

## Critical Temperatures of Ising Lattice Films

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Ising lattices consisting of  $n=2, 3, 4,$  and  $5$  interacting plane square lattice layers are studied by high-temperature series expansions. Specifically, seven to nine terms of the zero-field susceptibility expansion have been obtained for (a) free-surface boundary conditions (in which each surface spin interacts with only five nearest neighbors); and (b) periodic boundary conditions (in which all spins interact equivalently with six nearest neighbors). Estimates of the critical temperatures  $T_c(n)$  obtained by ratio and Padé approximant techniques are presented. These results are consistent with the conjectures that  $T_c(\infty) - T_c(n)$  varies with thickness  $n$ , as  $n^{-\lambda}$  with  $\lambda=1$  in case (a) and  $\lambda=1/\nu_c \approx 1.56$  in case (b); but other, somewhat larger, values of  $\lambda$  are not excluded.

### I. INTRODUCTION

THE properties of the Ising model have been investigated extensively for many two- and three-dimensional (and even higher-dimensional) lattices.<sup>1,2</sup> In addition, Kramers and Wannier,<sup>3</sup> Onsager,<sup>4</sup> and Fisher and Ferdinand<sup>4</sup> have considered one case of what might loosely be called "intermediate" dimensionality, namely, the  $n \times \infty$  square-lattice torus (or cylinder). In this case for finite  $n$  there is no true phase transition but the height and location of the maximum of the specific-heat peak were studied. In this paper we present some results for the analogous situation intermediate between the infinite two-dimensional square lattice and the infinite three-dimensional simple cubic lattice, namely, an  $n \times \infty \times \infty$  simple cubic lattice "film" formed by stacking  $n$  layers of the plane square lattice. (A section of such a lattice for  $n=3$  is shown in Fig. 1.)

The two-layer film ( $n=2$ ) has, in fact, been considered previously by Ballentine.<sup>5</sup> We have confirmed his numerical calculations and, in addition, obtained high-temperature expansions for the zero-field susceptibility for three- and four-layer lattices. A true phase transition occurs now for each value of  $n$ . Our ultimate aim has been not only to determine the critical temperatures  $T_c(n)$  for the individual values of  $n$ , but also to estimate, as far as possible, the form of the dependence of  $T_c(n)$  on the number of layers for large  $n$ . Knowledge of this dependence would be valuable in interpreting experiments on a variety of real films. The present work represents a first step in this program.

In the simple two-layer case ( $n=2$ ), all sites are equivalent (having a coordination number of 5) which

greatly simplifies the work of deriving the series coefficients. But this simplification is evidently lost for  $n \geq 3$  when the surface spins still have coordination 5, while the nonequivalent interior sites have coordination number 6. On the other hand, we may alternatively impose *periodic* or cyclic boundary conditions in the vertical direction by merely assuming that a spin on the top layer interacts via a direct nearest-neighbor bond with the corresponding spin on the bottom layer. In this case all sites are again equivalent (and no identifiable surface layers exist). This cyclic model is clearly analogous to the  $n \times \infty$  torus or cylinder considered in Refs. 3 and 4. Although it does not correspond closely to any physically realizable geometry, it is theoretically interesting in assessing the significance of surface effects as distinct from "finite-thickness" effects. We have therefore calculated series for cyclic multilayer lattices with  $n=2, 3, 4,$  and  $5$  layers.

The layout of the paper is as follows. The derivation of the series expansion coefficients is outlined in Sec. II. The series are analyzed by ratio and Padé approximant techniques in Sec. III to obtain estimates of the critical temperatures  $T_c(n)$ . The form of the variation of  $T_c$  with the number of layers  $n$  is discussed in Sec. IV, where plans for future work are mentioned.

