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# Phase boundary of Ising antiferromagnets near $H=H_c$ and $T=0$: Results from hard-core lattice gas calculations

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Ising antiferromagnets in a near-critical magnetic field at low temperatures are equivalent to hard-core lattice gases. Using this connection and the existing series-expansion results for hard-core lattice gases, we determine the slope of the phase boundary at $T=0$ for the square (sq), plane-triangular (pt), simple cubic (sc), and body-centered cubic (bcc) antiferromagnets. The slope is negative for the sq and pt lattices and nearly zero for the sc case. For the bcc lattice a positive slope is obtained, indicating that the phase boundary bulges above the zero-temperature critical field. We also test Müller-Hartmann and Zittarz's postulate for the critical curve of the sq Ising antiferromagnet. A renormalization-group treatment of the hard-square lattice gas yields a critical activity $z^*=3.7959\pm0.0001$, which is in agreement with series-expansion and finite-lattice estimates but at variance with the postulated $z^*=4$. The same calculation gives $\nu=0.999\pm0.001$ for the correlation-length exponent, thus supporting the conjecture that the transition of the hard-square lattice gas belongs to the Ising universality class.

## I. INTRODUCTION

The nearest-neighbor Ising antiferromagnet in a magnetic field is an interesting model from both theoretical and experimental points of view. In spite of its simplicity, this model provides a reasonable description for a variety of systems[1] like physisorbed monolayer films, uniaxial antiferromagnets, and binary alloys with variable composition.

Qualitatively, the phase diagram following from the model is well understood (Fig. 1): there are two phases (paramagnetic and antiferromagnetic) which are separated by a critical line running from the Nèel temperature at zero field ($T=T_N$, $H=0$) to the critical field at zero temperature ($T=0$, $H=H_c$). Since the nature of the symmetry breaking is not affected by the magnetic field, one expects that the transition at finite field is of second order and belongs to the same universality class as the zero-field Ising model.

The details of the shape of the phase boundary are known less well. Molecular-field calculations[2,3] resulted in curves bulging above the critical field (Fig. 1, curve a), a feature which was believed to be an artifact of the approximation. This belief was supported by Fisher's exact solution of a two-dimensional superexchange model[4] yielding a curve similar to that of b on Fig. 1, and also by series expansions[5] and renormalization-group calculations.[6] In contradiction to this postulated shape, two independent Monte Carlo calculations[7,8] showed clearly that in the case of the bcc lattice there were two phase transitions provided $H$ was slightly above $H_c$.

A recent development is Müller-Hartmann and Zittarz's (MHZ) derivation[9] of the critical line $T_c(H)$ for the sq lattice (Fig. 1, curve b). They use special interface configurations for calculating the surface tension of the system and then by setting it to zero, $T_c(H)$ is obtained. Although their method is approximate, it gives exact values for $T_c(0)$ and $H_c$, and the small $H$ expansion of $T_c(H)$ also agrees with series

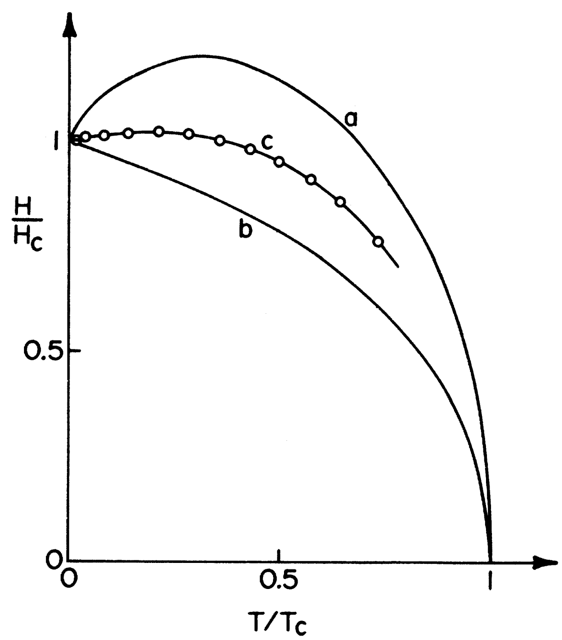


FIG. 1. Phase diagram of Ising antiferromagnets in a magnetic field: (a) molecular-field theory (Ref. 2), (b) Müller-Hartmann and Zittarz's approximation for the square lattice (Ref. 9), and (c) Monte Carlo calculations for the bcc lattice (Ref. 8).

