collisions, and the contribution of light particles to the energy density is known to be small.

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CRITICAL PROPERTIES OF ISOTROPICALLY INTERACTING CLASSICAL SPINS CONSTRAINED TO A PLANE

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High-temperature expansions of the susceptibility and internal energy (specific heat) are presented for general lattice structure for a system of isotropically interacting unit vectors (or "classical spins") which are constrained to lie in a plane. A phase transition ($T_c > 0$) is indicated for two-dimensional lattices; the expected result $T_c = 0$ is found in one dimension, but only upon choosing a more suitable expansion parameter than J/kT. Similarities with the corresponding expansions of the $S=\frac{1}{2}$ Ising and classical Heisenberg models are pointed out; in particular, it is found that certain critical properties of this planar model appear to be bounded on one side by the Ising model and on the other side by the Heisenberg model.

Consider a system of isotropically interacting unit vectors constrained to lie in the x-y plane and described by the Hamiltonian $\mathcal{H}^P = -2J\Sigma_{\langle ij\rangle}(S_{ix}S_{jx}+S_{iy}S_{jy})$, where $-2J$ is the energy of a nearest-neighbor pair of parallel "spins".[1] Bowers and Joyce[2] have very recently published, for the fcc, bcc, and sc lattices, the high-temperature expansions for the reduced susceptibility

$$\bar{\chi}^P \equiv \chi^P/\chi_{\text{Curie}}^P = 1+\sum_{n=1}^{\infty} a_n^P (J/kT)^n \qquad (1)$$

and the specific heat

$$C^P = k\sum_{n=2}^{\infty} c_n^P (J/kT)^n \qquad (2)$$

of this planar model. Here we present the corresponding expansions for general lattice structure (together with an additional term for loose-packed lattices), and analyze the series for one- and two-dimensional as well as for three-dimensional lattices. We also re-express the susceptibility and internal-energy series in terms of the new expansion parameter $w \equiv I_1(K)/$

$I_0(K)$, where $K \equiv 2J\beta \equiv 2J/kT$ and the $I_\nu(K)$ are modified Bessel functions of the first kind. We find that this new expansion for the planar model bears certain similarities to the corresponding expansions for the Ising model (one-dimensional spins) and the classical Heisenberg model (three-dimensional spins).

The expansions (1) and (2) can both be obtained directly from a knowledge of the spin correlation function $\langle \vec{S}_f \cdot \vec{S}_g \rangle_\beta{}^P$. The diagrams required to calculate $\langle \vec{S}_f \cdot \vec{S}_g \rangle_\beta{}^P$ are identical to those used in the calculation[3] of $\langle \vec{S}_f \cdot \vec{S}_g \rangle_\beta{}^H$ for the classical Heisenberg model; so one need only re-evaluate—for two-dimensional spins—the averages associated with each diagram The requisite single-spin averages are easily shown to be

$$\langle S_x^{2k} S_y^{2l} \rangle_0^P = \frac{\Gamma(k+\frac{1}{2})\Gamma(l+\frac{1}{2})}{\pi\Gamma(k+l+1)}, \tag{3}$$

where $\langle \mathcal{O} \rangle_0^P$ denotes the $\beta = 0$ thermal average of the operator $\mathcal{O}$.[4] We have obtained general-lattice expressions for the coefficients a_n^P in Eq. (1) (through order $n=8$ for close-packed and through order $n=9$ for loose-packed lattices), and for the coefficients c_n^P of Eq. (2) (through order $n=9$ for close-packed and order $n=10$ for loose-packed lattices).

We find the specific heat series for the two-dimensional lattices to be so irregular[5] that they cannot be readily used to estimate values of the critical temperatures T_c. However, the susceptibility series appear to diverge at nonzero T_c for the triangular and square lattices (see Fig. 1). This is particularly intriguing, since the indicated phase transition, if real (for there is nothing rigorous about extrapolating from a finite number of terms of an infinite series), would have to be to a new type of low-temperature phase[6] with no "infinite-range" order

$$M \propto [\lim_{R\to\infty} \langle \vec{S}_0 \cdot \vec{S}_R \rangle_\beta]^{1/2},$$

yet with sufficient "long-range" order so that $\chi \propto \sum_R \langle \vec{S}_0 \cdot \vec{S}_R \rangle_\beta$ diverges to infinity.

