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$$H = J[S_1 a_2^\dagger a_2 + S_2 a_2^\dagger a_1 + \sqrt{(S_1 S_2)}(a_1^\dagger a_2^\dagger + a_1 a_2)]$$

is $J(S_1 - S_2)$. If we now imagine $\vec{S}_1$ to be the total spin of the z nearest neighbors of the impurity and $\vec{S}_2$ to be the impurity spin, we can see that quadratic Bose Hamiltonian underestimates the energy of the s_0 mode when the impurity-host exchange is much larger than any other interactions present.

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Ising Model with Antiferromagnetic Next-Nearest-Neighbor Coupling. II. Ground States and Phase Diagrams*

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The critical values of the ratio of interaction energies at which the ordering of spins in the ground state is indeterminate (and probably $T_c = 0$) are calculated exactly for a variety of Ising lattices with antiferromagnetic next-nearest-neighbor coupling, and for anisotropic nearest-neighbor coupling. Using these results and the exact two-dimensional solutions, the general dependence of the critical point on interaction ratio is sketched. The complex case of the antiferromagnetic fcc is reconsidered.

The effects of introducing higher neighbor interactions in an Ising model have been investigated recently by series expansion methods,[1-3] by exact two-dimensional solutions [4,5] and by closed-form approximations.[6-8] One of the problems is to determine the dependence of the critical point (Curie point T_c) on the strength and sign of the next-nearest-neighbor interaction. It is generally expected that an antiferromagnetic next-nearest-neighbor interaction will depress the critical point, and if sufficiently

strong will alter the structure of the ground state. For some critical value of the ratio of next-nearest-neighbor (nnn) to nearest-neighbor (nn) interaction energy, it is expected that T_c will vanish. Domb and Potts[1] showed that for the square lattice $T_c = 0$ when the nnn interaction is antiferromagnetic and half the strength of the nn interaction. Some preliminary estimates of the critical ratio have been made on some three-dimensional Ising lattices by Dalton and Wood.[3] In the present paper, we calculate the critical interaction ratio *exactly* for a variety of Ising lattices, and sketch the general dependence of critical point T_c and disorder point[9] T_D on nnn interaction strength (Fig. 1). Finally the problem of the antiferromagnetic face-centered cubic (fcc) lattice with anisotropic nn interaction is considered in the same context.

Consider an Ising model with "spin" variables $\sigma_{\vec{r}} = \pm 1$ at lattice sites $\vec{r}$, and Hamiltonian

$$\mathcal{H} = -J_1 \sum_{nn} \sigma_{\vec{r}} \sigma_{\vec{r}'} - J_2 \sum_{nnn} \sigma_{\vec{r}} \sigma_{\vec{r}'}, \tag{1}$$

where the first sum is over all nn pairs of spins and the second sum over all nnn pairs of spins. Each coupling is ferromagnetic or antiferromagnetic according as $J_{1,2} \gtrless 0$. Let q_1, q_2 be the coordination numbers of lattices containing, respectively, nn and nnn bonds only. There will be two equivalent nnn sublattices for loose-packed (nn) lattices and more than two for close-packed lattices. At $T = 0$ the spins acquire a configuration with the energy of the ground state, which is at least twofold degenerate. If $|J_1| \gg |J_2|$ the ground state is that of a lattice of spins with nn interactions J_1 only, which is ferromagnetic if $J_1 > 0$. On the other hand, if $|J_2| \gg |J_1|$, the ground-state configuration of spins on *each* sublattice of nnn spins is that with respect to nnn interactions J_2 within each sublattice only. Ground-state configurations of the nn and nnn (sub)lattices may

be compatible, as in a completely ferromagnetic lattice with J_1, $J_2 > 0$, for which all spins are parallel in a ground state. But when nn interactions are ferromagnetic ($J_1 > 0$) and nnn interactions antiferromagnetic ($J_2 < 0$) then ground-state configurations for $|J_1| \gg |J_2|$ and $|J_2| \gg |J_1|$ are not compatible, in general. A similar situation obtains when all interactions are antiferromagentic ($J_1, J_2 < 0$). Let $r = J_2/|J_1|$ and $\rho = q_2 r/q_1$. If we start with a ground-state arrangement for $|J_1| \gg |J_2|$ and increase the strength of the antiferromagnetic nnn interaction, then at some "critical" value ρ_c of the ratio ρ, the ground-state configuration will change over to that appropriate to a lattice with $|J_2| \gg |J_1|$. We are assuming that there is no third configuration which is the ground state for some intermediate range of ρ. (This question is discussed further below.) ρ_c may be determined by equating the expressions for the ground-state energies calculated for the cases $|J_1| \gg |J_2|$ and $|J_2| \gg |J_1|$, and solving for $\rho = \rho_c$. The value of the ground-state energy is a maximum when $\rho = \rho_c$, and the critical temperature T_c is then expected to be a minimum (probably zero).

