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Correlation functions in decorated lattice models

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Abstract

Occupation probabilities for primary–secondary–primary cell strings and correlation functions for primary sites of a decorated lattice model are expressed through the well-studied partition function and correlation functions of the Ising model. The results are analogous to those found in related lattice models of hydrophobic interactions and are interpreted in similar terms.

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1. Introduction

Decorated-lattice-gas models that are equivalent to an underlying Ising model have been important as models of two-component mixtures with closed-loop solubility curves [1–3]. In summing over the states of the decoration sites in the partition function one obtains an Ising model in which the energy and field parameters are related to the parameters of the original mixture model by certain rules (transcription relations). The interactions in the Ising model or in the equivalent one-component lattice gas may then be understood to have arisen, or to have been altered, through the mediation of the particles occupying the decorated sites (cells). In particular, the correlation functions between the particles occupying the primary cells, which would be those of the underlying Ising model or one-component lattice gas, may be understood to have resulted from such mediation. This is then analogous to the circumstance in recent lattice models of hydrophobic interactions [4–6], in which the solvent-mediated potential of mean force between solute molecules is obtained in terms of the correlation functions of the

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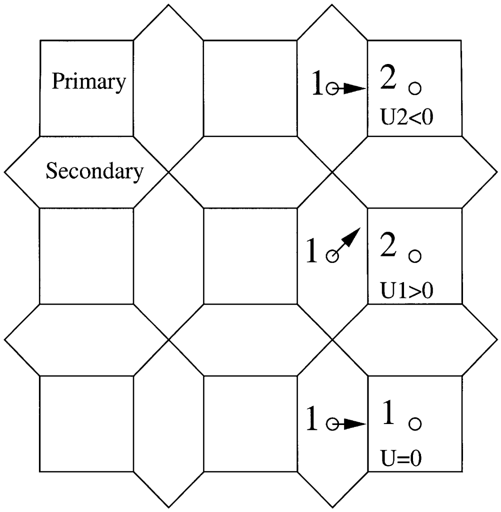

Fig. 1. Mermin's decorated lattice model.

pure solvent. In the present work the potentials of mean force between occupants of the primary cells in a class of decorated-lattice-gas models with closed-loop solubility curves [1–3] are calculated and interpreted in terms similar to those in the lattice models of hydrophobic interaction [4–6].

The model and the calculational machinery are outlined in the next section and the results are then illustrated by numerical examples in Section 3. In Section 4 the correlation lengths, which are the exponential decay lengths of the various correlation functions, are calculated from those of the underlying Ising model and are displayed numerically.

2. Decorated-lattice-gas model

We start with a brief review of a version of *Mermin's decorated lattice model* [1], Fig. 1, that describes a liquid mixture that possesses upper and lower critical solution points and a closed-loop temperature–concentration coexistence curve [2,3]. Each cell, primary or secondary, is occupied by a molecule, either of type 1 or 2. Each molecule of either type has ω possible orientations. The only interaction takes place between the molecules in adjacent primary–secondary cells. This interaction is defined to be 0 unless adjacent primary–secondary cells are occupied by molecules of different types, and its value then depends only on the orientation of the molecule in the secondary cell. For each primary–secondary neighboring pair, if the occupant of the secondary cell points to the primary cell occupied by an unlike molecule, the energy of interaction is $U_2 < 0$; if it points in any other of $\omega - 1$ directions, the energy is $U_1 > 0$.

To calculate the partition function for the whole system we first write the partition functions Q_{ijk} for all *primary–secondary–primary cell* strings for fixed occupations

i and k of primary cells,

$$\begin{aligned} &Q_{111} = Q_{222} = \omega\,, \\ &Q_{121} = Q_{212} = 2\mathrm{e}^{-(U_1+U_2)/kT} + (\omega-2)\mathrm{e}^{-2U_1/kT}\,, \\ &Q_{122} = Q_{211} = Q_{112} = Q_{221} = \mathrm{e}^{-U_2/kT} + (\omega-1)\mathrm{e}^{-U_1/kT}\,. \end{aligned} \tag{1}$$

We also introduce the activity ratio $\zeta = z_1/z_2$

$$\zeta = \mathrm{e}^{(\mu_1-\mu_2)/kT}\left[\frac{m_1}{m_2}\right]^{d/2}\,, \tag{2}$$

where m_i and μ_i are molecular masses and chemical potentials for each species and d is the dimensionality of space.

