planes; Dr. Yakov Eckstein, Argonne National Laboratory, for general discussions, correspondence, and specific advice to make certain that the senses of the axes we used are the same as his; Arthur Ballato, USAEL, for general discussions; and Dr. D. I. Bolef, Washington University, and Dr. H. B. Huntington, Rensselaer Polytechnic Institute, for general comments.

We also owe thanks to Dr. L. J. Teutonico, Republic Aviation Corporation, and Dr. P. C. Waterman, AVCO Research Laboratory, for discussions on pure-mode and energy-flux directions in trigonal crystals; and Dr. Rudolf Bechmann and Dr. Erich Hafner, USAEL, for a critical review of the final manuscript, for stimulating and encouraging discussions on elastic-wave propagation.

Support of USAEL'S Institute for Exploratory Research and Solid State and Frequency Control Division, in the persons of Dr. H. H. Kedesdy, I. N. Greenberg, and F. A. Brand is indicated.

PHYSICAL REVIEW VOLUME 138, NUMBER 3A 3 MAY 1965

Critical Properties of the Heisenberg Ferromagnet with Higher Neighbor Interactions ($S=\frac{1}{2}$)

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(Received 21 September 1964; revised manuscript received 28 December 1964)

The method of exact power-series expansions has been extended to include both nearest-neighbor and next-nearest-neighbor interactions in the Heisenberg model. The series expansions for the susceptibility in zero magnetic field and the free energy in zero magnetic field have been derived to the fifth power in reciprocal temperature for the simple cubic, body-centered cubic, and face-centered cubic lattices. For the special case when all interactions are equal (equivalent-neighbor model), an additional term has been obtained in these expansions. For purposes of discussing the susceptibility and magnetic specific heat, the series expansions have been derived for lattices in which third-neighbor interactions are included, but only for the equivalent-neighbor model. Estimates of critical points are given, and the Padé-approximant method is used to study the dependence of the critical properties (temperature, energy, and entropy) on the relative strength of the first- and second-neighbor interactions. It is found that the variation in the critical point is well represented by

$$T_c(\alpha)=T_c(0)[1+m_1\alpha],$$

where $\alpha=J_2/J_1$ and lies in the range $0\leqslant\alpha\leqslant 1$, and $T_c(0)$ is the critical temperature of the nearest-neighbor model. The values of m_1 are 0.76, 0.99, and 2.74 for the fcc, bcc, and sc lattices respectively. Both the second-neighbor model and the equivalent-neighbor model are used to investigate the behavior of χ_0 for values of T near T_c. It is found that all the coefficients in the magnetic-specific-heat series expansion are positive for the equivalent-neighbor model, and that for lattices with large coordination numbers, reliable estimates of the critical point may be obtained using this function.

I. INTRODUCTION

MUCH previous work has been done on the critical behavior of the Heisenberg model of a ferromagnet when it is assumed that exchange interactions ($-2J\mathbf{S}_i\cdot\mathbf{S}_j$) exist only between nearest-neighbor spins on the lattice. The most powerful theoretical approach towards obtaining estimates of critical constants is that introduced by Kramers and Opechowski.[1] In this method exact series expansions in ascending powers of reciprocal temperature are derived for the partition function and related thermodynamic functions for various lattice structures. In recent years much work has been done in extending the series expansions for the zero field susceptibility χ_0 and magnetic specific heat C_v to a high degree of approximation.[2] For the case where the spin variable S may take an arbitrary value the most extensive calculations have been performed by Rushbrooke and Wood.[3] These authors obtained the first six coefficients in the susceptibility series, and the first five coefficients in the magnetic specific-heat series. Recently a more powerful method of deriving these coefficients has been developed by

[1] H. A. Kramers, Commun. Kamerlingh Onnes Lab. Leiden, Suppl. No. 83. W. Opechowski, Physica **4**, 181 (1937); **6**, 1112 (1939).

[2] V. Zehler, Z. Naturforsch. **A5**, 344 (1950). H. A. Brown and J. M. Luttinger, Phys. Rev. **100**, 685 (1955). M. F. Sykes, thesis, Oxford, 1956 (unpublished). C. Domb and M. F. Sykes, Proc. Phys. Soc. (London) **B69**, 486 (1956).

[3] G. S. Rushbrooke and P. J. Wood, Proc. Phys. Soc. (London) **A68**, 1161 (1955); Mol. Phys. **1**, 257 (1958).

Domb and Wood[4] for the case $S=\frac{1}{2}$. This new method has been used to derive the series expansion of thermodynamic properties for loose-packed lattices to a higher degree of approximation. For these lattices the first eight terms in the susceptibility and the first seven terms in the magnetic specific heat have been derived.

Two ferromagnetic salts for which the isotropic Heisenberg model with a value of $S=\frac{1}{2}$ should be appropriate are $CuK_2Cl_4 \cdot 2H_2O$ and $Cu(NH_4)_2Cl_4 \cdot 2H_2O$. Experimental measurements of the critical constants of these two salts have recently been made by Miedema, van Kempen, and Huiskamp.[5] The bcc lattice is a good approximation to the structure of these two salts. The theoretical estimates of the critical constants for this lattice have been compared with these experimental values by Domb, Sykes, and Wood.[6] The results show that the experimental value of (kT_c/qJ) is 20% higher than the theoretical estimate, and both $(E_\infty - E_c)/kT_c$ and $(S_\infty - S_c)/k$ have experimental values 20% lower then the theoretical values.

It is likely that in these salts more-distant-neighbor interactions are present and are too large to be ignored in the theory. The above discrepancy between experiment and theory would certainly be taken into account by including second nearest-neighbor exchange interactions in the theoretical approach.

In many magnetic compounds it seems likely that the exchange interactions between second- and third-nearest-neighbor spins in the lattice are too large to be neglected. Low-temperature measurements of the magnetization in zero field and the specific heat of EuS have been compared with spin-wave calculations by Charap and Boyd.[7] These authors obtained reasonable agreement between the theoretical and experimental curves for both these properties using values of the nearest-neighbor exchange intergral J_1 and the second-nearest-neighbor exchange integral J_2 given by $J_1/k = 0.2 \pm 0.01$, $J_2/k = -0.08 \pm 0.02$. Wojtowicz[8] has extended the diagrammatic techniques of Rushbrooke and Wood and obtained the high-temperature series expansion for the magnetic specific heat to the fifth power in reciprocal temperature for the Heisenberg model where first- and second-neighbor interactions are present. By evaluating the truncated series expansion for the specific heat over a temperature range $0.44 < (T_c/T) < 0.94$, Wojtowicz has attempted to fit the high-temperature specific-heat measurements of EuS made by Moruzzi and Teaney,[9] using the values of J_1 and J_2 determined by Charap and Boyd. With such a small number of terms in the specific-heat series the error in truncating the series will increase rapidly for values of $(T_c/T) > 0.7$. It would be interesting to see if a closer fit could be obtained by calculating a sequence of Padé approximants to the speciffic heat series.

Wojtowicz and Joseph[10] have derived the high-temperature susceptibility series to the fourth power in reciprocal temperature for the Heisenberg model in which first- and second-neighbor interactions are present, and for an arbitrary spin value. Using experimental measurements of the high-temperature susceptibility of gadolinium these authors attempted to find unique values of J_1 and J_2 which would give a best fit over a temperature range $0.39 < T_c/T < 0.78$, by evaluating the truncated susceptibility series for various values of J_1 and J_2. They reported that all values of J_1 and J_2 satisfying

$$J_1/k + 0.621 J_2/k - 0.030 (J_2/k)^2 = 2.801$$
$$2.22 \leqslant J_1/k \leqslant 3.12$$
$$-0.5 \leqslant J_2/k \leqslant 1$$

gave equally good fits. It would be particularly interesting in this case to evaluate the sequence of Padé approximants to the susceptibility and to similarly compare these functions with the experimental data.

In this paper we discuss the effect of more-distant-neighbor interactions on the critical and thermodynamic properties of the Heisenberg model with a value of $S=\frac{1}{2}$. Exact power series expansions for thermodynamic properties have been derived using the cumulant method of Domb and Wood,[4] which is briefly described in Sec. II. All critical and thermodynamic properties that are discussed fall under two headings. The first of these will be referred to as the second-neighbor model. In this model, exchange interactions $-J_1 \mathbf{S}_i \cdot \mathbf{S}_j$ and $-J_2 \mathbf{S}_k \cdot \mathbf{S}_l$ are present between first- and second-neighbor spins, respectively, and J_2/J_1 is in the range $0 \leqslant J_2/J_1 \leqslant 1$. The second case will be referred to as the equivalent-neighbor model. In this model, exchange interactions are present between first-, second-, and third-nearest neighbor spins and are all equal. For the second-neighbor model the free energy F in zero magnetic field and the susceptibility in zero magnetic field have been expanded to the fifth power in reciprocal temperature. For the equivalent neighbor model six terms in these expansions have been derived.

It is generally found that the coefficients in the series expansions increase in smoothness as the coordination number q of the lattice increases, and as the magnitude of the second-neighbor interactions increases. To obtain information about the critical and high-temperature $(T > T_c)$ thermodynamic properties from the truncated series expansions two extrapolation procedures have been used. The Padé-approximant method has been

[4] C. Domb and D. W. Wood, Phys. Letters **8**, 20 (1964). C. Domb, *Advances in Physics* (Francis & Taylor, London, 1960), Vol. 9 pp. 149-245. C. Domb and D. W. Wood (to be published).

