6	16	Oguchi (Ref. 17)
1.03	1.82	3.23	Present	

ly, but not uniformly, α is the ratio of the biquadratic to the bilinear exchange integrals, occasionally it is the inverse, or something else.

The thermodynamic functions m, q, and ϵ [Eqs. (6), (7), and (14)] are easily obtained as functions of temperature by numerical means and are shown in Fig. 1. The shapes of the magnetization and energy curves are typical of a second-order transition. Both m and q go from saturation—or complete order—at $T = 0$ to disorder ($m = 0, q = \frac{2}{3}$) at T_c where their slopes are infinite. The exchange energy rises from $-z/2$ to $-z/6$ in going from zero to infinite temperature, showing a sharp rise just below T_c and a discontinuous slope there, i.e., a discontinuous heat capacity. Qualitatively at least, this behavior is in general agreement with the observed properties of ferromagnets. The figure also shows the results of the calculation[12] with

the S_z^2 terms not included, i.e., with a Hamiltonian like (3) but with $K = 0$. It is seen that the absence of these terms leads to a magnetization and exchange energy that change more abruptly just below T_c.

There is little from other methods with which to compare these results. The HTE methods, of course, do not apply below T_c. Biquadratic exchange calculations, by whatever means, usually evaluate only the α dependence of T_c and either predict a first-order transition for $\alpha = 1$ (Refs. 9 and 13) or give ambiguous results.[7]

Finally, values of kT_c/J from various theories are collected in Table I. There is little evidence of agreement but the present result is gratifying close to the HTE value, which is usually considered "exact." Considerable scatter exists in the GF results, reflecting their sensitivity to the mode of decoupling.

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