### II. CALCULATION OF SERIES

The Hamiltonian of the Ising model with nearest-neighbor interactions in an external magnetic field  $H$  is

$$\mathcal{H} = -J \sum_{\langle i,j \rangle} s_i s_j - mH \sum_i s_i, \quad (1)$$

where  $s_i = \pm 1$  is the spin variable associated with  $i$ th lattice site, ( $i=1,2,3,\dots,N$ ) and the first sum runs over all nearest-neighbor pairs of lattice sites  $\langle i,j \rangle$ . For a

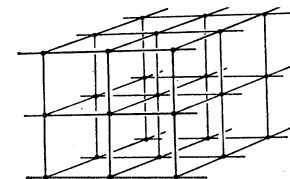


FIG. 1. A section of the three-layer simple cubic lattice with free surfaces.

<sup>1</sup> C. Domb, *Advan. Phys.* **9**, 149 (1960).

<sup>2</sup> M. E. Fisher, *Rept. Progr. Phys.* **30**, 615 (1967).

<sup>3</sup> H. A. Kramers and G. H. Wannier, *Phys. Rev.* **60**, 252 (1941); **60**, 263 (1941).

<sup>4</sup> L. Onsager, *Phys. Rev.* **65**, 117 (1944). Onsager confirmed the earlier conjecture of Kramers and Wannier (Ref. 3) that the height of the maximum in the specific heat is proportional to  $\ln n$ . See also A. E. Ferdinand and M. E. Fisher, *Phys. Rev.* **185**, 832 (1969), who consider a general  $m \times n$  torus and establish analytically the displacement of the position of the maximum from the limiting  $T_c$ .

<sup>5</sup> L. E. Ballentine, *Physica* **30**, 1231 (1964).

ferromagnet, the coupling parameter<sup>6</sup> or exchange constant  $J$  is positive; lastly,  $m$  denotes the magnetic moment per spin. The isothermal susceptibility,  $\chi = \partial M / \partial H$ , may be expanded at high temperatures in zero field in powers of

$$v = \tanh(J/kT), \quad (2)$$

in the form

$$(kT/m^2)\chi_0(v) = 1 + \sum_{r=1} a_r v^r, \quad (3)$$

where  $a_r$  is twice the term linear in  $N$ , the total number of lattice sites, in the number of ways of embedding all possible magnetic graphs of  $r$  lines on the lattice in question.<sup>1,2,6</sup> A magnetic graph is one which contains two, and only two, odd vertices, i.e., vertices at which an odd number of lines meet. The embeddings we consider are weak,<sup>7</sup> in the sense that two vertices of a graph  $G$  embedded on adjacent lattice sites need *not* be joined by a line of the graph. The number of (weak) embeddings per lattice site is the (weak) "lattice constant"<sup>11,2,7</sup> and is denoted  $(G)$ .

For  $n=2$ , it is convenient to distinguish

$$v = \tanh(J/kT),$$

for the inlayer or horizontal  $x$  bonds, from

$$v' = \tanh(J'/kT),$$

for the between-layer or vertical  $z$  bonds. The main feature in the calculation of the lattice constants is then a subdivision into contributions from embeddings involving zero, one, two, . . . ,  $z$  bonds, the remaining bonds being  $x$  bonds. As a trivial example, we see that the lattice constant  $(-)$  for a single line can be written

$$(-) = (2, \frac{1}{2}),$$

where the first figure, 2, refers to the embeddings with no  $z$  bonds, while the second figure,  $\frac{1}{2}$ , is the number of embeddings (per site) with one  $z$  bond. By calculating the more elaborate constants similarly, one generates a double-power series for  $\chi_0$  in the two variables  $v$  and  $v'$ . We have determined the lattice constants for the 50 magnetic graphs containing nine lines or less that can be embedded in the two-layer lattice thus obtaining all terms in the expansion of  $\chi_0$  of the form  $v^r v'^s$  for  $r+s \leq 9$ . On setting  $v=v'$ , we obtain the series for the standard two-layer lattice discussed by Ballentine.<sup>5</sup> Our coefficients agree exactly with those he found which provides a welcome check. On the other hand, by setting  $J'=2J$  and reexpanding  $v'$  in terms of  $v$ , we obtain the correct expansion for the two-layer *cyclic* lattice since, in effect, this lattice is simply the ordinary free-surface lattice with an extra vertical bond between each site in the top layer and its neighbor in the bottom layer.