For the one-dimensional linear chain, both the susceptibility and specific-heat series are too irregular to permit extrapolation to the

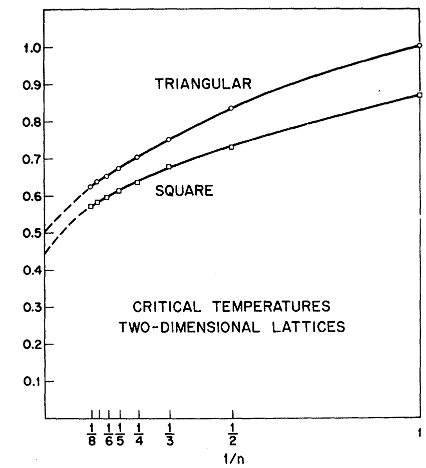

FIG. 1. The sequence $\rho_n \equiv a_n/a_1 a_{n-1}$ for the triangular lattice ($z=6$) and the sequence $\rho_{n+1}' \equiv (a_{n+1}/a_{n-1})^{1/2}/a_1$ for the square lattice ($z=4$). If these sequences approach limiting values as $n\to\infty$, these values are kT_c/zJ.

known[1,7] result $T_c = 0$.[8] However, we have obtained a much smoother series (and one which extrapolates directly to $T_c = 0$) by re-expanding the susceptibility in the new expansion parameter $w \equiv I_1(K)/I_0(K)$. Our choice of w was motivated by the similarity among the exact expressions

$$\bar{\chi}^I = (1+v)/(1-v), \tag{4a}$$

$$\bar{\chi}^P = (1+w)/(1-w), \tag{4b}$$

and

$$\bar{\chi}^H = (1+u)/(1-u) \tag{4c}$$

for the reduced susceptibilities of, respectively, the $S=\frac{1}{2}$ Ising, classical planar,[7] and classical Heisenberg models[9]; here $v \equiv \tanh K$ and $u \equiv \mathcal{L}(K) = \coth K - 1/K$.[10] The coefficients A_n^P in the new expansion

$$\bar{\chi}^P = 1 + \sum_{n=1}^{\infty} A_n^P w^n \tag{5}$$

are obtained from the general-lattice expressions for the a_n^P of Eq. (1) by substituting in Eq. (5) the small-argument expansion of $w = I_1(2j)/I_0(2j)$,

$$\left\{ j \sum_{n=0}^{\infty} \frac{j^{2n}}{n!(n-1)!} \right\} \left\{ \sum_{n=0}^{\infty} \frac{j^{2n}}{(n!)^2} \right\}^{-1} = j\{1 - \tfrac{1}{2}j^2 + \tfrac{1}{3}j^4 - (11/48)j^6 + (19/120)j^8 + \cdots\}, \tag{6}$$

Table I. General-lattice expressions for the D_n^V through order $n=8$ (through order $n=9$ for the subclass of loose-packed lattices). Notation as in C. Domb, Advan. Phys. 9, 149 (1960).

$$D_3^P = -6p_3$$

$$D_4^P = -8p_4 - 3p_3$$

$$D_5^P = -10p_5 - 4p_4 + 7.5p_3 + 6p_{5a}$$

$$D_6^P = -12p_6 - 5p_5 + 10p_4 + 13p_3 + 6\,[p_{6a} + p_{6b}] + 8p_{6c} + 38p_{5a}$$

$$D_7^P = -14p_7 - 6p_6 + 12.5p_5 + (7/3)p_4 + 5.25p_3 + 6\,[p_{7a} + p_{7b} + p_{7f}]$$
$$+ 10p_{7c} + 8\,[p_{7d} + p_{7e}] + 66p_{7g} + 18p_{6a} + 41p_{6b} + 24p_{6c} + 12p_{6d} + (244/3)p_{5a}$$

$$D_8^P = -16p_8 - 7p_7 + 15p_6 + (35/12)p_5 + (44/3)p_4 - (343/24)p_3 + 6\,[p_{8a} + p_{8b} + p_{8c} + p_{8d}]$$
$$+ 10\,[p_{8e} + p_{8f} + p_{8p}] + 8\,[p_{8g} + p_{8h} + p_{8j} + p_{8k}] + 12p_{8\ell} + 4p_{8q} + 72\,[p_{8r} + p_{8s}]$$
$$+ 44\,[p_{7a} + p_{7b}] + 74p_{7c} + 24\,[p_{7d} + p_{7e}] + 18p_{7f} + 64p_{7g} + 10p_{7h} + 105p_{6a}$$
$$+ (181/3)p_{6b} + 22p_{6c} - (37/3)p_{5a}$$