[5] A. R. Miedema, H. van Kempen and W. J. Huiskamp. Physica **29**, 1266 (1964).

[6] C. Domb and A. R. Miedema, *Progress in Low Temperature Physics* (North-Holland Publishing Company, Amsterdam, 1964), Vol. 4, p. 296.

[7] S. H. Charap and E. L. Boyd, Phys. Rev. **133**, A811 (1964).

[8] P. J. Wojtowicz, J. Appl. Phys. **35**, 991 (1964).

[9] V. L. Moruzzi and D. T. Teaney, Solid State Commun. **1**, 127 (1963).

[10] P. J. Wojtowicz and R. I. Joseph, Phys. Rev. **135**, A1314 (1964).

used to investigate all the properties that are discussed, and where the coefficients in the series expansions show a smooth behavior the ratios of successive terms in the expansion have been studied; this is referred to as the ratio method.[11] In Sec. III both these methods have been used to estimate the critical temperatures kT_c/J of the equivalent-neighbor model three-dimensional lattices. The estimates of KT_c/J obtained from the two methods are in very close agreement. Some consideration is also given to the two-dimensional equivalent-neighbor model lattices. Including more-distant-neighbor interactions in the equivalent-neighbor model is equivalent to increasing the coordination number of the lattice. The fcc lattice with first-, second-, and third-equivalent-neighbor interactions has a coordination number $q=42$. By examining the critical temperatures of three-dimensional lattices for the nearest-neighbor model, the second-neighbor model when $J_2/J_1=1$, and the equivalent-neighbor model it is found that for large values of $q(q>12)$ the critical temperature is accurately given by the relation

$$J/kT_c=(2/q)[1+(6/q)].$$

In Sec. IV the variation of the critical temperature with a gradual increase in the second-neighbor exchange interactions over a range $0\leqslant J_2/J_1\leqslant 1$ is discussed. Both Padé-approximant and ratio methods again give results which are in very close agreement. In recent papers Domb and Sykes,[12] Baker,[13] and Gammel[14] *et al.* have examined the behavior of the ferromagnetic susceptibility of the nearest-neighbor Heisenberg model for values of T near T_c. These authors found that the susceptibility behaves as

$$\chi_0(T)\sim A/[1-T_c/T]^\gamma.$$

The value of γ has been estimated by these authors using Padé-approximant methods and ratio methods. In Sec. V we have examined this asymptotic behavior for the second-neighbor model and the equivalent neighbor model three-dimensional lattices. In Sec. VI the effect of more-distant-neighbor interactions on the inverse susceptibility curves above the critical temperature is examined using the Padé-approximant method. The inverse susceptibility is found to be very insensitive to the presence of second-neighbor exchange interactions. This insensitivity probably explains why Wojtowicz and Joseph[10] were unable to obtain unique values of J_1 and J_2 in comparing their results with experimental data.

The critical energy $(E_\infty-E_c)/kT_c$ and the critical entropy $(S_\infty-S_c)/k$ provide information about the nature of the specific-heat curve above the Curie temperature. These two critical constants are suitable for comparison with experimental work. The thermal properties of a variety of ferromagnetic materials have been compared with the theoretical estimates of both the Heisenberg and Ising models with nearest-neighbor interactions by Domb and Miedema.[6] The thermal properties of the second-neighbor model and the equivalent-neighbor model are discussed in Sec. VII.

The high-temperature $(T>T_c)$ magnetic specific heat of the equivalent-neighbor-model lattices has been examined using Padé-approximant methods in Sec. VIII. For the nearest-neighbor-model three-dimensional lattices $(S=\frac{1}{2})$, the series expansion of the specific heat is very erratic and consists of positive and negative terms. For the equivalent-neighbor-model lattices, the coefficients in this expansion are all positive and much smoother. The presence of a singularity in this function is very clearly indicated by the Padé approximants.

For all the thermodynamic functions discussed in this paper we have derived the high-temperature series expansions for an arbitrary spin value S. We are also examining the critical behavior of the general-spin Heisenberg model where more distant-neighbor interactions are present. These results are being prepared for publication.

II. HIGH-TEMPERATURE SERIES EXPANSIONS

For the Heisenberg model where $S=\frac{1}{2}$ the Hamiltonian may be put in the form

$$\mathcal{H}=-\tfrac{1}{2}\sum_{i=1}^{N}\sum_{j=1}^{N}J(i,j)\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_j-mH\sum_{i=1}^{N}\boldsymbol{\sigma}_{zi}. \quad (1)$$

Equation (1) relates to a system of N spins each of which occupies a site of a regular lattice structure, where the sites are labeled $i=1, 2, 3, \cdots N$. The Pauli spin operator $\boldsymbol{\sigma}_i$ corresponding to the spin on the ith lattice site has a component $\boldsymbol{\sigma}_{zi}$ in the direction of the external magnetic field H. The energy of each spin in the magnetic field is $-mH\boldsymbol{\sigma}_{zi}$ where m is the magnetic moment on each lattice site. The exchange energy between the ith and jth sites is $-\frac{1}{2}J(i,j)\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_j$, where $J(i,j)$ is the exchange integral between sites i and j.

The partition function for the assembly is

$$Z_N(\beta,H)=\langle\exp(-\beta\mathcal{H})\rangle, \quad (2)$$

where $\beta=1/kT$ and $\langle\ \rangle$ denotes the trace. A high-temperature series expansion in ascending powers of β is obtained for the partition function by expanding the exponential in (2). The series expansion is then in the form

$$Z_N(\beta H)=1+\beta\langle\mathcal{H}\rangle+\beta^2\langle\mathcal{H}^2\rangle/2!+\cdots\beta^r\langle\mathcal{H}^r\rangle/r!+\cdots. \quad (3)$$

For the case where only nearest-neighbor exchange interactions are considered, $Z_N(\beta H)$ may be expanded

[11] G. A. Baker, Jr., Phys. Rev. **124**, 768 (1961); **129**, 99 (1963); J. W. Essam and M. E. Fisher, J. Chem. Phys. **38**, 802 (1963); M. F. Sykes and M. E. Fisher, Physica **28**, 919, 939 (1962); C. Domb and M. F. Sykes, J. Math. Phys. **2**, 63 (1961).

[12] C. Domb and M. F. Sykes, Phys. Rev. **128**, 168 (1962).

[13] G. A. Baker, Jr., Phys. Rev. **129**, 99 (1963).

[14] J. Gammel, W. Marshall and L. Morgan, Proc. Roy. Soc. (London) **A275**, 257 (1963).

in ascending powers of K, where $K=\beta J$ and J is the nearest-neighbor exchange integral. For the second-neighbor model, $J(i,j)=J_1$ between nearest-neighbor spins (n.n.) and J_2 between second-nearest-neighbor spins (n.n.n.). For the equivalent neighbor model, $J(i,j)=J$ between first-, second-, and third-nearest-neighbor spins.

The Hamiltonian for the second-neighbor model is of the form

$$\mathcal{H}=-\tfrac{1}{2}J_1\Big[\sum_{\text{n.n}}\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_j+\alpha\sum_{\text{n.n.n.}}\boldsymbol{\sigma}_k\cdot\boldsymbol{\sigma}_m\Big]-mH\sum_{i=1}^{N}\boldsymbol{\sigma}_{zi}, \quad (4)$$

where $\alpha=J_2/J_1$. For practical purposes of deriving the coefficients in the series expansions of thermodynamic functions, it is simpler to obtain a series expansion for $\ln Z_N(\beta,H)$. This may be expanded in the form[4]

$$\ln Z_N(\beta,H)=\sum_{(l,j)}P_l{}^j\phi_l{}^j(\beta,H). \quad (5)$$

The diagrammatic interpretation of (5) is as follows. The symbol (l,j) denotes a connected linear graph of l lines, and j serves to distinguish topologically distinct graphs with the same number of lines. The summation in (5) runs over all connected linear graphs. The total number of independent ways the graph (l,j) can exist on a particular lattice is denoted by $P_l{}^j$. The functions $\phi_l{}^j(\beta,H)$ are power series in ascending powers of β and are unique for each graph (l,j). The form of the expansion (5) applies to any system of N spins. By applying this form of the expansion to the graphs themselves the functions $\phi_i{}^j(\beta,H)$ can readily be obtained as functions of the partition functions of finite clusters of spins.

As an example of this procedure, consider the initial terms of (5) for the infinite lattice. The interactions between nearest-neighbor spins i and j are represented graphically by $\underset{i}{\bullet}\!-\!\underset{j}{\bullet}$, and between second-neighbor spins k and m by $\underset{k}{\bullet}\!-\!\|\!-\!\underset{m}{\bullet}$. The graphs appearing in Fig. 1 are the first five terms in (5) which now becomes

$$\ln Z_N(\beta,H)=P_1{}^0\phi_1{}^0+P_1{}^1\phi_1{}^1+P_2{}^0\phi_2{}^0+P_2{}^1\phi_2{}^1+P_2{}^2\phi_2{}^2+\cdots. \quad (5a)$$

The series expansions $\phi_l{}^j(\beta,H)$ are in the form

$$\phi_l{}^j(\beta,H)=\sum_{r=l}^{\infty}\psi_r\beta^r \quad (6)$$

and these may be obtained by applying (5) to the diagrams themselves, which are now considered as finite clusters of spins. If $f_l{}^j$ denotes $\ln Z_N(\beta,H)$ for the finite clusters we obtain

$$\begin{aligned}f_1{}^0&=\phi_1{}^0,\\ f_1{}^1&=\phi_1{}^1,\\ f_2{}^0&=2\phi_1{}^0+\phi_2{}^0,\\ f_2{}^1&=\phi_1{}^0+\phi_1{}^1+\phi_2{}^1,\\ f_2{}^2&=2\phi_1{}^1+\phi_2{}^2.\end{aligned} \quad (7)$$

From Eqs. (7) all the $\phi_l{}^j(\beta,H)$ in (5a) may be found, and from (6) $\ln Z_N(\beta,H)$ for the infinite lattice may be arranged as a power-series expansion in β such that the coefficients of β^r involve only graphs of l lines where $l\leqslant r$.

The thermodynamic functions we shall discuss in the following sections are related to two series expansions readily obtainable from (5) when expanded in powers of β. These are the expansions of the free energy F in zero magnetic field, and the susceptibility per spin χ_0 in zero magnetic field. For the second-neighbor model these expansions are in the form

$$\frac{-F}{kT}=N\ln 2+N\sum_{r=2}^{\infty}\frac{e_r(\alpha)K^r}{2^r r!}\cdots \quad (8)$$

and

$$\frac{kT}{m^2}\chi_0=1+\sum_{r=1}^{\infty}\frac{j_r(\alpha)K^r}{2^r r!}\cdots, \quad (9)$$

where $K=J_1/kT$, and the coefficients $j_r(\alpha)$ and $e_r(\alpha)$ are polynomials of degree r in α. We have derived the polynomials in (8) and (9) for values of r up to and including $r=5$. The labor involved in calculating these polynomials increases rapidly with r. For the calculation of $j_5(\alpha)$ more than 150 different graphs have to be considered. It is very important to have some reliable check on the series expansions (8) and (9). Our results have been checked in two ways and have been found to agree in both cases. The first checking procedure we adopted was to derive the series expansions (8) and (9) for a cluster of eight spins at the corners of a cube, where first-neighbor interactions were considered along the edges of the cube and second-neighbor interactions along the face diagonals, by using the exact solution of the partition function.[15] Using our calculated values of the $\phi_l{}^j(\beta,H)$ functions, we recalculated the same series expansions for the cube; and agreement was found. As a further check we have derived the series expansions (8) and (9) for an arbitrary spin value S, using the diagrammatic methods of Rushbrooke and Wood. Substituting $S=\frac{1}{2}$ we again obtain agreement.

For the equivalent-neighbor model the coefficients in (8) and (9) (e_r and j_r) are simply numerical coefficients of K^r, where $K=J/kT$. We have derived the first six coefficients for this model in both expansions. The series expansion coefficients for the second-

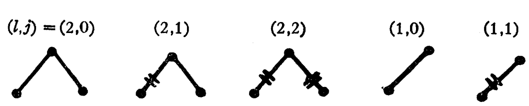

FIG. 1. One and two line diagrams in the expansion (5).

[15] E. M. Corson, *Perturbation Methods in the Quantum Mechanics of n-Electron Systems* (Blackie and Son, Ltd., London, 1951).

neighbor model and the equivalent-neighbor model are collected together in Appendixes I and II.

III. CURIE TEMPERATURES OF THE EQUIVALENT-NEIGHBOR MODEL

For the equivalent-neighbor model six terms in the susceptibility series expansions are available for all two- and three-dimensional lattices, where the expansion is of the form

$$\bar{\chi}_0=(kT/m^2)\chi_0=1+\sum_{r=1}^{\infty}A_rK^r. \quad (10)$$

For this model the following notation is used to refer to a particular lattice. The face-centered-cubic lattice with equivalent first-, second-, and third-neighbor interactions is denoted by fcc (1,2,3) and the body-centered-cubic lattice with first- and second-equivalent-neighbor interactions by bcc (1,2). The three-dimensional lattices that have been considered exhibit a very wide range of lattice structure with values of the coordination number ranging from $q=14$ for bcc (1,2) to $q=42$ for fcc (1,2,3). For these lattices the coefficients in (10) display very smooth behavior and provide a reasonable basis for extrapolations using the ratio methods developed by Domb and Sykes[11,12] for the Ising model susceptibility series. It is natural to try to fit the susceptibility in the region of the critical point ($K\sim K_c$) by an expression of the form

$$\bar{\chi}_0=A/[K-K_c]^{1+g}, \quad (11)$$

where A is a constant and $K_c=J/kT_c$. If the expression (11) is valid the coefficients A_n in (10) will have the asymptotic form

$$A_n\sim Cn^g/K_c^n, \quad (12)$$

where C is a constant. The ratios of successive coefficients in (10) will be of the form

$$\mu_n=\frac{A_n}{A_{n-1}}\sim\frac{1}{K_c}\left[1+\frac{g}{n}\right] \quad (13)$$

and hence a plot of μ_n versus $1/n$ should tend to a straight line as n increases. When the coefficients in the susceptibility are smooth this plot quickly settles down to a straight line whose intersection with $1/n=0$ determines the critical point K_c and whose slope determines g.

In Fig. 2 we have included plots of all the three-dimensional lattices with two and three equivalent-neighbor interactions. To compare the behavior of different lattices we have plotted μ_n/q versus $1/n$. As the coordination number of the lattice increases these plots exhibit a definite curvature. A similar effect was reported by Fisher and Gaunt[16] when considering the hypercubical lattices for the Ising model. Fisher has pointed out that the curvature could be reduced by plotting μ_n/q versus $1/n+k$, where k is a small positive or negative integer or fraction. For the equivalent-neighbor model lattices a slight improvement was obtained by plotting the graphs with $k=1$. Following Domb and Sykes, accurate estimates of the critical points for the lattices in Fig. 2 have been obtained using the following procedure. Estimates of g were obtained from the gradients of the plots in Fig. 2, and the functions

$$\beta_n=n\mu_n/q(n+g) \quad (14)$$

calculated for each lattice. The function β_n tends to $1/qK_c$ as $n\to\infty$. For a particular lattice an estimate of the critical point can be obtained by plotting μ_n/q versus $1/n$ and β_n versus $1/n$ on the same graph; both plots intersect $1/n=0$ at the common limit $1/qK_c$. If the value of g in (14) is a good estimate the plot of β_n versus $1/n$ will approach the limit $1/qK_c$ horizontally. Even if the estimate of g is in error, the limit of β_n is still $1/qK_c$; however, in this case the approach to the limit is not horizontal. Estimates of the critical temperatures of the equivalent-neighbor-model lattices in Fig. 2. obtained in this way are quoted in Table I.

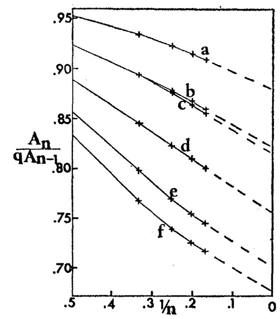

FIG. 2. Ratio plots for the equivalent-neighbor lattices (a) corresponds to the fcc (1,2,3), $q=42$, (b) corresponds to the bcc (1,2,3), $q=26$, (c) corresponds to the sc (1,2,3), $q=26$, (d) corresponds to the fcc (1,2) and sc (1,2), $q=18$, (e) corresponds to the bcc (1,2), $q=14$, (f) corresponds to the fcc (1), $q=12$.

Estimates of K_c for the lattices in Fig. 2 have also been obtained using the Padé-approximant method.[11] The $[M,N]$ Padé approximant to the susceptibility $\chi_0(K)$ is given by $P(K)/Q(K)$, where $P(K)$ is a polynomial $\sum p_rK^r$ of degree M and $Q(K)$ is a polynomial $\sum q_rK^r$ of degree N. The coefficients p_r and q_r in the two polynomials are chosen such that the series expansion of $\bar{\chi}_0(K)$ agrees with the expansion of $P(K)/Q(K)$ for the first $N+M+1$ terms. These coefficients can be found by equating the coefficients in the identity

$$\bar{\chi}_0(K)Q(K)-P(K)\equiv AK^{N+M+1}+BK^{N+M+2}+\cdots, \quad (15)$$

where $q_0=1$. These calculations have been performed on

TABLE I. Estimates of the Curie points K_c $(=J/kT_c)$ of three-dimensional lattices for the equivalent-neighbor model.

Lattice q	fcc (1,2) 18	sc (1,2) 18	bcc (1,2) 14	fcc (1,2,3) 42	sc (1,2,3) 26	bcc (1,2,3) 26
Ratio method	0.1475	0.1477	0.2031	0.0545	0.0948	0.0941
Padé approximant method	0.1475	0.1476	0.2048	0.0545	0.0950	0.0946

[16] M. E. Fisher and D. S. Gaunt, Phys. Rev. 133, A224 (1964).