<sup>6</sup> T. Oguchi, J. Phys. Soc. Japan 6, 31 (1951).

<sup>7</sup> M. F. Sykes, J. Essam, B. R. Heap, and B. Hiley, J. Math. Phys. 7, 1557 (1966).

In the case  $n=3$ , the cyclic lattice with periodic boundary conditions is close-packed in the sense that polygons of odd length, including triangles, occur on the lattice. Consequently, a number of additional lattice constants have to be evaluated in each order. In fact, 84 graphs now contribute up to eighth order which is as far as the calculations have so far been extended. Since all lattice sites are still equivalent, however, the calculation of these lattice constants follows standard procedures. The calculations for the four- and five-layer cyclic lattices are simpler because of the absence of triangles even though odd polygons of length 5, 7, 9, . . . can occur on the five-layer cyclic lattice. For this reason we have been able to extend the calculations up to ninth order on these lattices without excessive effort.

For the three- and four-layer lattices with free surfaces, the evaluation of the lattice constants is appreciably more arduous because of the nonequivalence of surface and interior sites, which prevents the straightforward application of the usual techniques. For these lattices we have decomposed each lattice constant into contributions from embeddings which span one, two, three, and four layers. If  $(G)^n$  denotes the lattice constant for a connected graph  $G$  on the cyclic lattice of  $n$  layers and  $(G)_l^n$  denotes the contributions to  $(G)^n$  from embeddings which span exactly  $l$  layers, we clearly have

$$(G)^n = \sum_{l=1}^n (G)_l^n. \quad (4)$$

When  $n=2$ , we see, recalling the previous discussion, that  $(G)_1^2$  is just the lattice constant for embeddings involving no  $z$  bonds while  $(G)_2^2$  is the sum over those embeddings involving one, two, . . .  $z$  bonds. It is also clear that

$$(G)_1^2 = (G)_1^3 = \dots = (G)_1^n, \quad (5)$$

for all  $n$ . More generally, if we consider the  $(n+1-l)$  vertical translations of an embedding which spans exactly  $l$  layers, we easily see that

$$(G)_l^n = n^{-1}(n+1-l)l(G)_l^l. \quad (6)$$

Thus for a three-layer lattice, for example, we can use the two-layer data to obtain  $(G)_2^3 = (\frac{2}{3})(G)_2^2$  and, hence, we need to count only those new embeddings of  $G$  that span all three layers, i.e., to evaluate the constant  $(G)_3^3$ . For any finite graph there is a maximum number of layers  $L(G)$  which can be spanned. A check on the calculations is thus provided by the tabulated constants for the simple cubic lattice<sup>1</sup> (corresponding to  $n=\infty$ ) since by combining (4) and (6) and letting  $n \rightarrow \infty$ , we obtain

$$(G)^{\infty} = (G)^{\infty} = \sum_{l=1}^{L(G)} l(G)_l^l. \quad (7)$$

Nevertheless, the labor involved, particularly in counting the linear chains manually, is considerable. (In this

initial study we have not used a computer to assist directly in the counting task.) We have thus obtained the series for the three- and four-layer free-surface lattices only up to seventh order.

The coefficients for all the series studied are presented in Table I.

III. ANALYSIS OF SERIES

To estimate the critical temperatures  $T_c(n)$  from the series given in Table I, we have utilized the well-known ratio and Padé approximant techniques.<sup>2,3</sup> Figure 2 illustrates the ratio plots for the various free-surface and cyclic lattices together with the limiting cases  $n=1$  (plane square) and  $n=\infty$  (simple cubic). For both free and cyclic boundary conditions, the plots for finite  $n \geq 2$  exhibit appreciable curvature. This is particularly noticeable for the cyclic lattices where, in fact, the ratios  $\mu_r(n) = a_r(n)/a_{r-1}(n)$  depart from the values  $\mu_r(\infty)$  for the simple cubic lattice, only for  $r \geq n$ . This phenomenon represents a change over from characteristically three-dimensional behavior at low values of  $r (< n)$  to characteristically two-dimensional behavior at high values of  $r (> n)$ . More specifically, the initial terms will tend to exhibit a slope  $g = \gamma - 1$  on the ratio plot corresponding to an exponent  $\gamma = \gamma_3 = 1.25$  while the high-order terms are expected to yield an asymptotic slope of  $\gamma = \gamma_2 = 1.75$  typical of the standard two-dimensional lattices.<sup>1,2</sup> Correspondingly, when  $T$  exceeds  $T_c$  by an appreciable amount,  $\chi_0$  will behave roughly like  $(T - T_c')^{-\gamma_3}$ , where  $T_c' \gg T_c$ , while close to the true critical point  $T_c$ , the characteristically two-dimensional variation as  $(T - T_c)^{-\gamma_2}$  is expected to take over. (This type of behavior can be seen explicitly in the spherical model in the corresponding situation intermediate between three- and four-dimensional lattices; then  $\gamma_3 = 2$  and  $\gamma_4 = 1$ .) For the two-layer lattices where the series are sufficiently long (about four times the number of layers) to span the change-over region, the apparent limiting behavior of the ratio plots provides direct support for the above surmise. For the lattices with

TABLE I. Series coefficients for high-temperature susceptibility of simple cubic lattices with  $n$  layers and cyclic or free-boundary conditions.

$n$	$n=2$		$n=3$		$n=4$		$n=5$	
	Free	Cyclic	Free	Cyclic	Free	Cyclic	Free	Cyclic
1	5	6	$5\frac{1}{2}$	6	$5\frac{1}{2}$	6	6	6
2	20	28	$23\frac{1}{2}$	30	25	30	30	30
3	80	130	$102\frac{3}{4}$	148	$114\frac{1}{2}$	150	150	150
4	304	564	$433\frac{3}{4}$	706	506	724	726	726
5	1 152	2 438	$1 822\frac{3}{4}$	3 322	2 234	3 490	3 508	3 508
6	4 236	10 132	$7 478\frac{3}{4}$	15 364	9 660	16 490	16 690	16 690
7	15 528	41 794	$30 569\frac{3}{4}$	70 222	41 648	77 826	79 234	79 234
8	55 924	169 652		317 574		362 356	373 106	373 106
9	200 808	682 870				1 684 966	1 751 810	1 751 810
10	712 868*							

\* From Ballentine (Ref. 5).

<sup>3</sup> G. A. Baker, Advan. Theoret. Phys. 1, 1 (1965).

TABLE II. Critical parameters for the simple cubic lattices of  $n$  layers.

$n$	$v_c = \tanh(J/kT_c)$		$kT_c(n)/\bar{q}J$		$T_c(n)/T_c(\infty)$	
	Cyclic	Free	Cyclic	Free	Cyclic	Free
1		0.4142136		0.5672963		0.50311
2	0.269 ± 0.001	0.301 ± 0.001	0.604	0.646	0.804	0.717
3	0.240 ± 0.001	0.267 ± 0.001	0.681	0.685	0.906	0.810
4	0.2305 ± 0.0010	0.252 ± 0.003	0.710	0.706	0.945	0.861
5	0.226 ± 0.002		0.725		0.964	
∞		0.21815		0.75172		1.0000

more layers, however, the present series are shorter, both in proportion to  $n$  and absolutely. Consequently, the available ratios do not clearly pass through the change-over region. Some estimate of the extent of curvature of the ratio plots must thus be made if the  $r = \infty$  intercepts, and hence the critical temperatures, are to be determined with reasonable precision. Accordingly, we have examined plots of  $\mu_r(n)$  versus  $1/(r + \epsilon)$  with various value of  $\epsilon$  (in the range 0-2). The variation of  $\epsilon$  has no effect on the leading asymptotic behavior but suitable values of  $\epsilon$  yield straighter plots at low values of  $r$ .<sup>2</sup> In estimating the limiting intercept we have chosen values of  $\epsilon$  such that the over-all slope of the plot was close to the expected value  $g = 1.75 - 1 = 0.75$ .

The values of  $v_c$  and the critical temperatures obtained with this procedure are listed in Table II with an indication of the range of uncertainty.