 

expansions combined with the smoothness postulate.[10] Within their 5% accuracy, subsequent Monte Carlo[11] and renormalization-group[12] calculations confirmed the MHZ result; so it was conjectured to be exact.

Baxter *et al.*[13] point out, however, that the above conjecture can be checked at low temperatures, as well, where the phase boundary has the form

$$H=H_c+a^*T \quad , \tag{1}$$

and the slope $a^*$ is related to the critical activity $z^*$ of the hard-square lattice gas by

$$a^*=-\tfrac{1}{2}\ln z^* \quad . \tag{2}$$

By extending the high-density series for the order parameter of the lattice gas up to 24 terms, they obtained $z^*=3.7962(1)$ with the estimated accuracy given in parentheses in units of the last significant digit. This value of $z^*$ was in agreement with earlier work,[14–16] but in conflict with the conjectured value $z^*=4$, so they concluded that although the MHZ $T_c(H)$ was a good approximation, it was not exact.

Our aim with this paper is twofold. First (Sec. II), the Baxter *et al.* conclusion is confirmed by determining $z^*$ from a renormalization-group calculation. Using Nightingale's[17] phenomenological scaling method in which the renormalization-group equation is derived by comparing the correlation lengths of finite width strips of the system, we obtain a series of estimates for $z^*$ which converges rapidly as the width of the strips increases. An extrapolation gives $z^*=3.7959(1)$ in good agreement with the result of Baxter *et al.*

Secondly (Sec. III), we explore the implications of the known nearest-neighbor exclusion lattice gas results for Ising antiferromagnets on different lattices. In particular, it is shown that, while for the plane-triangular (pt) and sq lattices the phase boundary at $T=0$ and $H=H_c$ starts with negative slope, for the simple cubic (sc) lattice the phase boundary is nearly horizontal, and for the bcc lattice the slope is positive; i.e., if $H$ is just above $H_c$, then there are two phase transitions as the temperature is increased.

As a by-product of our renormalization-group calculation, we obtained a series of estimates for the critical exponent of the correlation length of the hard-square lattice gas. The estimates extrapolate to $\nu=0.999(1)$, indicating that $\nu$ is equal to the Ising value $\nu_I=1$. This result, put together with the series estimate of the order-parameter exponent[13] $\beta=0.1249(1)$ $(\beta_I=0.125)$, shows that the hard-square lattice gas belongs to the Ising universality class. Although this is expected on the basis of ground-state symmetry considerations, series expansions had led to speculations about the possibility of another set of exponents.[13]

## II. RENORMALIZATION GROUP FOR THE HARD-SQUARE LATTICE GAS

The hard-square lattice gas is defined as a collection of particles restricted to the sites of a square lattice. Multiple occupancy of a site or simultaneous occupancy of neighboring sites are prohibited and an activity $z$ is assigned to each occupied site. Thus the partition function of the system is given by

$$Z=\sum_k A_k z^k \quad , \tag{3}$$

where $A_k$ is the number of ways $k$ lattice sites can be chosen so that none of them are nearest neighbors. It can be easily shown that in the limit $T\to 0$, the partition function of the square-lattice Ising antiferromagnet in a magnetic field $H=H_c+aT$ reduces to Eq. (3) with the activity related to $a$ by $z=\exp \times(-2a)$. It follows then that the approach of $H_c$ along the critical line $(a=a^*)$ corresponds to the critical point $(z=z^*)$ of the lattice gas [Eqs. (1) and (2)].

The critical properties of the hard-square lattice gas have been discussed in many works[14–16,18] using various analytical and numerical approaches. Here we present the first renormalization-group treatment of the system. Since our aim is the precise determination of $z^*$, we use Nightingale's method[17] which has proved to be reliable and accurate in finding the critical point and the critical exponents of two-dimensional systems like the Ising model[17] and its various generalizations.[19]

Nightingale's derivation of approximate renormalization-group equations is phenomenological. The critical properties of an infinite lattice are obtained from the properties of finite lattices by calculating the correlation lengths of strips of infinite length but finite width (say $L$ and $L'$ rows) and then relating them according to the Kadanoff picture:

$$\frac{1}{L'}\xi_{L'}(x')=\frac{1}{L}\xi_L(x) \quad , \tag{4}$$

where $x$ and $x'$ are the values of a parameter along a line in the space of coupling constants. In the simplest case of the Ising model, $x$ is the nearest-neighbor coupling while in our case of hard-square lattice gas $x$ is the activity $z$.

Once the renormalization-group transformation $x'=R(x)$ is known from Eq. (4), the calculation of the critical point and critical indices follows standard lines.[20]

The usefulness of Nightingale's method depends on whether $\xi_L$ can be computed for large enough $L$'s so that the estimates of the critical parameters following from Eq. (4) could be smoothly extrapolated to the exact values at $L,L'\to\infty$. A practical way of calculating the correlation length of two-dimensional systems is the transfer-matrix technique.[21] The eigenvalues of the transfer matrix which are the largest ($\Lambda_1$) and second largest ($\Lambda_2$) in absolute value yield $\xi_L$ through[22]

$$\xi_L^{-1} = \ln(\Lambda_1^{(L)}/|\Lambda_2^{(L)}|) \ , \tag{5}$$

and the calculation of $\Lambda_1^{(L)}$ and $\Lambda_2^{(L)}$ is greatly simplified by decomposing the $2^L \times 2^L$ transfer matrix into a direct sum of submatrices and finding the largest eigenvalues of only those submatrices which contain $\Lambda_1^{(L)}$ and $\Lambda_2^{(L)}$. The decomposition of the transfer matrix of the hard-square lattice gas is described in great detail in Ref. 16. We mention only that for the largest system we considered, a strip of 14 rows, $\Lambda_1$ and $\Lambda_2$ are determined from two $49 \times 49$ matrices. Thus the numerical part of our work is easily done by a computer.

When deriving a renormalization-group transformation, care must be taken about preserving the ground-state symmetry of the system.[23] In the high activity ($z \to \infty$) limit, the ground state of the hard-square lattice gas is twofold degenerate: the particles occupy one of the two interpenetrating square sublattices into which the square lattice can be divided. Since strips with periodic boundary conditions display twofold-degenerate ground state only if they have an even number of rows, $L$ and $L'$ in Eq. (4) must be even numbers.

A further consideration for the choice of $L$ and $L'$ comes from experience with the Ising model.[17] In that case the best converging sequences of critical parameters were obtained by proceeding towards an infinitesimal transformation: $L$ and $L'$ were increased simultaneously while the difference $L - L'$ was kept minimal. Following this line, we considered transformations with $L' = L - 2$ and increased $L$ from 4 to 14.

The results for the critical activity ($z^*$) and the correlation-length exponent ($\nu$) are displayed in Table I. Both sequences of estimates converge well as the system size increases and a simple extrapolation procedure can be designed[17] on the basis that in the limit $L \to \infty$ the deviations from the exact values ($z^* - z^*_\infty$, $\nu - \nu_\infty$) are expected to go as $L^{-p}$. One plots $\ln(z^* - z^*_\infty)$ and $\ln(\nu - \nu_\infty)$ against $\ln(L^{-1} + L'^{-1})$ and those values of $z^*_\infty$ and $\nu_\infty$ are chosen for which the linearity of the plots becomes optimal. This method gave extremely good results for the Ising model[17] and the quality of the fit is very good in our case as well (Fig. 2). A somewhat subjective assessment of the accuracy of the extrapolation can be obtained from the maximum changes induced in $z^*_\infty$ and $\nu_\infty$ by changing $\ln(L^{-1} + L'^{-1})$ into $\ln L^{-1}$ or $\ln[2/(L + L')]$ and leaving out one or two points from the small-$L$ region.

The extrapolated value of $z^* = 3.7959(1)$ agrees well with the series-expansion results[13,14] and with the estimates from finite-size systems.[15,16] Even if much more conservative error estimates are made, all

TABLE I. Critical activity ($z^*$) and correlation-length exponent ($\nu$) of the hard-square lattice gas as calculated from Nightingale's method using strips of widths $L$ and $L'$. Results of series expansions and extrapolations from finite-size systems are also included. The estimated accuracy is indicated between parentheses in units of the least significant digit.

| $L'/L$ | $z^*$ | $\nu$ |
|---|---|---|
| 2/4 | 4.1010 | 1.161 |
| 4/6 | 3.8538 | 1.058 |
| 6/8 | 3.8166 | 1.028 |
| 8/10 | 3.8056 | 1.016 |
| 10/12 | 3.8014 | 1.010 |
| 12/14 | 3.7992 | 1.007 |
| Extrapolation | 3.7959(1) | 0.999(1) |
| Series | | |
| Ref. 14 | 3.80(2) | |
| Ref. 13 | 3.7962(1) | |
| Transfer matrix | | |
| Ref. 15 | 3.80(4) | |
| Ref. 16 | 3.7966(3) | |

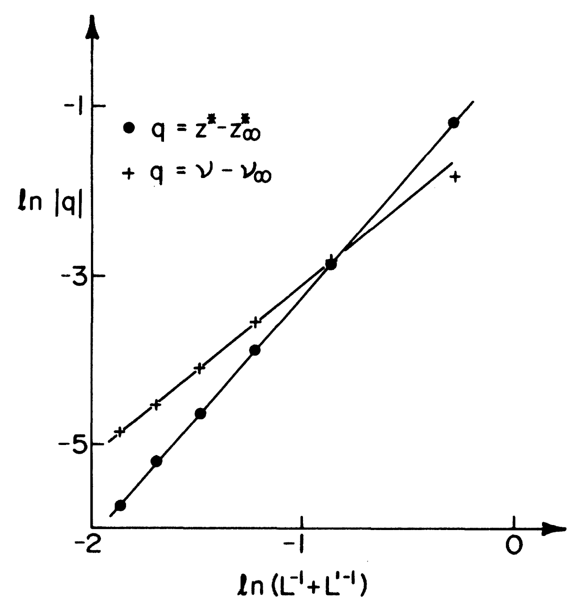


FIG. 2. Extrapolation of the critical activity ($z^*$) and correlation-length exponent ($\nu$) estimates obtained from renormalization-group transformations involving strips of widths $L$ and $L'$. The limiting values $z^*_\infty$ and $\nu_\infty$ are chosen from the requirement that the mean-square deviation from a linear fit would be minimal.

these numbers are still in disagreement with the MHZ value $z^* = 4$ indicating that the surface configurations they consider are not sufficient for an exact evaluation of the surface tension of the square-lattice Ising antiferromagnet in finite fields.

We note here that Nightingale's method has recently been applied[12] to the square-lattice Ising antiferromagnet in a magnetic field. In this case the reduction of the transfer matrix is not very effective so the calculation has been restricted to strips with maximum width of eight rows. Although the estimates of $T_c(H)$ approach a curve which is different from the MHZ result (see Table I in Ref. 12), the difference is small and Sneddon's conclusion is in favor of the MHZ curve attributing the difference to the uncertainties of the renormalization-group method. In view of the lattice gas results, Sneddon's conclusion should be reversed and the critical curve obtained by him should be considered as the more accurate one.

The order parameter of the hard-square lattice gas is one dimensional (twofold-degenerate ground state), and the interaction between the squares is of short range. Accordingly, the phase transition should be in the Ising universality class. Our results for the correlation length exponent $\nu = 0.999(1)$ (Table I) supports this notion, especially if it is considered together with the very accurate value of the order-parameter exponent $\beta = 0.1249(1)$ determined from high-density series[13] (the Ising values are $\nu_I = 1$ and $\beta_I = \frac{1}{8}$).

The result $\nu = \nu_I$ is in agreement with calculations on finite-size systems[15,16] which show that, to a high degree of accuracy, the maximum compressibility of finite width strips is proportional to the logarithm of the width; i.e., the compressibility has a logarithmic singularity ($\alpha = 0$). Assuming scaling, this means $\nu = 1$.

On the other hand, the $\nu = \nu_I$ result is in disagreement with series-expansion work, which, depending on the length of the series and on the method of their analysis, yield a nondiverging compressibility[14] or a compressibility with singularity stronger than logarithmic.[13] In our view, the series are not long enough to detect a logarithmic singularity reliably.