Our results are summarized for a variety of one-, two-, and three-dimensional lattices in Table I where values of r_c, ρ_c, and the ground-state energies per spin for $|J_1| \gg |J_2|$ and $|J_2| \gg |J_1|$ are presented. In part A of Table I we include the more familiar regular lattices with nnn antiferromagnetic interactions introduced. In part B, we consider some *anisotropic* Ising lattices, which with appropriate choice of J_1 and J_2 also fall into a scheme of nn lattices with nnn interactions. For example, the two-dimensional triangular lattice may be regarded as a square lattice, with nn interaction J_1, and one set of diagonal bonds, with nnn interaction J_2. Similarly, we may regard the fcc as a nn bcc lattice and a set of nnn quadratic layers, by choosing the interactions within a principal plane to be

TABLE I. $U^{(1)}(0)$, $U^{(2)}(0)$ are the ground-state energies per spin when $|J_1| \gg$ or $\ll |J_2|$. q_1, q_2 are the coordination numbers of nn and nnn lattices. The critical ratio is $\rho_c = q_2 r_c/q_1 = q_2 J_2/q_1 |J_1|$.

	Lattice	q_1	q_2	$U^{(1)}(0)$, $|J_1| \gg |J_2|$	$U^{(2)}(0)$, $|J_2| \gg |J_1|$	r_c	ρ_c
	lc (1, 2)	2	2	$-|J_1| + |J_2|$	$-|J_2|$	$-\frac{1}{2}$	$-\frac{1}{2}$
	lca (1, 2)	2	1^a	$-|J_1| + \frac{1}{2}|J_2|$	$-\frac{1}{2}|J_2|$	-1	$-\frac{1}{2}$
	sq (1, 2)	4	4	$-2|J_1| + 2|J_2|$	$-2|J_2|$	$-\frac{1}{2}$	$-\frac{1}{2}$
	h (1, 2)	3	6	$-\frac{3}{2}|J_1| + 3|J_2|$	$-|J_2| - \frac{1}{2}|J_1|$	$-\frac{1}{4}$	$-\frac{1}{2}$
A	sc (1, 2)	6	12	$-3|J_1| + 6|J_2|$	$-2|J_2| - |J_1|$	$-\frac{1}{4}$	$-\frac{1}{2}$
	bcc (1, 2)	8	6	$-4|J_1| + 3|J_2|$	$-3|J_2|$	$-\frac{2}{3}$	$-\frac{1}{2}$
	fcc (1, 2) $J_1 > 0$	12	6	$-6|J_1| + 3|J_2|$	$-3|J_2|$	-1	$-\frac{1}{2}$
	fcc (1, 2) $J_1 < 0$	12	6	$-2|J_1| + |J_2|$	$-3|J_2|$	$-\frac{1}{2}$	$-\frac{1}{4}$
	Diamond (1, 2)	4	12	$-2|J_1| + 6|J_2|$	$-2|J_2|$	$-\frac{1}{4}$	$-\frac{3}{4}$
	Triangular	4	2	$-2|J_1| + |J_2|$	$-|J_2|$	-1	$-\frac{1}{2}$
B	Union jack	4	2^a	$-2|J_1| + |J_2|$	$-|J_2|$	-1	$-\frac{1}{2}$
	fcc	8	4	$-4|J_1| + 2|J_2|$	$-2|J_2|$	-1	$-\frac{1}{2}$

[a]Effective q_2 is quoted.

J_2, and that in the four extraplanar directions to be J_1. Also the union-jack lattice may be constructed out of a square lattice by introducing nnn bonds along alternate diagonals. And the linear chain may be augmented by all nnn bonds lc (1, 2), or with only alternate nnn bonds, lca (1, 2). When the nn lattice is loose packed, we need not take special account of the sign of J_1, but the cases of the close-packed fcc (1, 2) with $J_1 \gtrless 0$ must be considered separately.