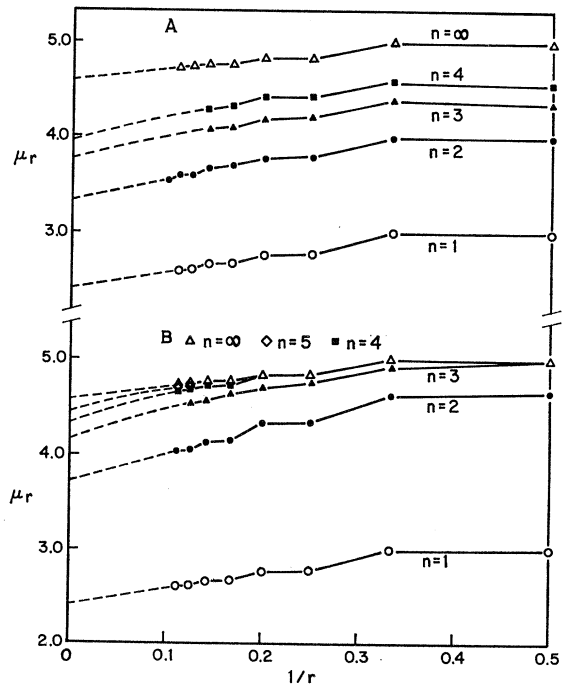


FIG. 2. Ratios  $\mu_r = a_r(n)/a_{r-1}(n)$  of successive coefficients of the susceptibility expansions versus  $1/r$  for (A) the free surface and (B) the cyclic  $n$ -layer lattices. (The simple cubic lattice corresponds to  $n = \infty$ .)

Owing to the relative shortness of most of our series, the precision is much less than obtained for the standard two- and three-dimensional lattices.<sup>1,2</sup> Just as in these cases, the error estimates have *not* been obtained by rigorous mathematical arguments. Rather, the range of values obtained for a particular model from the various extrapolation techniques described are carefully examined and an estimate, albeit partly subjective, of the probable range of uncertainty is made. We plan to obtain longer series which will yield increased precision and, in addition, enable us to study lattices with more layers.

Padé approximant analyses of the series yielded results similar to those found from the ratio plots. In particular, the location of the poles of direct Padé approximants to the function  $[\chi_0(v)]^{-4/7}$ , which is expected to have a simple pole at  $v_c$ , were quite consistent with the estimates in Table II. Thus, by way of example, a diagonal or near diagonal sequence of Padé estimates of  $v_c$  for the  $n=4$  periodic lattice is: 0.2392, 0.2335, 0.2314, 0.2304, 0.2303, and 0.2305. These compare with the corresponding linear extrapolants of the ratios  $\mu_r(4)$  with  $\epsilon = +2$ , viz, 0.2232, 0.2288, 0.2263, 0.2290, 0.2283, and 0.2306. As seen in Table II, our estimate of the critical parameter for this lattice is  $v_c = 0.2305 \pm 0.0010$ .

#### IV. DEPENDENCE OF CRITICAL TEMPERATURE ON NUMBER OF LAYERS

The solid curve labeled (a) in Fig. 3 shows the variation of  $v_c(n) = \tanh[J/kT_c(n)]$  versus  $1/n$  for the lattices with free surfaces. Figure 4(a) is the same plot for the cyclic lattices. In both cases there are clear departures from linearity with  $n^{-1}$ , but these are most pronounced for the cyclic lattices where  $v_c(n)$  approaches the three-dimensional value  $v_c(\infty)$  rather

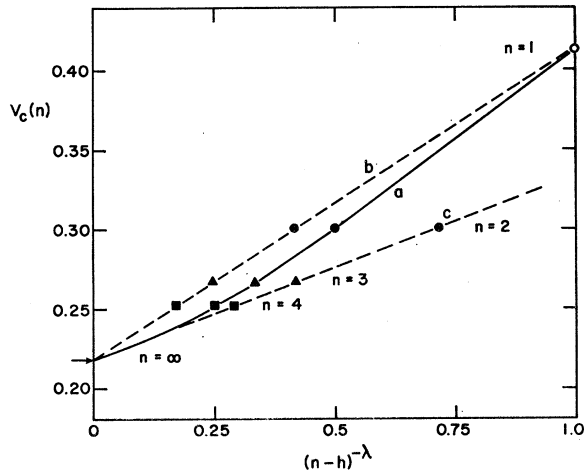


FIG. 3. Plots of the critical constants  $v_c = \tanh(J/kT_c)$  of the  $n$ -layer free-surface lattices versus  $(n-h)^{-\lambda}$  with (a)  $\lambda=1$ ,  $h=0$ , (b)  $\lambda=1.27$ ,  $h=0$ , and (c)  $\lambda=1$ ,  $h=0.60$ .

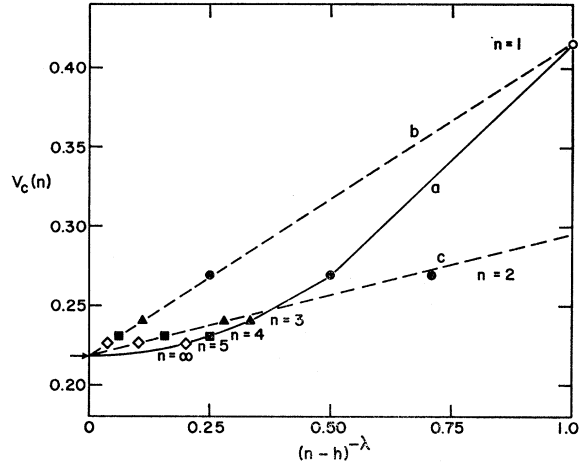


FIG. 4. Plots of the critical constants of the  $n$ -layer cyclic lattices versus  $(n-h)^{-\lambda}$  with (a)  $\lambda=1$ ,  $h=0$ , (b)  $\lambda=2$ ,  $h=0$ , and (c)  $\lambda=1/\nu_s \approx 1.56$ ,  $h=0.75$ .

rapidly. For large  $n$ , it is natural to expect a power law of the form

$$\epsilon_n = 1 - T_c(n)/T_c(\infty) \approx c_0/n^\lambda \quad (n \rightarrow \infty). \quad (8)$$

If we attempt to determine the constants  $\lambda$  and  $c_0$  by a direct fit to the simple two-parameter formula

$$v_c(n) - v_c(\infty) = A/n^\lambda, \quad (9)$$

which implies (8), we find that surprisingly good fits can be obtained for both free-surface and cyclic lattices with  $\lambda = 1.27 \pm 0.05$  and  $\lambda = 2.00 \pm 0.08$ , respectively, and  $A \approx 0.196$  (in both cases). These fits are shown by the dashed lines labeled (b) in Figs. 3 and 4, respectively. The uncertainties of the fits are principally determined by the lack of precision in the values of  $T_c(n)$ .

However, we are not aware of any other arguments that would support the validity of the fitted values of  $\lambda$ . On the contrary, Fisher and Ferdinand<sup>9</sup> have argued that for lattices with free surfaces one should rather expect  $\lambda=1$  (as  $n \rightarrow \infty$ ). This follows simply from a mean field argument. Thus if  $q$  is the coordination number of the lattice, mean field theory predicts

$$kT_c = qJ. \quad (10)$$

For an  $n$ -layer simple cubic lattice with free surfaces the mean coordination number is reduced from  $q=6$  to

$$\bar{q} = 6n - 2/n, \quad (11)$$

and so the mean field prediction becomes

$$\epsilon_n = \frac{1}{3}/n. \quad (12)$$

Similarly, the Bethe approximation for a uniform lattice gives

$$v_c = \tanh(J/kT_c) = 1/(q-1), \quad (13)$$

<sup>9</sup> M. E. Fisher and A. E. Ferdinand, Phys. Rev. Letters **19**, 169 (1967).