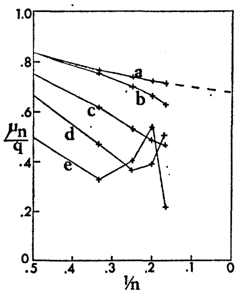

FIG. 3. Ratio plots for which (a) corresponds to the fcc (1), $q=12$, (b) corresponds to the triangular lattice with 2 equivalent neighbors, (c) corresponds to the simple-quadratic lattice with 2 equivalent neighbors, (d) corresponds to the triangular lattice with nearest neighbors only, (e) corresponds to the simple-quadratic lattice with nearest neighbors only.

an electronic digital computer which has been programmed to calculate the sequence of $[M,N]$ Padé approximants to any function such as $\bar{\chi}_0(K)$, which is given as a power series of the form

$$\bar{\chi}_0(K)=\sum_{r=0}^{\infty} A_r K^r.$$

If the reduced susceptibility diverges at the critical point in the form of (11), Baker[11] has suggested that the Padé approximants to the logarithmic derivative of $\bar{\chi}_0$ should first be examined. This has the form

$$\frac{d}{dK}\ln\bar{\chi}_0=-\frac{(1+g)}{K-K_c} \tag{16}$$

near the critical point, which contains a simple pole at K_c and a residue $-(1+g)$. We have examined the $[M,N]$ sequence of approximants to the logarithmic derivative of the susceptibility series of lattices in Fig. 2. Estimates of K_c have been obtained by selecting the first singularity on the positive real axis. These results are compared with the ratio method results in Table I. The two methods give results which are in very close agreement.

We have repeated the above analysis of the susceptibility series for the two-dimensional lattices when two equivalent-neighbor interactions are present ($J_1=J_2$). The ratio plots for the simple quadratic and triangular lattices with nearest-neighbor, and two equivalent-neighbor interactions present are shown in Fig. 3. The ratio plot for the fcc lattice with nearest-neighbor interactions is also shown for comparison. At first sight it appears that the plots *b* and *c* in Fig. 3 are smooth enough to be extrapolated to intersect $1/n=0$ at a finite critical temperature ($T_c>0$). This would imply the existence of ferromagnetism (spontaneous magnetization) for two-dimensional lattices with Heisenberg coupling between the spins. In contradiction to this are the results of spin-wave theory which are that the Heisenberg ferromagnet does not give rise to spontaneous magnetization in one and two dimensions ($T_c=0$). The divergence of the Bloch[17] integral for the magnetization in one and two dimensions indicates that there is no stability attached to the ferromagnetic ordered state.

[17] F. Bloch, Z. Physik **74**, 295 (1932).

This result is supported by an alternative argument put forward by Peierls,[18] who derived the changes in energy and entropy of an ordered spin system produced by introducing domains of reverse orientation. For Heisenberg coupling between spins on a two-dimensional lattice the energy of the domain border can be reduced by increasing the size of the Bloch wall.[18] If at the border there exists a transition layer in which the spins reverse their direction gradually the energy of the reversed domain becomes independent of its size, and no spontaneous magnetization is possible. In the same way Peierls was able to show that the two-dimensional Ising nets have a spontaneous magnetization at low enough temperatures. The essential difference between the Ising and Heisenberg interactions is that for the Ising model the entire change of spin direction must occur across a single atomic spacing.

The results of both the above arguments are still valid when the exchange interactions extend beyond nearest neighbors to any finite range. The ratio plots in Fig. 3 for the nearest-neighbor lattices (d and e) become erratic after two or three terms in the susceptibility series. It is very likely that the equivalent-neighbor-model lattices will show a similar effect when more terms in the series expansions for plots b and c are included. We conclude that the plots b and c in Fig. 3 are not in fact validly extrapolated to obtain nonzero critical points. Evidence for the different behavior of the Ising and Heisenberg models in two dimensions was obtained by examining the Padé approximants to the corresponding susceptibility series for the equivalent-neighbor-model lattices (b and c in Fig. 3). It was found that the approximants to the Ising model series gave consistent estimates of J/kT_c, whereas no clear indication of a singularity on the positive real axis was found for the Heisenberg-model series.

To consider how a gradual increase in the strength of the second-neighbor interactions affects the nature of the ratio plots μ_n versus $1/n$ we have examined the ratio of successive coefficients in the susceptibility series for the second-neighbor model. The ratio J_2/J_1 provides a measure of the relative strengths of the first- and second-neighbor interactions. For this purpose we have selected the simple cubic lattice, which for the case of nearest-neighbor interactions only ($\alpha=0$) gives a very irregular ratio plot. In Fig. 4 the ratio plots μ_n versus $1/n$ are shown for values of α from 0 to 0.25. It is clearly seen that as α increases a relatively small admixture of second-neighbor interactions is sufficient to render the coefficoents A_r quite smooth. It appears generally that at about $\alpha=0.25$ the second-neighbor interaction is sufficiently strong to produce a smooth ratio plot, and that this smooth behavior continues to exist for values of $\alpha>0.25$. Using these ratio plots

[18] R. Peierls, Proc. Cambridge Phil. Soc. **32**, 477 (1936); G. H. Wannier, *Elements of Solid State Theory* (Cambridge University Press, London, 1959).

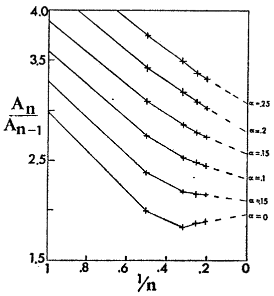

FIG. 4. Ratio plots for the simple-cubic lattice with second-neighbor interactions present.

accurate estimates of K_c for various value of α can be obtained by extrapolating the plots to intersect $1/n=0$.

It is interesting to consider how the Curie point of a lattice varies with q in the limit of large q. For small q the effect of lattice structure will play a dominant role in locating K_c, but as q increases the effects of lattice structure diminish [compare K_c for sc (1,2) and fcc (1,2) for which $q=18$]. The problem has been considered in detail by Sykes and Dalton[19] for both the Ising and Heisenberg models. For the Heisenberg model a good approximation for K_c is obtained by

$$K_c=(2/q)[1+(6/q)] \tag{17a}$$

for values of $q>12$. Reasonable results (within 1%) are obtained for $q=12$ and the accuracy rapidly increases with q. The asymptotic formula (17a) is useful for including even higher neighbor interactions (4, 5, 6 $\cdots$ equivalent-neighbor interactions) in the equivalent-neighbor model. The accuracy of (17a) is supported by calculations using the Green function theory of ferromagnetism as developed by Tahir Kheli.[20] We have used the theory to calculate the Curie temperatures of equivalent-neighbor-model lattices in which up to tenth-neighbor interactions have been included. Extrapolation of these results gives the asymptotic equation

$$qK_c=2+(11.9/q). \tag{17b}$$

IV. VARIATION OF THE CURIE TEMPERATURE WITH THE SECOND-NEIGHBOR INTERACTION

For the second-neighbor model we have investigated the variation of K_c $(=J_1/kT_c)$ with α $(=J_2/J_1)$ for values of α in the range $0\leqslant\alpha\leqslant 1$. To determine the Curie temperatures $K_c(\alpha)$ both Padé-approximant and ratio methods have been used. The sequence of $[M,N]$ Padé approximants to the series expansions of (d/dK) $\ln\bar{\chi}_0$ and $(\bar{\chi}_0)^{3/4}$ have been calculated for 20 values of α in the above range, and the appropriate singularities of each approximant have been selected.[21] For a particular lattice the sequence of estimates of $K_c(\alpha)$ from the sequence of $[M,N]$ Padé approximants to both the above functions was found to be convergent. For each function the convergence of $K_c(\alpha)$ increased rapidly with α, and for a particular value of α both functions gave consistent values of the critical point. The singularities of the [2,3] and [3,2] Padé approximants to $(\bar{\chi}_0)^{3/4}$ found for various values of α, which correspond to the critical temperature $K_c(\alpha)$ are listed in Table II. We expect the values of $K_c(\alpha)$ in Table II to be accurate to within 2% over the whole range of α, and that the error is probably much less than this for $\alpha>0.25$.

TABLE II. Padé approximant estimates of J_1/kT_c for the second-neighbor model for values of α in the range $0\leqslant\alpha\leqslant 1$, using the series for $(\chi_0)^{3/4}$.

$\alpha=J_2/J_1$	fcc lattice [2,3]	fcc lattice [3,2]	sc lattice [2,3]	sc lattice [3,2]	bcc lattice [2,3]	bcc lattice [3,2]
0.0	0.2458	0.2464	0.5686	0.5538	0.3914	0.3914
0.1	0.2289	0.2293	0.4357	0.4336	0.3564	0.3559
0.2	0.2145	0.2148	0.3531	0.3531	0.3279	0.3269
0.3	0.2020	0.2023	0.2979	0.2979	0.3044	0.3027
0.4	0.1912	0.1914	0.2583	0.2583	0.2887	0.2822
0.5	0.1816	0.1818	0.2284	0.2285	0.2621	0.2645
0.6	0.1731	0.1732	0.2051	0.2052	0.2482	0.2492
0.7	0.1655	0.1656	0.1864	0.1864	0.2350	0.2356
0.8	0.1586	0.1587	0.1709	0.1710	0.2231	0.2236
0.9	0.1525	0.1525	0.1580	0.1580	0.2123	0.2129
1.0	0.1468	0.1468	0.1470	0.1470	0.2025	0.2032

For each lattice we have obtained estimates of $K_c(\alpha)$ using the ratio methods described in the previous section. For each lattice and all values of α between 0 and 1 the estimates of $K_c(\alpha)$ from the ratio plots were found to be in very close agreement with the Padé-approximant estimates. This is clearly illustrated in Fig. 5, where kT_c/J_1 is plotted against α over the range $0\leqslant\alpha\leqslant 1$ for the bcc lattice. Estimates of the critical temperature kT_c/J_1 from the ratio plots are slightly and consistently less than the corresponding Padé-approximant estimates.