$$D_9^P = -8p_8 + 3.5p_6 + (26/3)p_4 + 6\,[p_{9k} + p_{9\ell}] + 8p_{9j} + 10p_{9m}$$
$$+ 18p_{8c} + 24p_{8h} + 96p_{8r} + 8p_{8t} + (97/3)p_{7a} + 53p_{6a}$$

in powers of $j \equiv \beta J = \frac{1}{2}K$, and then equating coefficients of successive powers of j in Eqs. (1) and (5).[11] We have in this fashion calculated general-lattice expressions for the coefficients A_n^P in Eq. (5).

These expressions are too lengthy to list here; they may, however, be directly obtained from the (much less lengthy) expressions in Table I for a new set of coefficients D_n^P defined by

$$\bar{\chi}^P \equiv (1-\sigma w)^{-2}[1-(\sigma-1)w-\sigma w^2 + \sum_{n=3}^{\infty} D_n^P w^n], \quad (7)$$

upon using the recursion relation

$$A_n^P = D_n^P + 2\sigma A_{n-1}^P - \sigma^2 A_{n-2}^P. \quad (8)$$

From Table I we see that for a "Bethe lattice" (a lattice with no polygons), $D_n^P = 0$ for $n \geq 3$ and Eq. (7) reduces to the result of the Bethe-Peierls approximation,

$$\bar{\chi}^P = (1+w)/(1-\sigma w). \quad (9)$$

In fact, the Bethe-Peierls result (9) is exact for lattices with no closed circuits. But common ("multiply connected") crystal structures found in nature possess many closed circuits, and the D_n^P are by no means zero. Thus including terms in the high-temperature expansion (7) beyond order $n=2$ corresponds in some sense to taking account of the "multiple connectivity" of the lattice, and one might expect extrapolations based upon high-temperature expansions carried beyond second order to be more realistic than the Bethe-Peierls approximation.[12]

The specific heat is by definition the temperature derivative of the internal energy, which for a linear chain is

$$E^I = -2Jv, \quad (10a)$$

$$E^P = -2Jw, \quad (10b)$$

and

$$E^H = -2Ju. \quad (10c)$$

Consequently, we have used Eq. (6) together with our general-lattice expressions for the c_n^P of Eq. (2) to obtain the coefficients B_n^P in the new expansion

$$E^P = -\frac{J}{w}\sum_{n=2}^{\infty} B_n^P w^n; \quad (11)$$

the general-lattice expressions for the B_n^P are given in Table II.[12]

The behavior of the new coefficients A_n^P and B_n^P is generally smoother than that of the coefficients a_n^P and c_n^P in the old expansions, thereby increasing the subjective reliability of extrapolations based thereon. Moreover, the planar critical properties which we studied—T_c, γ, α, ν, and η—appear to be bound-

Table II. General-lattice expressions for the $B_n{}^V$ through order $n=9$ (through order $n=10$ for the subclass of loose-packed lattices). Notation as in C. Domb, Advan. Phys. 9, 149 (1960).

$$B_2^P = z$$

$$B_3^P = 6p_3$$

$$B_4^P = 8p_4$$

$$B_5^P = 10p_5 - 9p_3$$

$$B_6^P = 12p_6 - 12p_4 - 10.5p_3 - 18p_{5a}$$

$$B_7^P = 14p_7 - 15p_5 + p_3 - 42p_{7g} - 21p_{6b} - 49p_{5a}$$

$$B_8^P = 16p_8 - 18p_6 - (41/3)p_4 + (71/4)p_3 - 48\,[p_{8r} + p_{8s}] - 24\,[p_{7a} + p_{7b}]$$
$$- 56p_{7c} - 96p_{6d} - 28\,[3p_{6a} + p_{6b}] + (89/3)p_{5a}$$