As in Eq. (62) of Ref. [2], we express the partition function of the model for the fixed number N of primary cells as

$$Y = \omega^N \sum_{\{n_i\}} \zeta^{N_1}(Q_{111}\zeta + Q_{121})^{N_{11}}(Q_{212}\zeta + Q_{222})^{N_{22}}(Q_{112}\zeta + Q_{122})^{N_{12}}\,. \tag{3}$$

Here N_i is the number of molecules of component i, N_{ij} is the number of i,j pairs of neighboring primary cells, and the sum is over all possible occupations "$\{n_i\}$" of the primary cells.

Now we can evaluate the probability P_{ijk} to find a primary–secondary–primary cell string being occupied by the molecules i, j, and k. It can be written as

$$\begin{aligned} P_{ijk} = {}& \frac{2}{qN}\frac{1}{Y}\omega^N \sum_{\{n_i\}} \zeta^{N_1} \frac{Q_{ijk}[1+\delta_{1j}(\zeta-1)]}{\sum_{n=1}^{2} Q_{ink}[1+\delta_{1j}(\zeta-1)]} N_{ik} \\ &\times (Q_{111}\zeta + Q_{121})^{N_{11}}(Q_{212}\zeta + Q_{222})^{N_{22}}(Q_{112}\zeta + Q_{122})^{N_{12}}\,. \end{aligned} \tag{4}$$

Here $qN/2$ is the total number of bonds between N primary cells of the lattice with coordination number q. If we multiply Q_{ijk} by an auxiliary factor α_{ijk}, so that Q_{ijk} is replaced by $Q_{ijk}\alpha_{ijk}$, the probability P_{ijk} can be expressed as

$$P_{ijk} = \frac{2}{qN}\left.\frac{\partial \ln Y}{\partial \alpha_{ijk}}\right|_{\alpha_{ijk}=0}\,. \tag{5}$$

We also can calculate the number of occupants of secondary cells pointing towards the opposite species occupants of primary cells, N_p. This quantity is a measure of orientational ordering of the system. Note that the orientations of molecules in primary cells do not enter the expression for the partition function and therefore are isotropic everywhere in the phase diagram. Since every secondary–primary cell neighboring pair where the cells are occupied by opposite species and secondary cell occupants point to the primary cell comes with a factor $\mathrm{e}^{-U_2/kT}$, there is no need to introduce any auxiliary multipliers:

$$N_p = -\frac{1}{kT}\frac{\partial \ln Y}{\partial U_2}\,. \tag{6}$$

The structure of this expression is similar to that of (4); it gives the probability of finding a particular ordered primary–secondary–primary string multiplied by the average number of such strings.

To get the corresponding intensive quantity n_p, we need to divide N_p by the number of primary–secondary cell neighboring pairs qN (twice the number of primary–primary neighboring pairs), $n_p = N_p/qN$.

Another important class of correlation functions it is possible to calculate is a connected pair and higher-order correlation functions for the primary cells. To underline the connection to the Ising model, let us introduce the primary cell occupation numbers, $n_j = +1$ for the jth cell being occupied by a molecule of species 1 and $n_j = -1$ if it is occupied by a molecule of species 2. Then N_1 and N_2 in Eq. (3) can be expressed as $N_1 = (N + \sum_{j=1}^{N} n_j)/2$ and $N_2 = (N - \sum_{j=1}^{N} n_j)/2$. We also introduce an auxiliary local field γ_j, in the presence of which the partition function takes the form

$$Y = \omega^N \zeta^{N/2} \sum_{\{n_i\}} e^{(1/2)\sum_{i=1}^{N} n_i(\gamma_i + (1/2)\ln\zeta)} A_{11}^{N_{11}} A_{22}^{N_{22}} A_{12}^{N_{12}} . \tag{7}$$

Here we used the shorthand notations: $A_{11} \equiv Q_{111}\zeta + Q_{121}$, $A_{22} \equiv Q_{212}\zeta + Q_{222}$, $A_{12} \equiv Q_{112}\zeta + Q_{122}$.