The variation of the Curie point with the strength of the second-neighbor exchange interactions for the three-dimensional lattices is shown in Fig. 6. In this diagram $T_c(\alpha)/T_c(0)$ has been plotted against J_2/J_1, where $T_c(0)$ is the critical temperature of the lattice

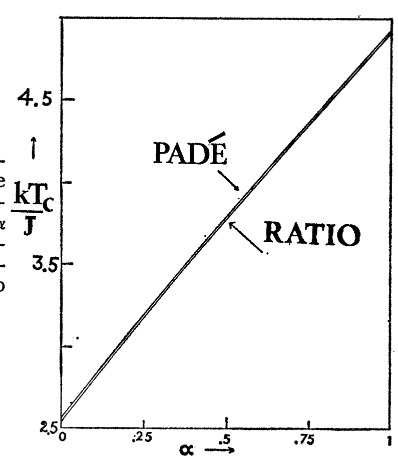

FIG. 5. The variation in the Curie point of the bcc, lattice for values of α in range $0\leqslant\alpha\leqslant 1$ obtained by using Padé-approximant and ratio methods.

[19] M. F. Sykes and N. W. Dalton (to be published).

[20] R. A. Tahir-Kheli and D. ter Haar, Phys. Rev. 127, 88 (1962).

[21] For some function $F(x)$ which has an asymptotic behavior of the form $F(x)\sim(x-x_c)^{-\gamma}$, the function $F(x)^{1/\gamma}$ will have a simple pole at $x=x_c$, which can be readily located by the approximants to $F(x)^{1/\gamma}$.

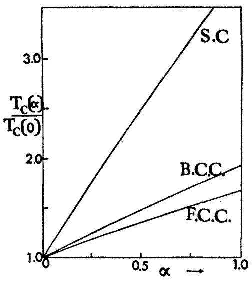

FIG. 6. Variation of the Curie point with the strength of the second-neighbor interactions.

when only nearest-neighbor interactions are present. The most striking feature of the function $T_c(\alpha)/T_c(0)$ is that it appears to have a very nearly linear variation with α. For the three lattices in Fig. 6 the plots appear to have a definite but very slight curvature. This effect is most evident in the case of the simple cubic lattice.

We may use the ratio method to examine the equation of the curve in Fig. 6. The coefficients A_n in (10) for the second-neighbor model are of the form

$$A_n=\frac{j_n(\alpha)}{2^n n!}=\sum_{r=0}^{n} a_{nr}\alpha^r,$$
$$=\frac{j_n(0)}{2^n n!}[1+\sum_{r=1}^{n} b_{nr}\alpha^r], \quad (18)$$

where $j_n(0)/2^n n!$ are the coefficients of K^n in the susceptibility series for the nearest-neighbor model. The ratio of successive terms in the susceptibility series for the second-neighbor model are now given by

$$\mu_n(\alpha)=\mu_n(0)[1+\sum_{r=1}^{\infty}\beta_r(n)\alpha^r], \quad (19)$$

where

$$\mu_n(\alpha)=j_n(\alpha)/2nj_{n-1}(\alpha),$$
$$\mu_n(0)=j_n(0)/2nj_{n-1}(0),$$
$$\beta_1(n)=(b_{n,1}-b_{n-1,1}),$$
$$\beta_2(n)=(b_{n,2}-b_{n-1,2})-b_{n-1}\beta_1(n), \text{ etc.}$$

As $n\to\infty$ the sequence $\mu_n(\alpha)\to kT_c(\alpha)/J_1$, and by virtue of (19) we obtain

$$\frac{T_c(\alpha)}{T_c(0)}=1+\sum_{r=1}^{\infty} m_r\alpha^r, \quad (20)$$

where

$$m_1=\lim_{n\to\infty}\beta_1(n), \quad m_2=\lim_{n\to\infty}\beta_2(n) \text{ etc.}$$

From Fig. 6 it appears that the sequence m_r is a rapidly decreasing one so that only the first few terms in (20) are significant. By examining the first five polynomial coefficients in (9) we find that $\beta_2(n)/\beta_1(n)$ is of the order of 0.05 for $n=5$ and is decreasing with n. At this stage a reasonable estimate of m_1 can be obtained but more polynomials in (9) are required to make a reliable estimate of m_2. To a good approximation the variation of the critical point with the strength of the second-neighbor interactions is given by (20) with

$$m_1=0.76,\ 0.99, \text{ and } 2.74,$$

for the fcc, bcc and sc lattices, respectively, and with all other $m_r=0$. The mean-field theory of the Heisenberg model predicts that $T_c(\alpha)/T_c(0)$ is exactly linear in α. The corresponding values of m_1 from mean-field theory are given by

$$m_1=q_2/q_1, \quad (21)$$

where q_1 and q_2 are the first- and second-nearest-neighbor coordination numbers of the lattice. For the lattices in Fig. 6 the values of m_1 given by (21) are $m_1=0.5$, 0.75, and 2.0 for fcc, bcc, and sc lattices, respectively.

In a recent paper Tahir-Kheli and Jarrett[22] have extended the Green function theory of the Heisenberg model to include exchange interactions between second-nearest neighbors for the bcc and fcc lattices. These authors have examined the behavior of $T_c(\alpha)$ for α in the range $-1\leqslant\alpha\leqslant 2$. The plots of $T_c(\alpha)/T_c(0)$ versus α shown in Fig. 6 exhibit a greater curvature than the corresponding plots predicted by Green function theory. In Table III we have compared the values of m_1 in (20) obtained from series expansions, mean-field theory, and Green-function theory for the sc, bcc, and fcc lattices.[23] The values of m_1 from the series expansion of χ_0 represent of course the initial gradients in Fig. 6. For the bcc and fcc lattices the Green function theory estimates lie between the mean-field theory and series expansion results. Tahir-Kheli and Jarrett have also calculated the Curie temperatures of the fcc and bcc lattices at the points $\alpha=0$ and $\alpha=1$. In Table IV we have compared their estimates of kT_c/J_1 with our estimates based on Padé-approximant and ratio methods.[24] The interesting behavior of $T_c(\alpha)$ occurs when α is in the range $-1<\alpha<0$, and Tahir-Kheli and Jarrett were able to calculate the values of α on the negative axis for which $T_c(\alpha)=0$. We have examined the series expansion for the susceptibility of lattices in Fig. 6 for values of α in the range $-1<\alpha<0$, but

TABLE III. Estimates of m_1 in (20) obtained from mean-field theory, Green-function theory, and series expansions.

Lattice	fcc	sc	bcc
Mean-field theory	0.5	2.00	0.75
Green-function theory	0.68	2.90	0.90
Series expansions	0.76	2.74	0.99

[22] R. A. Tahir-Kheli and H. S. Jarrett, Phys. Rev. **135**, A1096 (1964).

[23] The estimates of m_1 and kT_c/J for the simple cubic lattice using the Green function theory has been calculated by one of us (NWD).

[24] For the nearest-neighbor model ($\alpha=0$) the estimates of kT_c/J for series expansion are based on extrapolation of the susceptibility series with nine terms for the bcc and sc lattices and seven terms for the fcc lattice.

unfortunately the expansions become unreliable for fairly small negative values of α. More terms are needed in (9) to obtain reliable information in this region. The details of these calculations will be presented elsewhere.

V. ASYMPTOTIC BEHAVIOR OF THE SUSCEPTIBILITY

By using Padé-approximant and ratio methods it has been fairly well established by Domb and Sykes,[12] Baker,[13] and Gammel *et al.*,[14] that for the nearest-neighbor Heisenberg model the susceptibility at values of T near T_c behaves asymptotically as

$$\bar{\chi}_0 \sim A/[1-T_c/T]^{\gamma}. \tag{22}$$

Various authors report that the index γ is independent of lattice structure and appears to be close to the value $\gamma=\frac{4}{3}$. All previous work on this question has been confined to only three lattices with values of coordination number $q=6$, 8, and 12. These are the sc(1), bcc(1), and fcc(1) lattices, respectively. In the equivalent-neighbor-model lattices there exists a much wider range of lattice structure to be examined with values of $q=6$, 8, 12, 14, 18, 26, and 42. These are the sc(1), bcc(1), fcc(1), bcc(1,2), sc(1,2) and fcc(1,2), bcc(1,2,3) and sc(1,2,3), and the fcc(1,2,3) lattices, respectively.

TABLE IV. Estimates of kT_c/J_1 for the second-neighbor model at the points $\alpha=0$ and $\alpha=1$ in Fig. 6 obtained from Green-function theory and series expansions.

Lattice	fcc	sc	bcc
Green-function theory			
$\alpha=0$	4.08	1.75	2.60
$\alpha=1$	6.87	6.85	4.94
Series expansion			
$\alpha=0$	4.068	1.700	2.548
$\alpha=1$	6.780	6.770	4.925

The ratio plots for these lattices are shown in Fig. 2. The significant feature of these plots is that as the order of n increases the gradients become nearly equal. If the susceptibility has the form of (22) the gradients of the plots tend to limiting value $(\gamma-1)/qK_c$. A first approximation to K_c is given by $qK_c=2$. The plots for lattices in Fig. 2 suggest that γ is independent of lattice structure. Estimates of the limiting gradients in Fig. 2. give the result that $\gamma=1.31\pm0.03$ for all these lattices.