$$B_9^P = 18p_9 - 21p_7 + (5/3)p_5 + (485/24)p_3 - 54\,[p_{9e} + p_{9f} + p_{9h}] - 126p_{9g}$$
$$- 27\,[p_{8a} + p_{8b}] - 63\,[p_{8e} + p_{8p}] - 72\,[p_{8q} + p_{7h}] - 31.5p_{7b} + 40.5p_{7c}$$
$$- 63p_{7f} + 156p_{7g} + 0.75p_{6b} + 360p_{6d} + 180p_{5a}$$

$$B_{10}^P = 20p_{10} - 24p_8 + 2p_6 + 25p_4 - 60\,[p_{10b} + p_{10c}] - 30p_{9k} - 70\,[p_{9m} + p_{8c}] - 80p_{8t}$$
$$+ 72p_{8r} - (107/3)p_{7a} + 136p_{6a}$$

ed on one side by those predicted by Ising calculations, and on the other side by those predicted by the classical Heisenberg model. E.g., for the fcc lattice, $t_c \equiv T_c/T_M \cong 0.816$, 0.802, and 0.795 for the Ising, planar, and Heisenberg models, respectively; similarly, $\gamma \cong 1\frac{4}{16}$, $1\frac{5}{16}$, and $1\frac{6}{16}$; $\alpha \cong \frac{1}{8}$, 0, ~ -0.05.

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[1]This "planar" model was solved by E. Lieb, T. Shultz, and D. Mattis [Ann. Phys. (N.Y.) 16, 407 (1961)] for the linear antiferromagnetic chain for $S=\frac{1}{2}$ without the restriction that the spins interact isotropically. More recently, the classical isotropic planar model has received attention {V. G. Vaks and A. I. Larkin, Zh. Eksperim. i Teor. Fiz. 49, 975 (1965) [translation: Soviet Phys.—JETP 22, 678 (1966)]; L. P. Kadanoff et al., Rev. Mod. Phys. 39, 395 (1967); Ref. 2} as a lattice model for the λ transition in a Bose fluid; so far as we know, this has not been proven to be rigorous.

[2]R. G. Bowers and G. S. Joyce, Phys. Rev. Letters 19, 630 (1967). Note that Bowers and Joyce define $K \equiv J/kT$; whereas we define $K \equiv 2J/kT$.

[3]H. E. Stanley, Phys. Rev. 158, 537 (1967).

[4]Thus the illustrative example in Fig. 6 of Ref. 3 is changed only in that the averages on the second line of Fig. 6 must be evaluated using Eq. (3) [i.e., $\frac{1}{5}\frac{1}{3}$ is replaced by $\frac{3}{8}\frac{1}{2}$].

[5]The specific-heat series for the triangular, square, and honeycomb lattices in the classical Heisenberg model are also too irregular to predict a value of T_c. Indeed, it may well be that there exists no infinity in the specific heats of these models for two-dimensional lattices. See, e.g., the concluding remarks in B. Jancovici, Phys. Rev. Letters 19, 20 (1967).

[6]The spontaneous magnetization, order parameter, or "infinite-range order" M has been proven to be zero for the planar model by N. D. Mermin and H. Wagner, Phys. Rev. Letters 17, 1133 (1967). However, the proof fails to exclude the possibility of a novel type of second-order phase transition with infinite susceptibility but zero order parameter. E.g., low-temperature approximations for the Heisenberg model [F. J. Dyson, unpublished] and for the classical isotropic planar model [F. Wegner, to be published] predict $\langle \vec{S}_0 \cdot \vec{S}_R \rangle_\beta \sim R^{\lambda T}$ so that $M=0$; yet for sufficiently small T, $\chi = \infty$. See also J. W. Kane and L. P. Kadanoff, Phys. Rev. 155, 80 (1967); T. M. Rice, Phys. Rev. 140, A1889 (1965); H. E. Stanley and T. A. Kaplan, Phys. Rev. Letters 17, 913 (1966), and J. Appl. Phys. 38, 975 (1967).

[7]G. S. Joyce, Phys. Rev. 155, 478 (1967).

[8]The linear chain susceptibility $\bar{\chi}^H = \sum_n a_n{}^H (J/kT)^n$ for the classical Heisenberg model is also too irregular to extrapolate; a much smoother series which extrapolates directly to $T_c = 0$ was recently obtained [H. E. Stanley, Phys. Rev. 164, 709 (1967)] by re-expanding $\bar{\chi}^H$ in the new parameter $u \equiv \mathcal{L}(K) = \coth K - 1/K$.

