Journal of Statistical Physics, Vol. 98, Nos. 3/4, 2000

A New Statistical Aspect of the Cluster Variation Method for Lattice Systems

Hiromu Asada[1]

Received July 9, 1999; final September 20, 1999

This paper presents an alternative statistical way to derive the cluster variation method (CVM) for lattice systems. The formulation is developed for a series of different clusters, each of which is the largest overlap cluster between two clusters of the next larger type. We arrive at the CVM expression of the lattice configuration factor by deriving the number of different ways of distributing clusters of a selected type in the lattice so that they overlap each other at the largest overlap clusters in a physically correct manner. The essential assumption employed is that individual overlapping events are statistically independent of each other. This reveals a new statistical aspect of the CVM: The CVM is based on a Bethe tree of clusters of the selected type.

KEY WORDS: Cluster variation method; Kikuchi approximation; lattice system; configuration factor; Bethe tree; cluster distribution; cluster series.

1. INTRODUCTION

The cluster variation method (CVM)[1,2] has been recognized by many authors to be one of the most powerful theories to treat analytically lattice systems. The original CVM was started by Kikuchi,[3,4] who derived the lattice configuration factor by considering the process of adding one lattice point of either of the two kinds to the lattice in harmony with the equilibrium distribution of clusters over different cluster configurations. However, complicated steps composing the formulation make it difficult to generalize his method for larger clusters. Later, he and his coworkers[5] reformulated the CVM by introducing a pseudo-assembly of subclusters in which the probability of the subclusters being distributed in a physically

[1] Department of Materials Science, Faculty of Science, Ehime University, Matsuyama 790-8577, Japan.

0022-4715/00/0200-0621$18.00/0

correct manner was assumed to be equal to that of parent clusters (we use *parent* instead of *main* in their papers) in their assembly. However, physical implications of their assumption is not necessarily clear, as mentioned by themselves.[5] Hijmans and de Boer[6, 8] generalized the pseudo-assembly method to treat larger parent clusters. There are different shapes of overlap clusters between two neighboring parent clusters, depending on their separation, and they incorporated all kinds of overlap clusters with their theory.

On the other hand, