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Exact Analysis of a Lattice Gas on the 3-12 Lattice with Two- and Three-Site Interactions

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Exact results are obtained for a lattice gas on the 3-12 lattice with two- and three-site interactions. By using a decoration transformation, we map the lattice gas into one on the honeycomb lattice with pure two-site interactions. This procedure permits us to draw exact results for the original 3-12 lattice gas. In particular, we obtain its exact two-phase boundary, and confirm the fact that an experimentally observed anomaly in the critical behavior of the coexistence-curve diameter is present if, and only if, the three-site interactions are present.

KEY WORDS: Lattice gas; three-site interactions; two-phase region; coexistence-curve diameter.

1. INTRODUCTION

In 1952 Lee and Yang[(1)] formulated the consideration of a lattice gas as a realization of liquid–gas transitions. This seemingly artificial formulation of a real gas has, however, proven to be extremely useful in explaining experimental observations. In a recent paper Goldstein *et al.*[(2)] suggested that the experimentally observed anomaly of the critical behavior of the coexistence-curve diameter in real gases can be explained by the existence of three-site interactions. This suggestion has subsequently been verified by Wu and Wu,[(3)] who showed rigorously that the introduction of three-site interactions in a Kagomé lattice gas indeed induces the observed anomaly. But in the Kagomé lattice the two- and three-site interactions are not "separable" in the sense that they involve the same cluster of spins. It is

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therefore desirable to further test the relevance of three-site interactions on the observed anomaly in systems with completely separable two- and three-site interactions. In this paper we take up this consideration.

We consider the lattice gas on the 3-12 lattice shown in Fig. 1, where J and J' are the (reduced) nearest-neighbor interactions, and J_3 the (reduced) three-site interactions among three spins surrounding a triangle. Note that the J' interactions are between sites belonging to different triangles, and are therefore completely separated from the three-site interactions J_3. The grand partition function of this lattice gas is

$$\Xi(z, J, J', J_3) = \sum_{n_i=0,1} \left[\prod_i z^{n_i}\right]\left[\prod e^{Jn_in_j}\right]\left[\prod e^{J'n_in_j}\right]\left[\prod_\Delta e^{J_3n_in_jn_k}\right] \tag{1}$$

where the second and the third products are taken over nearest neighboring sites, and the last product is over all triangular faces. The pressure of this lattice gas is

$$p = \frac{kT}{N} \ln \Xi(z, J, J', J_3) \tag{2}$$

where N is the total number of sites and the fugacity z is to be eliminated by fixing the density at

$$\rho = z\left(\frac{\partial}{\partial z}\frac{p}{kT}\right)_{J,J',J_3} \tag{3}$$

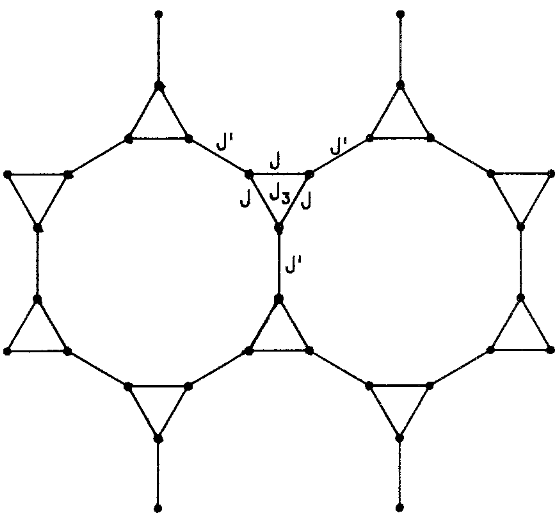

Fig. 1. The 3-12 lattice.

2. EQUIVALENCE WITH AN EIGHT-VERTEX MODEL

Our analysis is based on a decoration transformation, which maps the 3-12 lattice gas into one on the honeycomb lattice with pure two-site interactions. Our first step is to relate the lattice gas in (1) to an eight-vertex model on the underlying honeycomb lattice. First, to each J' interaction we introduce a decorating site as shown in Fig. 2. Writing the equivalence of Boltzmann factors

$$z^{n_1+n_2}e^{J'n_1n_2}=F\sum_{n=0,1} y^n e^{Rn(n_1+n_2)} \tag{4}$$

where y is the fugacity of the decorating site, and $n_1, n_2 = 0, 1$ are the occupation numbers of the end sites, we obtain

$$\begin{aligned} e^R &= z(1-zu)/(1-z) \\ y &= (1-z)^2/z^2(u'-1) \\ F &= z^2(u'-1)/(1-2z+z^2u) \end{aligned} \tag{5}$$

where here, and in (8) below, we introduce the notation

$$u=e^J, \qquad u'=e^{J'}, \qquad v=e^{J_3} \tag{6}$$

The introduction of the decorating sites permits us to group the Boltzmann factor in (1) by elementary triangles. Thus, we can write