The mean-field approximation gives

$$T_c(\rho)/T_c(0) = 1 + \rho$$

so that $T_c = 0$ when $\rho = \rho_c = -1$. However, the mean-field method does not take lattice structure into account properly, and so cannot be expected to give correct results for the Ising model. We now show that $\rho_c = -\frac{1}{2}$ quite generally for *loose-packed* nn lattices with *loose-packed* nnn sublattices, so that both q_1 and q_2 are even. The ground-state energy per spin when $|J_1| \gg |J_2|$ is, independent of the sign of J_1,

$$U^{(1)}(0) = \tfrac{1}{2}(-q_1|J_1| + q_2|J_2|). \qquad (2)$$

Next, since each nnn sublattice may be subdivided into two equivalent sublattices (by meaning of loose packed), a twofold degenerate ordered antiferromagnetic ground state is obtained on each nnn sublattice when $|J_2| \gg |J_1|$. The net energy contribution from nn interactions is then *zero*, since the q_1 nn spins to a given central spin lie in pairs on adjacent sites of the other nnn sublattice, which has antiferromagnetic order. So the ground-state energy per spin when $|J_2| \gg |J_1|$ is, independent of the sign of J_1,

$$U^{(2)}(0) = -\tfrac{1}{2}q_2|J_2|. \qquad (3)$$

Equating (2) and (3) gives $\rho_c = -\frac{1}{2}$. The fcc (1, 2) may be dealt with similarly when $J_1 > 0$, but requires special consideration when $J_1 < 0$. Lattices with close-packed nnn sublattices, such as the sc and diamond which have two fcc nnn sublattices, must be treated separately. For comparison with our exact results, we quote the "bounds" for r_c estimated by Dalton and Wood[3]: sc, $-0.4 < r_c < -0.2$; bcc, $-0.8 < r_c < -0.6$; fcc, $-1.0 < r_c < -0.7$.

Our implicit assumption that only two types of ground-state configuration occur, and that a unique ρ_c is determined by equating their ground-state energies needs justification. A rigorous proof seems difficult, but we are able to provide a heuristic argument. The energy of any specific arrangement of spins on a lattice is a linear function of the interaction J_1 and J_2, and is given by (2) and (3) for the two types of configuration which yield a ground state for an appropriate range of values of J_1 and

J_2. Let ρ_c be determined by the intersection of (2) and (3). We have to demonstrate that no linear combination of J_1 and J_2 corresponding to the energy of a spin configuration can lie below the energies in (2) and (3). It is sufficient to consider the energies of spin configurations at $\rho = \rho_c$ for a fixed value of $|J_1|$. The effect of fixing $|J_1|$ is to normalize the energy. Clearly, the ground-state energies are given correctly by (2) when $|J_1| \gg |J_2|$ and by (3) when $|J_2| \gg |J_1|$. (2) and (3) continue to be correct right up to ρ_c unless a linear combination of J_1 and J_2 corresponding to the energy of some other spin configuration lies below the energy value $U^{(3)} = -\tfrac{1}{4}q_1|J_1|$ appropriate to (2) and (3) at ρ_c. A proof that $U^{(3)}$ is indeed the ground-state energy at ρ_c has eluded us. The ground-state spin arrangements when $|J_1| \gg |J_2|$ and $|J_2| \gg |J_1|$ do exhibit simple periodicity compatible with the lattice structure, and have energy $U^{(3)}$ at ρ_c. But they are not necessarily the only spin arrangements with energy $U^{(3)}$ at ρ_c. It is our belief that other less simply periodic spin arrangements have energies greater than or equal to $U^{(3)}$ at ρ_c.