We have also obtained estimates of γ from the residues of the singularities in the sequence of $[M,N]$ Padé approximants to the logarithmic derivatives of the susceptibility series for all the lattices in Fig. 2. These estimates are given in Table V. We have often observed that a small variation in the position of K_c given by the approximant can produce a large change in the value of the residue. Although the values of γ in Table V show a fairly wide spread, we regard these results as consistent with $\gamma=\frac{4}{3}$ for all the lattices.

TABLE V. Estimates of γ from the residues to the singularities in χ_0 obtained from the $[M,N]$ sequence to $(d/dK)\log(\chi_0)$.

Lattice / q	fcc (1,2) 18	sc (1,2) 18	bcc (1,2) 14	fcc (1,2,3) 42	sc (1,2,3) 26	bcc (1,2,3) 26
[2,2]	1.374	1.380	1.582	1.297	1.339	1.537
[3,2]	1.375	1.379	1.261	1.261	1.351	1.348
[2,3]	1.375	1.382	1.403	1.255	1.348	1.388

The second-neighbor-model series expansions for $\bar{\chi}_0(\alpha,K)$ have also been used to obtain estimates of γ. This has been done by forming a power-series expansion in K_c for γ, which on assuming the above asymptotic form of $\bar{\chi}_0$ is given by

$$\gamma \simeq K_c(1-K/K_c)(d/dK)\ln\bar{\chi}_0. \tag{23}$$

Evaluating this series expansion for γ at $K=K_c$ estimates of γ can be obtained. This has been done by calculating the sequence of $[M,N]$ Padé approximants to the series expansion of γ, and evaluating these functions at the point $K=K_c$. These calculations have been performed for various values of α. Estimates of γ obtained in this way are given in Table VI, where it can be seen that γ is close to 1.33 for each lattice. Similar calculations have been performed for the nearest-neighbor-model and equivalent-neighbor-model susceptibility series. For the nearest-neighbor-model bcc lattice γ was very close to the value $\frac{4}{3}$ and for the equivalent-neighbor model, estimates of γ obtained were much more consistent and nearer to $\frac{4}{3}$ than the estimates in Table V. We conclude that the nature of the transition in the susceptibility is probably independent of lattice structure.

VI. MAGNETIC SUSCEPTIBILITY ABOVE THE CURIE POINT

Much previous work has been done on investigating how the reciprocal susceptibility of the Heisenberg ferromagnet above the Curie point is affected by changes in lattice structure and changes in the spin variable.[3,12] It was reported by Rushbrooke and Wood that a striking feature of the reciprocal susceptibility of a given lattice was its insensitivity to large changes in the spin variable.

TABLE VI. Estimates of γ for the second-neighbor model.

α	sc [1,3]	sc [2,2]	bcc [1,3]	bcc [2,2]	fcc [1,3]	fcc [2,2]
0.	...	1.313	1.333	1.331	1.323	1.330
0.2	1.333	1.333	1.349	1.315	1.326	1.336
0.4	1.332	1.335	1.336	1.414	1.329	1.339
0.6	1.330	1.344	1.336	1.354	1.330	1.337
0.8	1.319	1.342	1.337	1.350	1.328	...
1.0	1.397	1.384	1.315	1.335	1.313	1.332

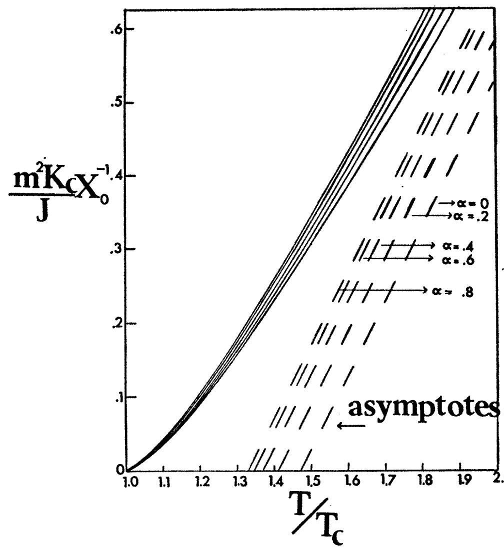

FIG. 7. Inverse susceptibility of the fcc lattice with second-neighbor interactions present.

To investigate the response of the inverse susceptibility to the presence of second-neighbor exchange interactions we may write the series expansion in the form

$$\frac{J\chi_0}{m^2K_c}=t^{-1}\left[1+\sum_{l=1}^{\infty}\frac{P_l t^{-l}}{2^l l!}\right], \tag{24}$$

where $t=K_c/K$ and $P_l=j_l(\alpha)K_c(\alpha)^l$. Expanded in this way the susceptibility is a power series in reduced temperature T/T_c.

The sequence of $[M,N]$ Padé approximants to the series (24) have been evaluated over the range $T=T_c$ to $T=2T_c$ and for various values of α between 0 and 1 for the three-dimensional lattices. By examining the behavior of these Padé approximants we can pick out the trend in the susceptibility curves obtained by including successively higher order terms in (24). The convergence was found to be quite rapid and we expect the results to be accurate to within 2% except in regions within 20% of T_c. The changes in the susceptibility curves above T_c produced by varying α are similar for all three lattices. In Fig. 7 the inverse susceptibility of the fcc lattice is plotted against the reduced temperature for values of $\alpha=0$, 0.2, 0.4, 0.6, and 0.8. For a given value of the reduced temperature the inverse susceptibility increases with α. A plot of $m^2K_c/J\chi_0$ against the reduced temperature tends to a straight line for large values of temperature. The equation of the asymptotes to the curves in Fig. 7 is

$$m^2K_c/J\chi_0=T/T_c-P_1/2; \tag{25}$$

these intercept the temperature axis at the Curie-Weiss points T_w given by

$$T_w=\tfrac{1}{2}(q_1+\alpha q_2)T_c(\alpha)K_c(\alpha), \tag{26}$$

where q_1 and q_2 denote the first- and second-neighbor coordination numbers, respectively. The most striking feature of the curves in Fig. 7 is the insensitivity of the inverse susceptibility to the magnitude of the second-neighbor interaction. We conclude from this that the theoretical susceptibility curves are not very suitable for comparison with experimental data for the purpose of obtaining estimates of J_1 and J_2 for ferromagnetic substances.

Using the same methods we have also examined the behavior of the susceptibility of the equivalent-neighbor-model lattices. For comparison with Fig. 7, in Fig. 8 are the plots of the inverse susceptibility for the bcc(1), fcc(1,2), sc(1,2,3), and fcc(1,2,3) lattices, for which the coordination numbers are 8, 18, 26, and 42, respectively. The inverse susceptibility has been evaluated over the range $T=T_c$ to $T=3T_c$. The inverse susceptibility increases by about 30% at $T=2T_c$ for a change in q from $q=8$ to $q=42$. As the coordination number of the lattice increases the inverse susceptibility becomes increasingly linear.

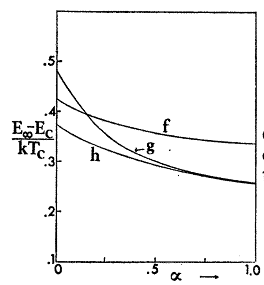

FIG. 9. The variation of the critical energy with the strength of the second-neighbor interaction, (f) bcc lattice (g) sc lattice (h) fcc lattice.

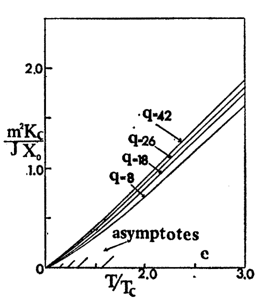

FIG. 8. Variations in the inverse susceptibility with increasing coordination numbers.

VII. CRITICAL ENERGY AND ENTROPY

Domb and Sykes[12] have recently examined the effects of spin on the critical and thermal properties of the fcc lattice with both Ising and Heisenberg nearest-neighbor-exchange interactions. We have examined the effects of more-distant-neighbor interactions on these properties for the bcc, sc, and fcc lattices.

An expansion of the internal energy function $E_\infty-E(K)$ may be obtained from the expansion of the free energy F in zero magnetic field given in (8) from the relation

$$\frac{E_\infty - E(K)}{J} = \frac{d}{dK}\left[\frac{-F(K)}{kT}\right]. \quad (27)$$

The internal-energy function evaluated at $K=K_c$ represents the critical energy and is denoted by $(E_\infty - E_c)/kT_c$. This is obtained from (27) as a power series in K_c. The critical energy is a very important critical parameter and represents the area below the high-temperature specific-heat curve ($T>T_c$) plotted on a reduced scale of temperature T/T_c

$$\frac{(E_\infty - E_c)}{kT_c} = \frac{1}{k}\int_1^\infty C_v dt. \quad (28)$$

Evaluation of the power series (27) at $K=K_c$ provides an estimate of this area.