## III. IMPLICATIONS OF THE HARD-CORE LATTICE GAS RESULTS

So far, we have discussed the square lattice. Equations (1) and (2), however, are not restricted to this case. For any given lattice, the low-temperature phase boundary [parameter $a^*$ in Eq. (1)] of the Ising antiferromagnet in a magnetic field and the critical activity $z^*$ of the nearest-neighbor exclusion lattice gas are related by Eq. (2). Since series-expansion estimates of $z^*$ are available[13,14,24] for the sq, pt, sc, and bcc lattices, they can be used to determine $a^*$ for the corresponding antiferromagnets.

The results of translating $z^*$ into $a^*$ are displayed in Table II. Besides $a^*$, we included there another quantity of interest, the critical magnetization $m_c$ which is the limiting value of the magnetization as the $T = 0$ and $H = H_c$ point is approached along the critical line. It can be obtained from the critical density ($\rho_c$) of the lattice gas through

$$m_c = 1 - 2\rho_c \quad . \tag{6}$$

The following aspects of the results should be remarked.

*a. sq lattice.* The slope of the phase boundary has been discussed in detail in Sec. II. The value of $m_c = 0.264(2)$ is in good agreement with the estimates from finite-size systems $m_c = 0.258(8)$ (Ref. 15) and $m_c = 0.26448(1)$ (Ref. 16).

*b. pt lattice.* The pt Ising antiferromagnet does not order in zero field ($T_N = 0$). Its finite field ($0 < H < H_c$) ground state is threefold degenerate, and the phase transition in finite field belongs to the same universality class as the three-state Potts model.[25] In contrast, the sq, sc, and bcc Ising antiferromagnets have finite Nèel temperature, display a doubly degenerate ground state, and consequently their phase transition belongs to the Ising universality class. In view of the above differences, it is not surprising that the value of $a^*$ for the pt lattice does not fit into the trend which can be seen for the other lattices, namely that $a^*$ increases as the number of nearest neighbors is increased.

The numerical values of both $a^* = -1.20(1)$ and $m_c = 0.445(6)$ compare well with finite lattice[15] [$a^* = 1.205(5)$ and $m_c = 0.442(10)$] and Monte Carlo[26] estimates ($a^* \approx 1.15$) but they differ from the renormalization-group result[27] $a^* = 0.91$ and $m_c = 0.52$.

*c. sc lattice.* The phase boundary near $T = 0$ is almost horizontal [$a^* = -0.04(3)$]. Although $a^*$ is

TABLE II. Slope of the phase boundary ($a^*$) and critical magnetization ($m_c$) for the square (sq), plane-triangular (pt), simple cubic (sc), and body-centered cubic (bcc) antiferromagnets at their zero-temperature critical point. The estimates are from series-expansion works on nearest-neighbor exclusion lattice gases (Refs. 13, 14, and 24).

| Lattice | $a^*$ | $m_c$ |
|---|---|---|
| sq | −0.66700(2) | 0.260(8) |
| pt | −1.20(1) | 0.445(5) |
| sc | −0.04(3) | 0.573(20) |
| bcc | 0.13(3) | 0.645(20) |

determined with quite a large relative error, the negative value indicates that there is no bulge above $H_c$. This conclusion is supported by calculations of the magnetization in the limit of a horizontal approach to the zero-temperature critical point[28] ($H = H_c$ and $T \to 0$). The resulting values of $m_0 = 0.62$ (finite lattice) and $m_0 = 0.59$ (Monte Carlo) are larger than the critical magnetization $m_c = 0.57(2)$, indicating that the phase boundary near $T = 0$ is below the critical field ($a^* < 0$). It should be noted, however, that the uncertainties in $m_0$ and $m_c$ are of the same order as their difference, so the magnetization results present only a weak case for $a^* < 0$.

*d. bcc lattice.* $a^* > 0$, so the phase boundary near $T = 0$ is above $H_c$. While renormalization-group[6] and high-temperature series[5] calculations do not reproduce this feature of the phase diagram, the Monte Carlo findings[8] [$a^* = 0.16(2)$ and $m_c = 0.644(6)$] are in excellent agreement with our results [$a^* = 0.13(3)$ and $m_c = 0.645(20)$].

The existence of the bulge is a "lattice effect." For $H > H_c$ the ground state of the antiferromagnet is ferromagnetic and the low-temperature excitations are flips of an arbitrary number of non-neighboring spins. Close to $H_c$, the question of how many spins are flipped and whether their density is sufficient for their ordering on one of the sublattices is decided by the combinatorics of the corresponding nearest-neighbor exclusion lattice gas. The trend for two-sublattice antiferromagnets (Table II) is that as the coordination number of the lattice is increased the transition occurs at lower and lower densities of flipped spins. With the bcc lattice we arrive at high enough coordination number so that the low-temperature spin flips are numerous enough for antiferromagnetic ordering to take place.