As in the Ising model, the two-body connected correlation function for the primary cells i and j, $C_{i,j} \equiv \langle n_i n_j \rangle - \langle n_i \rangle \langle n_j \rangle$, can be expressed as

$$C_{i,j} = \left. \frac{\partial^2 \ln Y}{\partial\gamma_i \partial\gamma_j} \right|_{\{\gamma_k\}=0} . \tag{8}$$

In order to complete the calculation of correlation functions P_{ijk} and C_{ij}, we express the partition function $Y(\{\gamma_i\})$ through the known partition and correlation functions for the Ising model. Using lattice identities, $qN_1 = 2N_{11} + N_{12}$ and $qN_2 = 2N_{22} + N_{12}$, we reduce Eq. (7) to

$$Y = \omega^N \zeta^{N/2} (A_{11}A_{22})^{qN/4} \sum_{\{n_i\}} e^{(1/2)\sum_{i=1}^{N} n_i(\gamma_i + (1/2)\ln\zeta + (q/4)\ln(A_{11}/A_{22}))} \left[\frac{A_{12}}{\sqrt{A_{11}A_{22}}} \right]^{N_{12}} . \tag{9}$$

On the other hand, for the Ising model with the Hamiltonian

$$\mathscr{H} = -J \sum_{\langle i,j \rangle} s_i s_j - \sum_i s_i(H + \gamma_i) , \tag{10}$$

the partition function Z and the connected two-point correlation function $C_{i,j}^{Ising}$ can be expressed as

$$Z = e^{-qN/2} \sum_{\{s_i\}} e^{(1/kT)\sum_{i=1}^{N} n_i(H+\gamma_i) + 2JN_{+-}} , \tag{11}$$

$$C_{i,j}^{Ising} = \left. \frac{\partial^2 \ln Z}{\partial\gamma_i \partial\gamma_j} \right|_{\{\gamma_k\}=0} . \tag{12}$$

Here N_{+-} is the number of bonds between up and down spins, which is equivalent in the mixture model to the number of 1–2 neighboring pairs of primary cells, $N_{+-} = N_{12}$.

We observe that the connected correlation function for the decorated lattice model and Ising model are identical if the coupling constant J and external field H in the Ising model are defined as

$$C_{i,j} = C_{i,j}^{Ising} ,$$
$$\frac{J}{kT} \langle = \rangle -\frac{1}{2} \ln \left[\frac{A_{12}}{\sqrt{A_{11}A_{22}}} \right] , \quad \frac{H}{kT} \langle = \rangle \frac{1}{2} \ln \zeta + \frac{q}{4} \ln \frac{A_{11}}{A_{22}} . \tag{13}$$

We next proceed to the calculation of the k-point potential of mean force between the occupants of the primary cells, $W_k(\{i \dots j\}) = -kT \ln [\langle \rho_i \dots \rho_j \rangle / \langle \rho \rangle^k]$. Since the effective interaction between occupants of primary cells depends on the species and state of the occupants of the intervening secondary cells, the corresponding primary–primary cell correlation functions may be viewed as having been mediated by those secondary-cell occupants. This is analogous to the solvent-mediated correlations between solute molecules in related hydrophobic-interaction models [4–6], but it is to be understood that in the present model the effective correlations are not those between molecules of the same species since molecules of both species may be the occupants of either kind of site. Indeed, the correlations calculated here are those between the occupants (whatever their species) of the same kind of site (the primary cells).

In the definition of W, ρ_i is the non-negative occupation number (density) of the ith cell. Since the actual amplitude of ρ in the definition of W cancels, it is convenient to let it vary between 0 and 1, i.e., to define it as $\rho_i \equiv (n_i + 1)/2$. It allows a k-point potential of mean force to be expressed through the k- and lower-order connected correlation functions in the Ising model. For example, for $k = 2$,

$$W_2(\{i,j\}) = -kT \ln \left[\frac{C_{i,j}}{(\langle n \rangle + 1)^2} + 1 \right] . \tag{14}$$

Here $\langle n \rangle = \partial \ln Z / \partial H$ is the average occupation number or magnetization for the corresponding Ising model. The correspondence between the parameters of the decorated lattice model and Ising model is given by (13). Similarly, one can calculate the higher-than-second-order correlation functions and potentials of mean force.