$$\Xi(z, J, J', J_3)=F^{N/2}\sum_{n_i=0,1}\prod_{\Delta}\omega(n_1, n_2, n_3) \tag{7}$$

where the product is taken over all triangles, and

$$\begin{aligned}\omega(n_1, n_2, n_3) = y^{(n_1+n_2+n_3)/2}[&1+e^{Rn_1}+e^{Rn_2}+e^{Rn_3}+ue^{R(n_1+n_2)} \\ &+ue^{R(n_2+n_3)}+ue^{R(n_3+n_2)}+u^3ve^{R(n_1+n_2+n_3)}]\end{aligned} \tag{8}$$

is the Boltzmann factor of a unit cell shown in Fig. 3. Now the Boltzmann factor (8) can be regarded as the vertex weights of an eight-vertex model on the underlying honeycomb lattice[4,5] with vertex weights

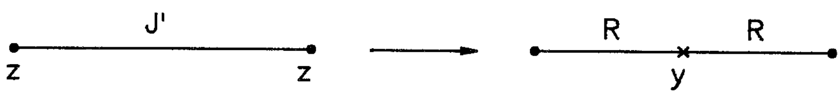

Fig. 2. The introduction of a decorating site with fugacity y to the J' interaction.

$$
\begin{aligned}
a &= \omega(0, 0, 0) = 4 + 3u + u^3 v \\
b &= \omega(1, 0, 0) = \sqrt{y}\,[3 + u + (1 + 2u + u^3 v)\, e^R] \\
c &= \omega(1, 1, 0) = y[2 + 2(1 + u)\, e^R + u(1 + u^2 v)\, e^{2R}] \\
d &= \omega(1, 1, 1) = y^{3/2}[1 + 3e^R + 3ue^{2R} + u^3 v e^{3R}]
\end{aligned} \tag{9}
$$

This leads to the identity

$$\Xi(z, J, J', J_3) = F^{N/2} Z_{8v}(a, b, c, d) \tag{10}$$

where $Z_{8v}(a, b, c, d)$ is the eight-vertex model partition function.

3. EQUIVALENCE WITH AN ISING MODEL

It has been established[4,5] that the eight-vertex model partition function $Z_{8v}(a, b, c, d)$ is further equivalent to that of an Ising model on the honeycomb lattice with reduced nearest-neighbor interactions K and an external field L. For completeness, we quote here all relevant expressions for this equivalence as needed in our calculations:

$$Z_{8v}(a, b, c, d) = \left(\frac{\tilde{a}}{2\cosh L}\right)^{N/3} (\cosh K)^{-N/2}\, Z^{\mathrm{HC}}_{\mathrm{Ising}}(K, L) \tag{11}$$

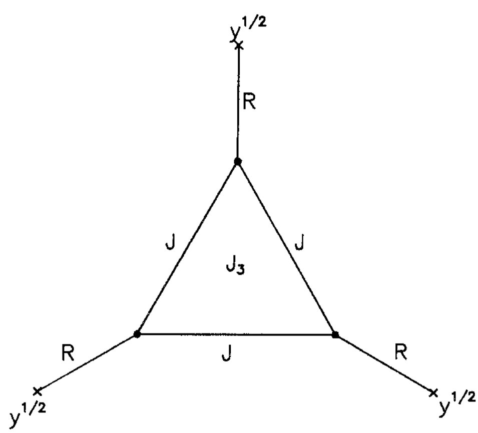

Fig. 3. A unit cell of the decorated 3-12 lattice.

where $Z^{\mathrm{HC}}_{\mathrm{Ising}}(K, L)$ is the Ising partition function, and

$$\begin{aligned}
\tilde{a} &= (a + 3by_0 + 3cy_0^2 + dy_0^3)/(1 + y_0^2)^{3/2} \\
y_0 &= (\delta - A - C)/B \\
e^{4K} &= [\delta/(A - C)]^2 \\
\tanh L &= \frac{V}{U}\left[\frac{\delta - (A - C)}{\delta + A - C}\right]^{1/2} \\
U &= (b + d)\, y_0 + a + c \\
V &= (a + c)\, y_0 - (b + d) \\
A &= c^2 - bd, \qquad B = ad - bc \\
C &= ac - b^2, \qquad \delta = [(A - C)^2 + B^2 + 4AC]^{1/2}
\end{aligned} \tag{12}$$

Using (9) and (5) and after some algebra, we obtain the following explicit expressions:

$$\begin{aligned}
C - A &= t^3(u^3u'vz^2 - u^2u'z^2 + u^3vz - uz + u - 1)/z^4(u' - 1)^2 \\
B^2 + 4AC &= t^6u^2(u^4v^2 - 6u^2v + 4uv + 4u - 3)/z^6(u' - 1)^3 \\
t &= u'z^2 - 2z + 1
\end{aligned} \tag{13}$$

The substition of (11) into (10) and (2) now yields

$$\frac{p}{kT} = \frac{1}{3}\ln\left(\frac{\tilde{a}}{2\cosh L}\right) + \frac{1}{2}\ln\left(\frac{F}{\cosh K}\right) + \frac{1}{3} f_{\mathrm{HC}}(K, L) \tag{14}$$

where $f_{\mathrm{HC}}(K, L)$ is the per-site "free energy" of the honeycomb Ising model. Here the factor 1/3 in the last term of (14) takes into account that the honeycomb lattice has $N/3$ lattice point.

We shall need to locate the locus $L = 0$ in the ensuing discussions. From the expressions of K and L in (12), we see that $L = 0$ must occur at $V = 0$. Using (9) and (5) we find[3] after some reduction both V and U containing a factor t^2. As a result, $L = 0$ must occur at the locus obtained by setting the remaining factor in V aqual to zero. After some lengthy algebra, this leads to[4]

[3] In addition, we also find $B \sim t^2$, $a + c \sim t$, $b + d \sim t$, $(a + c)(A + C) + (b + d)\, B \sim t^4$, $(b + d)(A + C) - (a + c)\, B \sim t^4$.

[4] If (31) of ref. 7 for $L = 0$ is used in place of $V/U = 0$, one finds (as a result of the quoted t dependences) the spurious solution $t^6 = 0$, in addition to (15).

$$u^6u'^3v(u^3v^2-3uv+2)\,z^6+6u^4u'^2(u^3v^2-u^2v^2-uv+1)\,z^5$$
$$+3u^3u'(u^2u'v-2uu'v+4u^2v-8uv+u'+4)\,z^4$$
$$+4u^3(-3u'v-2v+3u'+2)\,z^3$$
$$+3u(-u^2u'v-4u^2v+2uu'-u'+8u-4)\,z^2$$
$$+6(-u^3v+u^2+u-1)\,z$$
$$-(u^3v-3u+2)=0 \qquad (L=0) \tag{15}$$

4. TWO-PHASE REGION AND COEXISTENT CURVE DIAMETER

The equation of state of the lattica gas (1) is now given by (14) with the fugacity z contained therein eliminated by using (3). However, the Ising free energy $f_{\mathrm{HC}}(K, L)$ is known[6] only for $L=0$, which lies on the boundary of the two-phase region. Explicitly, this boundary is given by

$$\frac{p}{kT}=\frac{1}{3}\ln\left(\frac{\tilde{a}}{2}\right)+\frac{1}{2}\ln\left(\frac{F}{\cosh K}\right)+\frac{1}{3}f_{\mathrm{HC}}(K,0) \tag{16}$$

at the two densities

$$\rho_{\pm}=\rho_d\pm\frac{z}{3}\left(\frac{\partial L}{\partial z}\right)I_0(K) \tag{17}$$

Here,

$$f_{\mathrm{HC}}(K,0)=\frac{1}{8\pi}\int_0^{2\pi}d\theta\,\ln[\alpha+(\alpha^2-2\cos\theta-2)^{1/2}]$$
$$+\frac{1}{2}\ln(2\alpha+2\cos\theta) \tag{18}$$

where $\alpha=-\cos\theta+(1+\cosh^3 2K)/\sinh^2 2K$,

$$\rho_d=z\frac{\partial}{\partial z}\left(\frac{1}{3}\ln\tilde{a}+\frac{1}{2}\ln F\right)+z\left(\frac{\partial K}{\partial z}\right)\left[-\frac{1}{2}\tanh K+\frac{1}{3}\frac{\partial}{\partial K}f_{\mathrm{HC}}(K,0)\right] \tag{19}$$

is the coexistence-curve diameter, and

$$I_0(K)=\left(1-\frac{16x^3(1+x^3)}{(1-x)^3\,(1-x^2)^3}\right)^{1/8} \tag{20}$$

where $x=e^{-2K}$ is the spontaneous magnetization[7] of the honeycomb Ising lattice. In both (16) and (17), the fugacity z is to be determined from

(15). The critical regime of the lattice gas now corresponds to the regime $K > K_c$ or $e^{2K} > e^{2K_c} = 2 + \sqrt{3}$, where K_c is the critical point of the honeycomb Ising model satisfying $I_0(K_c) = 0$.

The procedure of computing the boundary of the two-phase region is as follows: For fixed J, J', J_3, we solve z from (15). Then, the substitution of this z into (16) and (17) yields two points $\{p, \rho_\pm\}$ on the phase boundary. We have carried out this calculation for $J = J'$. The result is shown in Fig. 4. Numerically, we have found that the critical regime occurs only when $J_3 > 0$, $J > 0$, and $J' > 0$. For $J_3 > 0$, $J < 0$, and $J' < 0$, for example, it can be readily seen from (12) and (13) that we have always $K < 0$. Our numerical finding is consistent with the fact that $K < K_c$ if any of J, J', J_3 becomes negative.