*e. sq, sc, and bcc lattices.* As the number of nearest neighbors is increased, $a^*$ and $m_c$ tend towards their molecular-field values[2,29] $a^* = +\infty$ ($H \approx H_c - \frac{1}{2}T \times \ln T$) and $m_c = 1$. This is in accord with the expectation that the molecular-field theory becomes exact as the coordination number goes to infinity.

In summary, we can see that the connection between Ising antiferromagnets and hard-core lattice gases is extremely useful for obtaining qualitative as well as quantitative results about the low-temperature ordering of Ising antiferromagnets in a magnetic field.

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[1]See, e.g., M. E. Fisher, Rep. Prog. Phys. 30, 615 (1967) and references cited therein.

[2]C. G. B. Garrett, J. Chem. Phys. 19, 1154 (1951).

[3]J. M. Ziman, Proc. Phys. Soc. London Sect. A 64, 1108 (1951).

[4]M. E. Fisher, Proc. R. Soc. London Ser. A 254, 66 (1960).

[5]A. Bienenstock and J. Lewis, Phys. Rev. 160, 393 (1967).

[6]G. D. Mahan and F. H. Claro, Phys. Rev. B 16, 1168 (1977).

[7]T. E. Shirley, Phys. Rev. B 16, 4078 (1977).

[8]D. P. Landau, Phys. Rev. B 16, 4164 (1977).

[9]E. Müller-Hartmann and J. Zittartz, Z. Phys. B 27, 261 (1977).

[10]D. C. Rapaport and C. Domb, J. Phys. C 4, 2684 (1971).

[11]D. C. Rapaport, Phys. Lett. A 65, 147 (1978).

[12]L. Sneddon, J. Phys. C 12, 3051 (1979).

[13]R. J. Baxter, I. G. Enting, and S. K. Tsang, J. Stat. Phys. (in press).

[14]D. S. Gaunt and M. E. Fisher, J. Chem. Phys. 43, 2840 (1965).

[15]L. K. Runnels and L. L. Combs, J. Chem. Phys. 45, 2482 (1966).

[16]F. H. Ree and D. A. Chesnut, J. Chem. Phys. 45, 3983 (1966). In this work, the remarkable accuracy in determining the critical parameters is achieved by considering the transfer matrix of strips with up to 22 rows and extrapolating the parameters along the minimum of $\Lambda_1^{(L)}/\tilde{\Lambda}^{(L)}$ where $\tilde{\Lambda}^{(L)}$ is the second-largest eigenvalue of the submatrix containing the largest eigenvalue $\Lambda_1^{(L)}$.

[17]M. P. Nightingale, Physica (Utrecht) A 83, 561 (1976).

[18]Earlier works are cited in Ref. 14.

[19]M. P. Nightingale, Phys. Lett. A 59, 486 (1977).

[20]K. G. Wilson and J. Kogut, Phys. Rep. C 12, 76 (1974).

[21]R. H. Fowler and G. S. Rushbrooke, Trans. Faraday Soc. 33, 1272 (1937).

[22]C. Domb, Adv. Phys. 9, 149 (1960).

[23]J. M. J. van Leeuwen, Phys. Rev. Lett. 34, 1056 (1975).

[24]D. S. Gaunt, J. Chem. Phys. 46, 3237 (1967).

[25]S. Alexander, Phys. Lett. A 54, 353 (1975).

[26]B. D. Metcalf, Phys. Lett. A 45, 1 (1973).

[27]M. Schick, J. S. Walker, and M. Wortis, Phys. Rev. B 16, 2205 (1977).

[28]B. D. Metcalf and C. P. Yang, Phys. Rev. B 18, 2304 (1978).

[29]More sophisticated mean-field approximations [D. M. Burley, Physica (Utrecht) 27, 768 (1961)], yield finite and lattice-dependent $a^*$ and $m_c < 1$. Since mean-field theories overestimate the ordering tendencies, the results for $a^*$ are always larger than the "exact" series-expansion estimates.