Finally, we show how to calculate the primary–secondary cell and secondary–secondary cell correlation functions and potentials of mean force. Taking into account Eqs. (1), we calculate a probability $S_{\{i1j\}}$ to find a particle of species 1 in the secondary cell situated between primary cells occupied by molecules of species i and j:

$$S_{\{111\}} = \frac{\omega}{\omega + 2\mathrm{e}^{-(U_1+U_2)/kT} + (\omega - 2)\mathrm{e}^{-2U_1/kT}} ,$$
$$S_{\{112\}} = S_{\{122\}} = \frac{1}{2} ,$$
$$S_{\{212\}} = \frac{2\mathrm{e}^{-(U_1+U_2)/kT} + (\omega - 2)\mathrm{e}^{-2U_1/kT}}{\omega + 2\mathrm{e}^{-(U_1+U_2)/kT} + (\omega - 2)\mathrm{e}^{-2U_1/kT}} . \tag{15}$$

Taking into account the definition of ρ for primary cells given above, we express an average density of molecules of the species 1 in secondary cells, $\rho^\dagger$, as

$$\rho^\dagger = \langle \rho_i \rho_j \rangle S_{\{111\}} + 2 \langle (1 - \rho_i) \rho_j \rangle S_{\{211\}} + \langle (1 - \rho_i)(1 - \rho_j) \rangle S_{\{212\}} . \tag{16}$$

Here ρ_i and ρ_j are the occupation numbers of the neighboring primary cells i and j. Similarly, the primary–secondary-cell correlation function $\langle \rho_k \rho_l^\dagger \rangle$ is expressed through the following average:

$$\langle \rho_k \rho_l^\dagger \rangle = \langle \rho_k \rho_i \rho_j \rangle S_{\{111\}} + \langle \rho_k (1-\rho_i) \rho_j \rangle S_{\{211\}} + \langle \rho_k \rho_i (1-\rho_j) \rangle S_{\{112\}} + \langle \rho_k (1-\rho_i)(1-\rho_j) \rangle S_{\{212\}} \,. \tag{17}$$

Here i and j are the primary cells between which the secondary cell l is situated. It follows that the primary–secondary cell correlation function of the decorated lattice model can be expressed through the energy-magnetization (3-point) correlation function of the underlying Ising model and various 2-point correlation functions. Finally, for the primary–secondary cell potential of mean force $\tilde{W}_2(\{k,l\})$, one obtains

$$\tilde{W}_2(\{k,l\}) = -kT \ln \frac{\langle \rho_k \rho_l^\dagger \rangle}{\langle \rho \rangle \langle \rho^\dagger \rangle} \,. \tag{18}$$

A similar approach shows that the secondary–secondary cell correlation function and potential of mean force for the decorated lattice model can be expressed through energy–energy (4-point), energy–magnetization (3-point) and 2-point correlation functions.

Because of the generic nature of Mermin's decorated lattice model, these results are applicable for lattices in arbitrary dimension d with various coordination numbers q.

3. Examples

Using the formal mapping of the correlation functions of the decorated lattice model onto the correlation functions of an Ising spin system, we perform computations of the two- and three-body potential of mean force in two dimensions. The following values were chosen for the model parameters: $U_1 = -U_2$ (with U_1, unspecified, then a scale factor for the temperature), $\omega = 100$, $\zeta = 1$. The phase diagram for these values of the parameters is shown in Fig. 2.

Despite the fact that the correlation functions for the Ising model are available analytically in the form of series expansions (see, for example, Ref. [7]), we found it more convenient to generate them each time directly using Monte Carlo simulations. We used Metropolis and Wolff algorithms (see, for example, Ref. [8]) [1] with the lattice size varying from 100×100 to 200×200. All the results presented below were averaged over the whole system and over 200 configurations; configuration were considered different if they were separated by about 10 flips for each spin in the Wolff algorithm. The correlation functions and potentials of mean force were measured along the lattice ("normal") unit vectors ($(1,0)$ and $(0,1)$) and along the main diagonal $(1,1)$. We found that in general the results for "normal" and "diagonal" measurements are very similar. An example is in Fig. 3, where the two-body potential of mean force is shown.

[1] We believe that there is a typographical error in Eqs. (4.53)–(4.54): a coefficient in front of βJ in the exponentials should be 2 instead of 4.

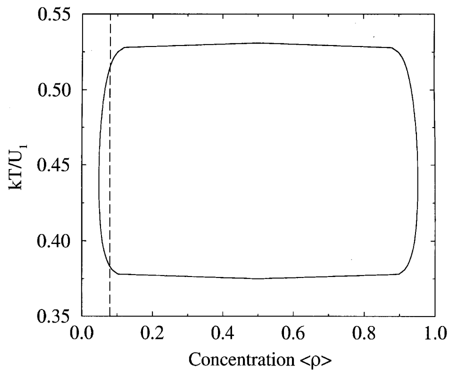

Fig. 2. Closed loop temperature–concentration solubility curve. The dashed line marks the concentration $\langle\rho\rangle \approx 0.08$.

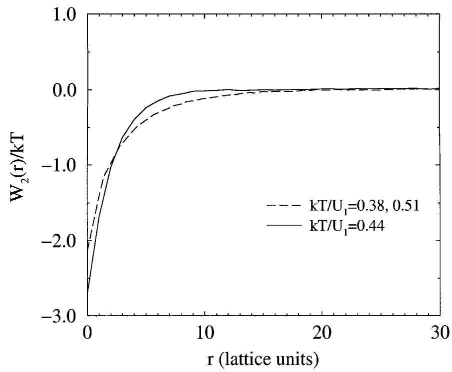

Fig. 3. Plots of the two-body potential of mean force at $kT/U_1 = 0.44$ (solid line) and $kT/U_1 = 0.38, 0.51$ (dashed line).

The dashed curve is a result of the superposition of normal and diagonal plots, with normal and diagonal points occurring at integer and $\sqrt{2} \times$ integer lattice separation.

The potentials of mean force W_2 and W_3 were calculated for the temperatures corresponding to the widest point of the phase diagram ($\langle\rho\rangle \approx 0.05$, $kT/U_1 \approx 0.44$), and also for the two temperatures corresponding to the intersections of the dashed line and the coexistence curve in Fig. 2, at both of which there is the same minority species concentration $\langle\rho\rangle \approx 0.08$ ($kT/U_1 \approx 0.384$ and $kT/U_1 \approx 0.513$). The effective Ising coupling constant is $J/kT = 0.47$ for the first point and $J/kT = 0.45$ for the next two.

The potentials of mean force for primary–secondary cell occupants is shown in Fig. 4. The original primary cell and two primary cells that surround the secondary cell lie on the same lattice vector; the distance between primary and secondary cell is assumed to be equal to half-sum of the distances between the original and two neighboring primary cells.

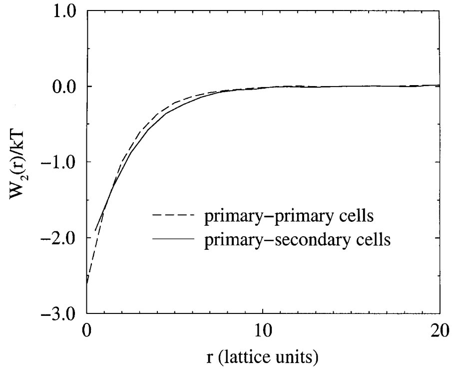

Fig. 4. Plots of the two-body potential of mean force at $kT/U_1 = 0.44$ for primary–secondary cells (solid line) and primary–primary cells (dashed line).

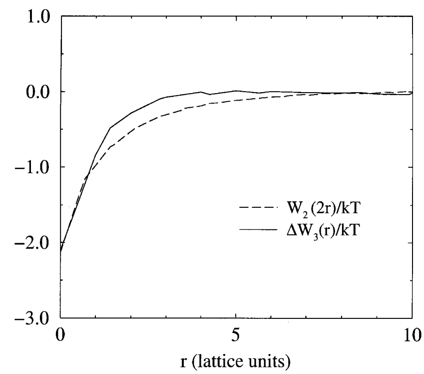

Fig. 5. Difference between the sum of the three pair potentials and the true three-body interaction, $\Delta W_3(r) \equiv 2W_2(r) + W_2(2r) - W_3(r,r,2r)$ (solid line), and the pair potential $W_2(2r)$ (dashed line), for the linear configuration of points and the temperatures $kT/U_1 = 0.384$ or 0.513.

For the three-body potential, we chose the two simplest configurations: In the first one, particles 1, 2, and 3 lie on a straight line with the distance r between particles 1 and 2 being equal to the distance between particles 2 and 3. In the second configuration particles 1, 2, and 3 form a right triangle in which the legs 1,2 and 1,3 are of equal length r. We are mainly interested in the question of how well the effective three-body interaction is approximated by the sum of the three pair interactions.