The critical energy is also a useful parameter for comparison with experimental work as it is independent of the exchange integral. Similar considerations apply to the critical entropy denoted by $(S_\infty - S_c)/k$, which

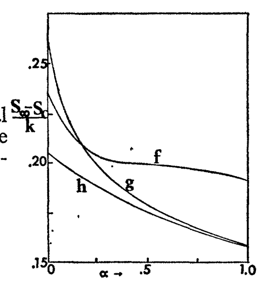

FIG. 10. The variation in the critical entropy with the strength of the second-neighbor interaction (g) sc lattice, (f) bcc lattice, (h) fcc lattice.

may be obtained as a power series in K_c (see Appendix II). The critical entropy provides another measure of the area below the high-temperature specific-heat curve given by

$$\frac{S_\infty - S_c}{k} = \frac{1}{k}\int_1^\infty \frac{C_v}{t} dt. \quad (29)$$

For the second-neighbor model the variations in the critical energy and entropy with α have been examined for values of α in the range $\alpha=0$ to $\alpha=1$. This has been done by forming the sequence of $[M,N]$ Padé approximants to the series (28) and (29) and evaluating each approximant at $K=K_c$ for various values of α. The convergence of the estimates for $(E_\infty - E_c)/kT_c$ and $(S_\infty - S_c)/k$ obtained in this way is generally fairly poor. The reason for this is that for values of $\alpha<0.25$ the series are erratic and consist of positive and negative terms. As the value of α increases beyond 0.25 the terms in the series are all positive but the coefficients do not show any smooth behavior as found in the susceptibility. The convergence of the approximants at $K=K_c$ is much better for the energy and improves greatly for both properties for values of α greater than 0.25. The results of this analysis are shown graphically in Figs. 9 and 10.

TABLE VII. Estimates for critical properties using the polynomials $e_r(\alpha)$, $j_r(\alpha)$ $r=1, \rightarrow 5$.

Property	Critical temperature		Energy $(E_\infty - E_c)/kT_c$		Entropy $(S_\infty - S_c)/k$	
Value of α	0	1	0	1	0	1
sc	1.759	6.801	0.495	0.260	0.203	0.159
bcc	2.555	4.921	0.426	0.336	0.212	0.191
fcc	4.069	6.812	0.385	0.259	0.204	0.158

Both $(E_\infty - E_c)/kT_c$ and $(S_\infty - S_c)/k$ decrease smoothly as α increases indicating that the area below the specific-heat curve decreases and that C_v approaches T_c more sharply.

To assess the accuracy of these estimates we have compiled a table of critical constants for various lattices obtained by using five terms in the corresponding series expansions. These are listed in Table VII. Similar estimates obtained from both ratio and Padé-approximant methods using more terms in the expansions (six in the case $\alpha=1$ and nine in the case $\alpha=0$) are given in Table VIII. Using these tables for comparison the critical energies and entropies of Table V (and those included in Figs. 9 and 10) are probably between 5% and 10% too low. In constructing Fig. 10, that part of the curve between $\alpha=0$ and $\alpha=0.25$ has been drawn by continuing the curves above $\alpha=0.25$ through the estimates at $\alpha=0$ from Table VIII.

Domb and Sykes[12] obtained a value of 0.265 for the critical entropy of the fcc lattice. These authors think that this estimate which differs from the value in Table VIII by 0.045 may need revising. The critical constants in Table VIII may be compared with the experimental work of Meidema, van Kempen, and Huiskamp[5] who measured $(E_\infty - E_c)/kT_c$ and $(S_\infty - S_c)/k$ for the ferromagnetic salts $CuK_2Cl_4 \cdot 2H_2O$ and $Cu(NH_4)_2Cl_4 \cdot 2H_2O$.

VIII. THE MAGNETIC SPECIFIC HEAT

Owing to the very poor nature of the magnetic specific-heat series for the nearest-neighbor model ($S=\frac{1}{2}$) it is difficult to obtain any clear indications of a singularity in this function. For the nearest-neighbor model, nine terms in expansion of the free energy in zero magnetic field for the loose-packed lattice are available.[4] The higher order $[M,N]$ Padé approximants to the logarithmic derivative of C_v for the bcc lattice indicate the presence of a singularity on the positive

TABLE VIII. Estimates for critical properties using six or more terms in corresponding series expansion.

Property	Critical temperature		Energy $(E_\infty - E_c)/kT_c$		Entropy $(S_\infty - S_c)/k$	
Value of α	0	1	0	1	0	1
sc	1.700	6.770	0.595	0.312	0.265	0.186
bcc	2.5477	4.925	0.460	0.357	0.235	0.210
fcc	4.0684	6.780	0.430	0.310	0.220	0.185

TABLE IX. The specific-heat singularity and its residue obtained from [1,2] of $(d/dK)\ \ln C_v$.

Lattice q	bcc (1,2) 14	sc (1,2) 18	fcc (1,2) 18	sc (1,2,3) 26	bcc (1,2,3) 26	fcc (1,2,3) 42
Critical point from C_v	···	0.1348	0.1375	0.1214	0.0963	0.05635
Residue at singularity in C_v	···	0.3096	0.3281	1.290	0.6798	0.943
Critical point from χ_0	0.2031	0.1477	0.1475	0.0948	0.0941	0.0545

real axis. More terms in this series are needed before any serious estimate of this transition temperature can be made. Previous authors[3,12] found that at high values of spin the series became smoother, but extrapolations are still unreliable. We have examined the specific heat series for the equivalent-neighbor-model lattices using six terms in (8).

To locate the transition temperature in the magnetic specific heat we have examined the [1,2] Padé approximant to the logarithmic derivative of the specific-heat series. These results are given in Table IX. Except for the bcc(1,2) lattice there is a very clear indication of a singularity in the approximants. An important feature of the results in Table IX is the fair agreement between the critical points obtained from the approximants to the specific heat and susceptibility series; the latter of course are very reliable estimates of the transition temperatures. The two estimates converge rapidly with increasing coordination number.

The specific heat may be expanded as a power series in reduced temperature $t(=T/T_c)$ by writing the series in the form

$$\frac{C_v}{k}=\sum_{r=2}^{\infty} c_r t^{-r}, \tag{30}$$

where K_c is obtained from the susceptibility series. We have calculated a sequence of Padé approximants to (30) and evaluated them over a range T_c/T from 0 to 1.5. Remembering that $t=1$ represents the true transition temperature and using the [2,3] approximant for comparison between the lattices, it was found that the specific heat gave a transition temperature at the values of $t^{-1}=t_c^{-1}$ given in Table X.

These results clearly demonstrate that as q increases t_c tends to unity, where the specific heat and the susceptibility predict the same transition temperature.

TABLE X. Values of t_c^{-1} for the equivalent-neighbor model.

Lattice q	bcc (1,2) 14	sc (1,2) 18	fcc (1,2) 18	sc (1,2,3) 26	bcc (1,2,3) 26	fcc (1,2,3) 42
t_c^{-1}	···	1.33	1.31	1.15	1.15	1.05

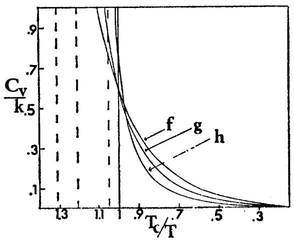

FIG. 11. The specific-heat curves above the Curie point, (f) fcc (1,2), (g) bcc (1,2,3), (h) fcc (1,2,3).

In Fig. 10 these evaluations are plotted for three lattices to illustrate this effect.

The data available are not sufficient to attach any significance to the residues of the singularities in the approximants, which are included in Table IX. To determine the nature of the transition in the specific heat, more terms are needed in the series expansion (8). The specific heat curves in Fig. 11 are in accord with the consideration of Sec. 7: that the area below the curves decreases as q increases, and that the curves become sharper in the critical region. For comparison we have repeated the above calculations for a Bethe lattice with $q=42$. The approximants gave no indication of a singularity on the real axis. In Fig. 12 an approximant for this lattice is compared with the corresponding approximant for the fcc(1,2,3) lattice, which has the same coordination number.

The marked difference in the curves f and g illustrate the dominant effects of the lattice structure in determining the onset of ferromagnetic ordering.

IX. SUMMARY

In this paper we have discussed some of the effects of more-distant-neighbor interactions on the critical and thermodynamic properties of the Heisenberg model of a ferromagnet. The theoretical approach used is the one originally introduced by Opechowski[1] where high-temperature $(T>T_c)$ power-series expansions are derived for all the thermodynamic functions. We have shown that for the three-dimensional cubic lattices the coefficients in some of these series expansions behave smoothly and provide a basis for accurate extrapolations. By applying the Padé-approximant and ratio methods[11] to these expansions we have discussed the effects of second-neighbor interactions and in some cases second- and third-neighbor interactions on the thermodynamic functions for values of T near T_c.

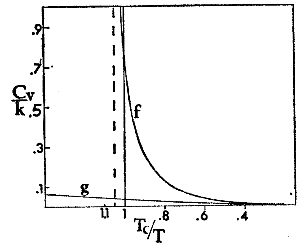

FIG. 12. Effects of the lattice structure on the specific-heat curve, (f) fcc (1,2,3) lattice (g) Bethe lattice with $q=42$.

In Sec. III it is shown that the coefficients in the susceptibility series for the equivalent-neighbor-model three-dimensional lattices behave very smoothly. The smooth behavior of these coefficients increases as the strength of the higher neighbor interactions increases. This effect is clearly seen in Fig. 4 where the ratio plots for the simple cubic lattice are shown for various values of α $(=J_2/J_1)$. It can be seen from Table I that the estimates of the Curie points (J/kT_c) of the three-dimensional equivalent-neighbor-model lattices obtained by using both Padé-approximant and ratio methods are in very good agreement.