The expression (19) gives rise to an exact expression of the coexistence curve diameter ρ_d. According to the law of *rectilinear diameter*,[8] ρ_d increases from its critical value ρ_c linearly as the temperature decreases from the critical temperature T_c. However, Goldstein *et al.*[2] presented experimental evidence in real gases of an anomalous behavior of $(T_c - T)^{1-\alpha}$, where α is the specific heat exponent, and argued that this anomalous behavior can be explained by the existence of three-site interactions. In the present case the exact critical behavior of ρ_d of the lattice gas (1) can be examined. The rhs of (19) contains four terms; the first three of which are analytic functions in T, which, indeed, gives rise to the rectilinear critical behavior. The last term in (19), however, possesses a

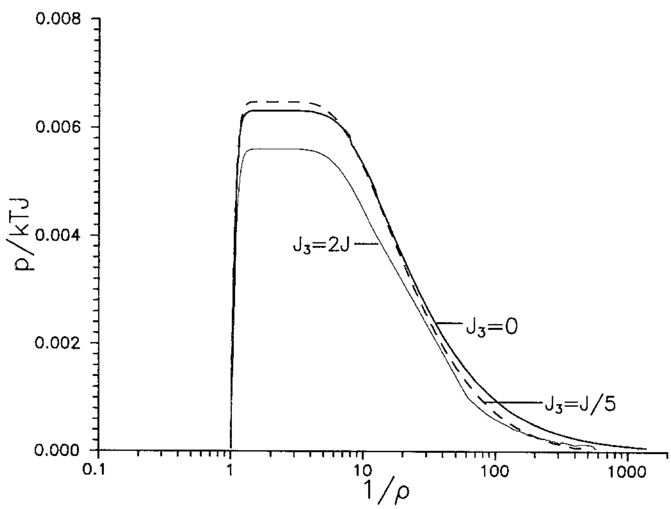

Fig. 4. The exact boundary of the two-phase region of the lattice gas (1) ($J = J'$).

critical behavior $(T_c - T)^{1-\alpha'}$ with an amplitude proportional to $\partial K/\partial z$, which after some algebra becomes

$$\frac{\partial K}{\partial z} = u^2(1-u')[u^2u'(uv-1)z^2 - (u-1)]\frac{u^4v^2 - 6u^2v + 4uv + 4u - 3}{P} \tag{21}$$

where P is a polynomial in u, u', v, and z whose explicit form does not affect us and therefore is not given. In the most general case $\partial K/\partial z$ given by (21) does not vanish when substituted with the value of z solved from (15), and hence the critical behavior of ρ_d is $(T_c - T)^{1-\alpha'}$, as observed experimentally. When $J_3 = 0$ ($v = 1$), we find (15) factorized into

$$\begin{aligned}(u^2u'z^2 - 1)(u-1)^2\,[u^4u'^2(u+2)\,z^4 + 6u^2u'(u+1)\,z^3 \\ + u(u^2u' + 2uu' + 3u' + 12)\,z^2 + 6(1+u)\,z + u + 2] = 0\end{aligned} \tag{22}$$

for which the only solution is $z = 1/u\sqrt{u'}$. At this value of z the factor inside the square brackets in (21), hence $\partial K/\partial z$, vanishes identically (using $v = 1$). As a result, the anomalous critical behavior $(T_c - T)^{1-\alpha'}$ is absent. Thus, we have established that, for the lattica gas (1), the observed anomalous critical behavior $(T_c - T)^{1-\alpha'}$ is present if, and only if, the three-site interaction J_3 is present.

It should be noted that for two-dimensional models the anomalous critical behavior is, in fact, $(T_c - T)\ln|T_c - T|$. However, our main conclusion on the relevance of the anomaly of the critical behavior with the presence of three-site interactions is also valid in three dimensions. In three dimensions one considers an "extended" hyper-Kagomé lattice derived by decorating a three-coordinated hydrogen peroxide lattice,[9] much in the same fashion as the 3-12 lattice is derived from the honeycomb lattice. Again, using the consideration of an eight-vertex model, one establishes our main conclusion for a lattice gas with three-site interactions on the extended hyper-Kagomé lattice. In this case the anomalous critical behavior is $(T_c - T)^{1-\alpha'}$, where α' is the specific heat exponent of the three-dimensional Ising model.

Finally, we remark that the method of an effective decimation of three-site interactions such as that employed here has proven to be an extremely useful tool, and has very recently been used to establish the existence of an asymmetry in the two-phase coexistence surface of a ternary solution of molecules with three-body interactions.[10]

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