In Fig. 5 we present a plot of the difference between the sum of the three pair potentials and the true three-body potential for a linear configuration, $\Delta W_3(r) \equiv 2W_2(r) + W_2(2r) - W_3(r,r,2r)$. Since all potentials W_2 and W_3 are negative for any r, one can observe that the sum of the three pair interactions clearly overestimates (the absolute magnitude of) the true three-body potential. However, unlike in some one-dimensional models of hydrophobic interactions [6], this overestimate is less (in absolute value) than the interaction between the furthermost particles, 1 and 3. To

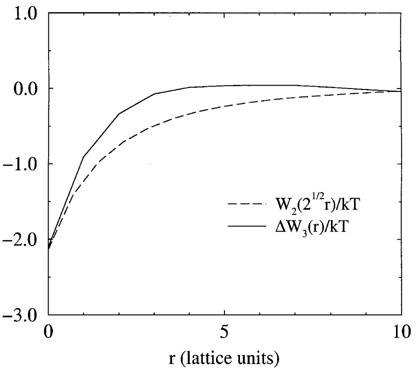

Fig. 6. Difference between the sum of the three pair potentials and the true three-body interaction, $\Delta W_3(r) \equiv 2W_2(r) + W_2(\sqrt{2}r) - W_3(r, r, \sqrt{2}r)$ (solid line), and the pair potential $W_2(\sqrt{2}r)$ (dashed line), for the triangular configuration of points and the temperatures $kT/U_1 = 0.384$ or 0.513.

show that, a plot of the corresponding two-body potential $W_2(2r)$ is presented in the same Fig. 5.

Similar results for the triangular configuration of points are presented in Fig. 6. Here again, as in the linear case, the sum of all pairwise interactions overestimates the true three-body potential, but, as in the linear case, by less than the magnitude of the pairwise potential between two most remote points, $W_2(\sqrt{2}r)$.

4. Correlation length

In this section the correlation lengths for the decorated lattice model are calculated from those of the underlying Ising model. Before we proceed to the calculation, we note that the correlation lengths for any pair of species (i.e., 1–1, 1–2, 2–2) are all identical in this model. The exponential range of correlation for a pair of molecules of species α and β occupying the primary cells is defined by

$$1/\xi_{\alpha\beta} = -\lim_{|\boldsymbol{R}|\to\infty} |\boldsymbol{R}|^{-1} \ln |h_{\alpha\beta}(\boldsymbol{R})| \,, \tag{19}$$

where $h_{\alpha\beta}(\boldsymbol{R})$ is the pair correlation function for species α and β. However, all the pair correlation functions h_{11},h_{12}, and h_{22} are expressed as

$$h_{\alpha\beta}(\boldsymbol{R}) = k_{\alpha\beta} C(\boldsymbol{R}) \,, \tag{20}$$

where $k_{\alpha\beta}{=}(2\delta_{\alpha\beta}-1)/4\rho_\alpha\rho_\beta$ with $\rho_\alpha{=}N_\alpha/N$. Note that $C(\boldsymbol{R})$ is the two-body connected correlation function $C_{i,j}$ defined in Eq. (8), but now expressed as a function of $\boldsymbol{R} = \boldsymbol{R}_i - \boldsymbol{R}_j$ instead of as a function of the primary cell addresses i,j. Since the factor $k_{\alpha\beta}$ is independent of $\boldsymbol{R}$, all the correlation lengths for any pair of species in this model are identical to the length defined by

$$1/\xi = -\lim_{|\boldsymbol{R}|\to\infty} |\boldsymbol{R}|^{-1} \ln |C(\boldsymbol{R})| \,. \tag{21}$$

On the other hand, the correlation length for the Ising model is defined by

$$1/\xi^{Ising} = -\lim_{|\boldsymbol{R}|\to\infty} |\boldsymbol{R}|^{-1} \ln |C^{Ising}(\boldsymbol{R})| \,. \tag{22}$$

Therefore the correlation length ξ for the decorated lattice model is calculated from ξ^{Ising} through the transcription given by Eq. (13).