In Sec. IV we discussed the variation of the Curie temperature $T_c(\alpha)$ with the relative strengths of the first- and second-neighbor interactions J_2/J_1 for values of J_1 and J_2 in the range $0\leqslant J_2/J_1\leqslant 1$. In this range it was found that to a good approximation $T_c(\alpha)$ is given by the linear relation

$$T_c(\alpha)=T_c(0)[1+m_1J_2/J_1].$$

Both the mean-field theory and the Green-function[22] theory of ferromagnetism predict a similar variation of $T_c(\alpha)$ for positive values of the exchange integrals. The values of kT_c/J_1 and m_1 obtained from series expansions, mean-field theory, and Green-function theory are compared in Tables III and IV. This analysis shows that the Green-function theory gives fairly reliable estimates of critical constants. The estimates of m_1 predicted by the mean-field theory are between 15% and 30% in error.

In Sec. V we used both the equivalent-neighbor-model and the second-neighbor-model series to investigate the high-temperature susceptibility for values of T near T_c. It has been shown by previous authors[12–14] that in this region χ_0 has the asymptotic form

$$\chi_0\sim A[1-(T_c/T)]^{-\gamma}.$$

By using ratio methods and Padé-approximant techniques we obtained estimates of γ for a wide range of lattice structures. These estimates are shown in Tables V and VI, where the coordination number of the lattices varies from $q=6$ to $q=42$. For this range of lattice structure the index γ appears to be independent of lattice structure and has a value $\gamma=1.31\pm0.03$.

In Sec. VI we discussed the effects of second-neighbor interactions $(-2J_2\sigma_k\cdot\sigma_l)$ on the reciprocal susceptibility curves in the high-temperature region $(T>T_c)$. The response of χ_0^{-1} to the presence of second-neighbor interactions is very small. The susceptibility is therefore very insensitive to the relative strengths of the first- and second-neighbor exchange integrals and consequently not very suitable for comparison with experimental work.

The thermal properties of the Heisenberg model where second- and third-neighbor interactions are included are discussed in Sec. VII. The variation of the critical energy and entropy $[(E_\infty-E_c)/kT_c$ and $(S_\infty-S_c)/k]$ with the magnitude of the second-neighbor interactions is shown in Figs. 8 and 9 for the three-dimensional lattices. Estimates of these two critical constants based on Padé-approximant methods are listed in Table VIII. Although the smoothness of the coefficients increases rapidly with α, the series expansion for the entropy and energy do not provide a very reliable basis for extrapolations, and consequently the values of $(E_\infty-E_c)/kT_c$ and $(S_\infty-S_c)/k$ in Table VIII may be between 5% and 10% in error. In Sec. VIII it is shown that as the coordination number of the lattice increases the Padé approximants to the magnetic specific-heat series give reliable estimates of the transition temperature but more terms in this expansion are needed to determine the nature of this transition.

In this paper we have been concerned only with the $S=\frac{1}{2}$ system and for the case where both J_1 and J_2 are positive. We have also derived the expansions (8) and (9) for the general spin case to the fifth order in reciprocal temperature. These expansions have been derived independently by Wojtowicz and Joseph[10] to the fourth order in reciprocal temperature. We are presently engaged upon the analysis of the general spin series and these results will be published elsewhere.

A preliminary analysis of the region for which $\alpha<0$ shows that the series expansions soon become erratic for small negative values of α. We are presently examining the susceptibility series (9) in this region. We hope that a more detailed analysis will enable us to investigate this interesting region more accurately.

ACKNOWLEDGMENTS

We are grateful to Professor C. Domb and to Dr. M. E. Fisher for the interest which they have taken in this work, and also to Dr. M. F. Sykes for his help in the configurational aspects of the work. We would like to express our thanks to the University of London Computer Unit for the use of their computational facilities, and to the Department of Scientific and Industrial Research for the award of a maintenance grant.

APPENDIX I

The polynomial coefficients of Eqs. (8) and (9) are listed below.

a. The simple cubic lattice

$\frac{1}{2}j_1=6\alpha+3\,,$

$\frac{1}{2}j_2=120\alpha^2+144\alpha+24\,,$

$\frac{1}{2}j_3=3312\alpha^3+6912\alpha^2+2664\alpha+264\,,$

$\frac{1}{2}j_4=117\,360\alpha^4+366\,336\alpha^3+239\,328\alpha^2+52\,416\alpha+3960\,,$

$\frac{1}{2}j_5=5\,104\,416\alpha^5+21\,764\,160\alpha^4+21\,043\,200\alpha^3+7\,583\,040\alpha^2+1\,196\,160\alpha+74\,928\,.$

$e_2=18\alpha^2+9$,

$e_3=108\alpha^3+216\alpha-18$,

$e_4=180\alpha^4+5760\alpha^2-576\alpha-162$,

$e_5=-5040\alpha^5+187\,200\alpha^3-17\,280\alpha^2-14\,400\alpha+2520$.

b. The body-centered-cubic lattice

$\frac{1}{2}j_1=3\alpha+4$,

$\frac{1}{2}j_2=24\alpha^2+96\alpha+48$,

$\frac{1}{2}j_3=264\alpha^3+2016\alpha^2+2520\alpha+832$,

$\frac{1}{2}j_4=3960\alpha^4+44\,544\alpha^3+100\,512\alpha^2+75\,072\alpha+18\,400$.

$\frac{1}{2}j_5=74\,928\alpha^5+1\,100\,160\alpha^4+3\,788\,160\alpha^3+4\,767\,360\alpha^2+2\,558\,400\alpha+504\,384$,

$e_2=9\alpha^2+12$,

$e_3=-18\alpha^3+216\alpha-24$,

$e_4=-162\alpha^4+2016\alpha^2-576\alpha+168$,

$e_5=2520\alpha^5+17\,280\alpha^3-5760\alpha^2-5760\alpha+1440$.

c. The face-centered-cubic lattice

$\frac{1}{2}j_1=3\alpha+6$,

$\frac{1}{2}j_2=24\alpha^2+144\alpha+120$,

$\frac{1}{2}j_3=264\alpha^3+3024\alpha^2+6552\alpha+3312$,

$\frac{1}{2}j_4=3960\alpha^4+66\,816\alpha^3+268\,128\alpha^2+323\,136\alpha+117\,360$,

$\frac{1}{2}j_5=74\,928\alpha^5+1\,650\,240\alpha^4+10\,211\,520\alpha^3+21\,816\,000\alpha^2+17\,917\,440\alpha+5\,104\,416$.

$e_2=9\alpha^2+18$,

$e_3=-18\alpha^3+216\alpha+108$,

$e_4=-162\alpha^4+576\alpha^2+4608\alpha+180$,

$e_5=2520\alpha^5+5760\alpha^3+112\,320\alpha^2+37\,440\alpha-5040$.

APPENDIX II

Equivalent-neighbor model zero-field partition function.

$$\log Z_{\text{per spin}}=\log 2+\sum_{l=2}^{\infty} e_l\frac{K^l}{2^l l!}$$

The expressions for the thermodynamic functions are given by the following formulas; internal energy, $(E_\infty-E)/J=(\partial/\partial K)[(\log Z(K)]$; specific heat, $C_v/k=K^2(\partial/\partial K)[(E_\infty-E)/J]$; entropy

$$\frac{(S_\infty-S)}{k}=\frac{1}{k}\int\frac{C_v(K)}{K}dK.$$

	fcc (1,2) $q=18$	sc (1,2) $q=18$	bcc (1,2) $q=14$
e_2	27	27	21
e_3	306	306	174
e_4	5 202	5 202	1 446
e_5	153 000	153 000	9 720
e_6	6 975 864	7 028 280	344 808

	fcc (1,2,3) $q=42$	sc (1,2,3) $q=26$	bcc (1,2,3) $q=26$
e_2	63	39	39
e_3	2 106	714	714
e_4	144 810	27 834	23 514
e_5	14 983 560	1 479 240	1 300 680
e_6	2 025 047 448	105 115 032	98 908 632

Equivalent-neighbor-model susceptibility series.

$$\frac{kT}{m^2}\chi_0=1+\sum_{l=1}^{\infty} j_l\frac{K^l}{2^l l!}$$

	fcc (1,2) $q=18$	sc (1,2) $q=18$	bcc (1,2) $q=14$
j_1	18	18	14
j_2	576	576	336
j_3	26 304	26 304	11 264
j_4	1 558 800	1 558 800	484 976
j_5	113 549 088	113 531 808	25 586 784
j_6	9 812 816 160	9 809 009 184	1 600 639 968

	fcc (1,2,3) $q=42$	sc (1,2,3) $q=26$	bcc (1,2,3) $q=26$
j_1	42	26	26
j_2	3 360	1 248	1 248
j_3	395 424	87 008	87 008
j_4	61 287 120	7 922 000	7 941 200
j_5	11 770 526 112	889 245 216	894 957 216
j_6	2 695 241 747 616	118 618 042 272	119 970 832 800

Series for the Bethe lattice. Susceptibility series.

$$\frac{kT}{m^2}\chi_0=1+\sum_{l=1}^{\infty} j_l\frac{K^l}{2^l l!}$$

	$q=42$	$q=26$
j_1	42	26
j_2	3 360	1 248
j_3	402 864	89 648
j_4	64 473 360	8 612 240
j_5	12 897 844 512	1 034 251 296
j_6	3 096 087 564 192	149 008 068 768

$$\log Z_{\text{per spin}}=\log 2+\sum_{l=2}^{\infty} e_l\frac{K^l}{2^l l!}$$

	$q=42$	$q=26$
e_2	63	39
e_3	−126	−78
e_4	−20 790	−7 878
e_5	209 160	79 560
e_6	37 064 664	8 497 944