[9]M. E. Fisher, Am. J. Phys. 32, 343 (1964).

[10]A clearer and more precise terminology is to call the Ising, planar, and Heisenberg models "classical isotropic models" for, respectively, one-, two-, and three-dimensional spins (or "vectors"). It is possible

to solve the linear chain exactly (as well as to calculate high-temperature expansions) for arbitrary-dimensionality spins [H. E. Stanley, to be published].

[11]Thus, e.g., $A_1^P = a_1^P = z$; $A_2^P = a_2^P = z\sigma$; $A_3^P = a_3^P + \frac{1}{2}A_1^P = z\sigma^2 - 6p_3$ [notation as in H. E. Stanley, Phys. Rev. 158, 546 (1967)]. The A_n^P are easily deduced from Table I and Eqs. (7) and (8).

[12]Expansions analogous to Eq. (7) and Eq. (11) have been carried out for the Ising and classical Heisenberg models by, respectively, M. F. Sykes [J. Math. Phys. 2, 52 (1961)] and Stanley [Ref. 8]. The remarks concerning the Bethe-Peierls approximation [Eq. (9)] also apply to these other two models, with w replaced by v and u, respectively.

CALORIMETRIC INVESTIGATION OF HYPERFINE INTERACTIONS IN RARE-EARTH METALS*

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Specific-heat measurements have been the principal source of information relating to hyperfine interactions in the rare-earth metals. This has been true primarily because the method is applicable to any sample. The low-temperature specific heat C of these metals may be written

$$C = C_L + C_E + C_M + C_N, \tag{1}$$

where the terms represent the lattice, electronic, magnetic, and nuclear (hyperfine) contributions, respectively. For most of the rare-earth metals $C_L + C_E + C_M$ contribute less than 1% to C at temperatures below ≈0.2°K. With few exceptions previous specific-heat measurements have extended only down to ≈0.4°K, hence leaving C_N partially obscured by the other contributions. In order to determine C_N uniquely we have made heat-capacity measurements on the metals Pr, Nd, Sm, Tb, Dy, Ho, and Tm within the temperature range 0.02-0.4°K. Our data suggest the existence of a cooperative interaction between nuclei.

The polycrystalline metal samples[1] of roughly 10-g mass were cooled in an adiabatic-demagnetization cryostat. Heat capacities were measured to an accuracy of about ±1%. Temperatures were determined to ±0.2%+Δ, where Δ (0<Δ<0.001°K) is due to the shape correction of the cerium-magnesium-nitrate magnetic thermometer. Details will be presented in a forthcoming publication.[2]

Of the rare earths the metal most amenable to analysis should be Tm, since there is only one stable isotope (Tm^{169}) and that has a nuclear spin I of $\frac{1}{2}$ resulting in only two energy levels. Our data for Tm are shown in Fig. 1 with the data of Trolliet[3] and of Lounasmaa.[4] In view of the obvious discrepancies let it be noted that our sample was rerun one month after the original run, having been remounted with a different heater and with the thermometer recalibrated. It was run after exposure to magnetic fields <10 G and >10^4 G. All data are included in Fig. 1; the data therefore represent a unique heat capacity for our sample. The arrow in Fig. 1 indicates that temperature below which $C = C_N$ to within roughly 1%.[4]

C_N is a Schottky-type heat capacity given by

$$C_N = \frac{R}{(kT)^2} \frac{\sum_{i=-I}^{I} \sum_{j=-I}^{I} (W_i^2 - W_i W_j) \exp[-(W_i + W_j)/kT]}{\sum_{i=-I}^{I} \sum_{j=-I}^{I} \exp[-(W_i + W_j)/kT]}, \tag{2}$$

where R and k are the gas and Boltzmann constants and W_i are the energy levels of the nucleus which are typically expressed as

$$W_i/k = a' I_z + P[I_z^2 - \tfrac{1}{3}I(I+1)]. \tag{3}$$

The two terms arise from the interaction of the hyperfine fields with the nuclear magnetic dipole moment and the nuclear electric quadrupole moment, respectively. The hyperfine fields are primarily due to unpaired $4f$ electrons which are rather well isolated within the host ion, and which are polarized at the temperatures of interest here.

For Tm no attempts at nmr have been successful as yet, but Mössbauer measurements[5]