For the Ising model an accurate expression for the correlation length at zero field below T_c is available as a function of $\exp(-2J/kT)$ [7]. Thus, we can calculate ξ for the decorated lattice model along the coexistence curve from the upper to the lower critical solution point. For the simple cubic lattice, the following expression is used:

$$\frac{1}{\xi} = \frac{1}{f}\cosh^{-1}\left(1 + \frac{f^2}{2\Lambda_2'}\right) \tag{23}$$

or

$$\frac{1}{\xi} = \frac{1}{f}\ln\left[1 + \frac{f^2}{2\Lambda_2'} + \sqrt{\left(1 + \frac{f^2}{2\Lambda_2'}\right)^2 - 1}\right] , \tag{24}$$

where for the simple cubic lattice in the direction $\boldsymbol{e}_0 = (1,0,0)$,

$$\Lambda_2'(x) = x^4 - x^6 + 10x^8 - 14x^{10} + 93x^{12} - 201x^{14} + \cdots \quad \text{and} \quad f^2 = 1 \tag{25}$$

and for the same lattice in the direction $\boldsymbol{e}_1 = (1,1,0)/\sqrt{2}$,

$$\Lambda_2'(x) = x^4 - \frac{3}{4}x^6 + 9\frac{7}{16}x^8 - 13\frac{11}{32}x^{10} + \cdots \quad \text{and} \quad f^2 = \frac{1}{2} \,. \tag{26}$$

The variable x in terms of which Λ_2' is expanded is to be understood as $A_{12}/\sqrt{A_{11}A_{22}}$ for the decorated lattice model, instead of $\exp(-2J/kT)$ for the Ising model.

Here, we show the numerical results for the correlation length for the decorated lattice model of the simple cubic lattice. The results are obtained for various values for the parameter ω at $|U_2|/U_1 = 1$. Shown in Fig. 7(a) is the behavior of the correlation

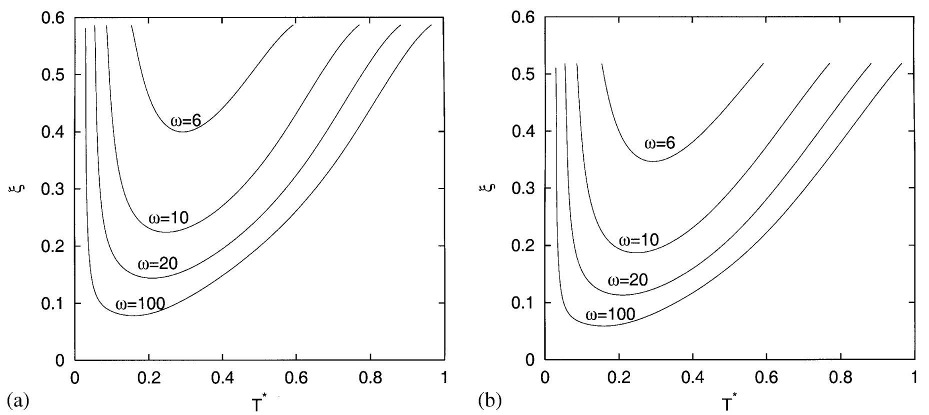

Fig. 7. Behavior of the correlation length along the coexistence curve for the decorated lattice-gas model for various values of ω at $|U_2|/U_1 = 1$. The lattice type is the simple cubic and the lattice direction in which ξ is calculated is (a) $\boldsymbol{e}_0 = (1,0,0)$ and (b) $\boldsymbol{e}_1 = (1,1,0)/\sqrt{2}$. Temperature is in units of U_1/k.

length ξ in the direction $\boldsymbol{e}_0$ along the coexistence curves as a function of temperature between the upper and lower critical solution points. For any given ω, the correlation length takes a minimum value at a temperature lower than the midpoint between the upper and lower critical temperatures. With increasing ω, the minimum of the curve becomes deeper and shifts toward the lower critical temperature. This means that a larger number of possible orientations ω gives rise to a correlation length that increases more rapidly as the system approaches the lower critical solution temperature. Similar results are obtained for the direction $\boldsymbol{e}_1$ as shown in Fig. 7(b).

5. Summary

We have calculated and illustrated the correlations between primary cells for a decorated-lattice-gas model that exhibits a closed-loop phase diagram. The three-body potential of mean force was also calculated and compared with the sum of the three two-body interactions. The calculations are related to those in earlier models of hydrophobic interactions and are interpreted in similar terms.

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