which on using (11)  $q$  leads to the relative shift

$$\epsilon_n = [c_0 + (c_1/n) + \dots]/n, \quad (\text{free surface}) \quad (14)$$

with  $c_0 = 0.4132$  in place of  $c_0 = \frac{1}{3}$ . Higher-order terms like those in (14) must be expected generally. They may be allowed for simple and conveniently by fitting to the modified expression

$$v_c(n) - v_c(\infty) = A/(n-h)^\lambda, \quad (15)$$

which is asymptotically equivalent to (14). Such a fit with the exponent fixed at  $\lambda = 1$  is shown in Fig. 3, curve (c): The parameter values are  $h = 0.60$  and  $A = 0.116 \pm 0.001$ . Evidently, the fit is quite satisfactory and certainly no worse than the *ad hoc* fit (8). The value of  $A$  corresponds to a constant  $c_0 \simeq 0.55$ , which compares reasonably with the Bethe prediction of 0.41.

In the case of the cyclic lattices the coordination number  $q$  remains unchanged (at  $q = 6$ ) and the mean field and Bethe approximations, both of which take no account of closed polygons on the lattice, predict *no change* in  $T_c$ . To the extent that  $T_c(n)$  approaches  $T_c(\infty)$  much more rapidly for the cyclic lattices than for those with free surfaces this prediction is, roughly, confirmed. Indeed the fit (b) in Fig. 4, corresponding to  $\lambda = 2$ , is consistent with (14) if  $c_0 = 0$  but  $c_1 \neq 0$ . On the other hand, some general heuristic arguments<sup>9</sup> suggest that when  $n$  is large the deviations from the infinite simple cubic lattice susceptibility should first become significant at temperatures so close to  $T_c(\infty)$  that the range of correlation in the infinite lattice,  $\xi(T) \simeq a(\Delta T/T_c)^{-\nu_3}$ , is of the same order as  $na$ , the circumference of the finitely-layered lattice ( $a$  being the lattice spacing). One imagines the mode of propagation of the correlations must change when significant correlation occurs around the lattice. If the deviation  $T_c(\infty) - T_c(n)$  were of the same order as this effect, one would expect,  $\lambda = 1/\nu_3$ , that is

$$\epsilon_n \simeq c_0/n^{1/\nu_3} \quad (\text{cyclic lattices}). \quad (16)$$

To test this hypothesis (while allowing for the higher-order corrections which must still be expected), we have fitted the cyclic critical points to (16) with  $\lambda = 1.56 \simeq 1/\nu_3$ .<sup>10</sup> As shown by the line (c) in Fig. 4,

<sup>10</sup> M. E. Fisher and R. J. Burford, Phys. Rev. **156**, 583 (1967); M. A. Moore, D. Jasnow, and M. Wortis, Phys. Rev. Letters **22**, 940 (1969).

the fit is very reasonable for  $n \geq 3$  with  $h = 0.75$  but is less satisfactory for  $n = 1$  and 2; but this may not be very surprising since the underlying picture loses credibility for such thin lattices.

In summary then, the conjectures (a) [free surface]  $\lambda = 1$ , and (b) [cyclic]  $\lambda = 1/\nu_3$ , based on the various heuristic theoretical arguments, are quite consistent with the estimated critical points and yield reasonable two-parameter fits for  $n \geq 2$ . However, the shortness of the present series limits both the precision of the estimated critical temperatures and the number of layers that can be studied. As a consequence, the theoretical conjectures must not be regarded as unambiguously confirmed; in particular, the larger values (a)  $\lambda \simeq 1.27$  and (b)  $\lambda \simeq 2.0$  provide *ad hoc*, but equally satisfactory, fits to the available results. To resolve this question, longer series are clearly essential. We are undertaking the calculation of further terms by using the computer-based methods of enumerating lattice configurations developed by Sykes, Martin, and their co-workers. Just how many terms can be obtained is of course uncertain at this stage. We feel that at least eleven- or twelve-term series for lattices with up to seven or eight layers will be necessary to enable more definite conclusions to be drawn regarding the form of the dependence of the critical temperatures on the number of layers. [We do not, however, anticipate that the present estimates will be invalidated and these already confirm a number of significant features of the Fisher-Ferdinand conjectures.] We also hope to extend the work to the Heisenberg and related models which have a bearing on experiments on superfluidity in helium films, as well as to magnetic materials.

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