Let us next summarize the qualitative dependence of T_c on ρ for exactly soluble two-dimensional models. (Assume $J_1 > 0$ for simplicity in describing the phases.) Figure 1(a) shows this dependence for the *union-jack* lattice.[4,9] While $\rho_c < \rho < 0$, T_c is a monotonic increasing function of ρ, with $T_c(\rho) = 0$ at $\rho_c = -\frac{1}{2}$. At temperatures below T_c there is ferromagnetic long-range order. In a range of temperatures above T_c there is ferromagnetic short-range order up to a temperature T_D, the disorder point[9] above which the short-range order is oscillatory. The nnn axis pair correlations are antiferromagnetic above T_D. If $-|J_1| < J_2 < -0.907 \times |J_1|$ there is an intervening antiferromagnetic ordered phase, with upper and lower critical points T_c^*. The ordering of this antiferromagnetic phase, which extends below T_c^* for $-\infty < \rho < \rho_c$, is with respect to the nnn lattice, which is itself a *two*-dimensional quadratic lattice. The dependence of T_c on ρ for the *triangular* lattice,[10,11] regarded as a quadratic lattice with a single nnn interaction, is displayed in Fig. 1(b). There is no antiferromagnetic ordered phase ($T_c^* = 0$) for $-\infty < \rho < \rho_c = -\frac{1}{2}$. This may be attributed to the fact that the nnn lattice is a set of disconnected *one*-dimensional chains, which do not exhibit long-range order. The short-range order above T_D is oscillatory with *temperature-dependent* wavelength.[9,11] [The situation for the completely anisotropic triangular lattice is more complex. Different sets of bonds may be regarded as forming the effective nn lattice above and below ρ_c. The dependence of T_c and T_c^* on ρ is similar to

that in Fig. 1(c).]

On the basis of these results for the two-dimensional lattices, and the existence of a critical ratio ρ_c, we suggest that the qualitative features of the phase diagram for three-dimensional lattices will be as follows when $J_1 > 0$:

(i) There is a critical point $T_c(\rho)$ when $\rho_c < \rho < \infty$, below which there is ferromagnetic long-range order ($\rho_c < 0$). $T_c(0)$ is that of the nn lattice, and $T_c(\infty)$ is that of the nnn (sub) lattice. $T_c(\rho_c) = 0$ and $T_c(\rho)$ increases with ρ at least up to $J_2 = J_1 > 0$.

(ii) When $\rho > 0$ there is ferromagnetic short-range order above T_c.

(iii) When $\rho_c < \rho < 0$, there is ferromagnetic short-range order between T_c and the disorder point T_D.

(iv) Above T_D the short-range order is oscillatory, at least along the nnn axes. [The wavelength of oscillation may be dependent on temperature, as for the triangular lattice (T_D point of first kind), or independent of temperature, as for the union-jack lattice (T_D point of second kind).]

(v) There *may* be an intervening antiferromagnetic long-range order phase between upper and lower critical temperatures T_c^* [Fig. 1(a).] Otherwise the phase diagram is similar to Figs. 1(b) or 1(c).

(vi) When $\rho < \rho_c$, there is a critical point T_c^* (ρ) below which there is antiferromagnetic long-range order on the nnn (sub)lattices, provided these are two or three dimensional. $T_c(-\infty)$ is that of the nnn (sub)lattice alone. [Neighboring spins on different (sub)lattices are uncorrelated at $T = 0$ when both nn and nnn lattices are loose packed, and generally are (weakly) correlated by the nn interaction J_1 at finite temperatures.]

(vii) There is antiferromagnetic short-range order above T_c^* on the nnn lattices, whose nature is probably related to the type of short-range order above T_D when $\rho_c < \rho < 0$. [If T_D is of the second kind, we expect that the antiferromagnetic short-range order above T_D, for $\rho_c < \rho < 0$, continues above T_c^* for $\rho < \rho_c$. Whereas, if T_D is of the first kind, so the short-range order above T_c for $\rho_c < \rho < 0$ has temperature dependent wavelength, then there may be a disorder point T_D^* which terminates the antiferromagnetic short-range order phase above T_c^*. Allowance may be made for the intervening ordered phase in (v) if necessary.]

The case $J_1 < 0$ may be dealt with similarly, provided the nn lattice is loose packed.

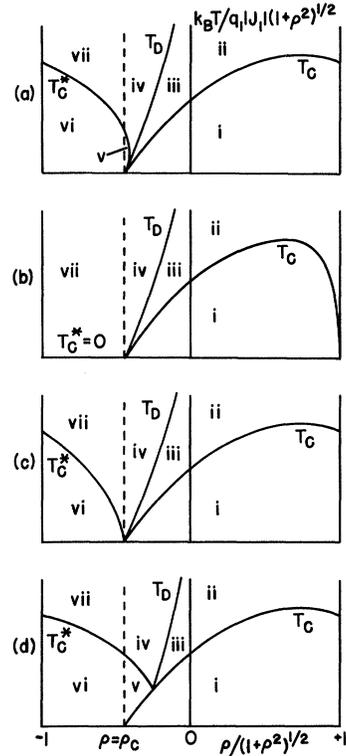To provide discussion on these suggestions, we take as an example the antiferromagnetic fcc

lattice,[12,13] which may be assembled from quadratic layers, so that the spins of one layer are located over the squares of adjacent layers. The bcc lattice constructed from all interlayer bonds will be taken as the nn lattice with interaction $J_1 < 0$. The quadratic layers form the nnn lattices with interaction $J_2 < 0$. Then $q_1 = 8$, $q_2 = 4$, and $\rho_c = -\frac{1}{2}$. When $\rho_c < \rho < \infty$, bcc antiferromagnetic ordering is predominant at sufficiently low temperatures, and we expect there to be a critical point $T_c(\rho)$. When $\rho < \rho_c$, the ordering is that of the antiferromagnetic quadratic layers (i. e. two dimensional), so we expect T_c^* (ρ) to exist. In fact, estimates of the critical point T_c^* ($-\frac{1}{2}$) on the basis of *quadratic layer ordering* have been made for the "isotropic" fcc by Danielian,[12] by Betts and Elliott,[13] and more recently by Wortis,[14] who obtains $k_B T_c^* / |J|$ = 0.61 ± 0.06. This suggests that the fcc behaves as in Fig. 1(a), though some other possibility, such as Fig. 1(d) cannot be ruled out.

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Possibility of Multipolar Ordering in the Exchange-Interaction Model of Ferromagnetism

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It is shown that for the spin-S exchange-interaction model of ferromagnetism the $2S$-"independent" multipolar ($4S$-polar) phase transitions are in fact exactly degenerate with the usual dipolar transition.

The Heisenberg Hamiltonian linear in $\vec{S}_f \cdot \vec{S}_g$ which forms the point of departure for most modern theories of magnetism is in fact only the lowest-order significant term in a perturbation expansion[1] which when carried further leads to terms nonlinear in $\vec{S}_f \cdot \vec{S}_g$. Because of the presence of these (small?[2]) nonlinear terms in $\vec{S}_f \cdot \vec{S}_g$ for systems of spin $S > \frac{1}{2}$, there exists the possibility of phase transitions associated with the various multipole moments of the system[3,4] in addition to the one usually associated with its dipole moment. There is as yet no rigorous statistical mechanical calculation which goes very far in taking such terms into account.[5] Recently, however, the Schrödinger exchange operator[6] which is an essentially nonlinear operator in $\vec{S}_f \cdot \vec{S}_g$ (for $S > \frac{1}{2}$) has been used to form an exchange-interaction model of ferromagnetism.[7,8] The coefficients of the terms in $(\vec{S}_f \cdot \vec{S}_g)^n$ ($1 \le n \le 2S$) are chosen such that this operator permutes the spin coordinates of atoms labeled f and g. For the case $S = 1$, the coefficients of the terms linear and quadratic in $\vec{S}_f \cdot \vec{S}_g$ have equal magnitude. A large number of terms in the high-temperature expansion of various thermodynamic quantities have been obtained by the use of group-theoretic techniques.[9] While these combinations of nonlinear terms may not be realized in nature, considerable insight into the effect of nonlinear terms in the Hamiltonian on critical prop-

erties has been obtained.[10] For this model, the critical index γ which characterizes the divergence of the (dipolar) susceptibility is quite different from that of any other model (for $S > \frac{1}{2}$). Furthermore, it appears that experimental[11] values of γ are bracketed from below by those of the exchange model[10] and from above by those predicted for the Heisenberg model.[12] The purpose of the present paper is to show that for this model all of the $2S$-independent multipolar ($4S$-polar) transitions are in fact *exactly* degenerate with the dipolar transition. This means that considerable care must be exercised in the interpretation of proposed experimental attempts to observe such transitions. For the case $S = 1$, a null result in attempts to resolve dipolar and quadripolar ordering effects could equally well be used as evidence for the fact that the ratio of biquadratic to bilinear terms in the Hamiltonian is very small, or quite close to unity.

The Hamiltonian for the exchange-interaction model of ferromagnetism is

$$\mathcal{H} = -J\vec{\mathcal{P}} - mH\vec{Q} = \mathcal{H}_0 - mH\vec{Q} , \qquad (1)$$

where

$$\vec{\mathcal{P}} = \sum_{\langle f,g \rangle} \vec{P}_{fg}, \quad \vec{Q} = \sum_{f=1}^{N} \vec{S}_{zf} , \qquad (2)$$

and J is the magnitude of a nearest-neighbor exchange interaction, $\vec{P}_{fg}$ is the Schrödinger exchange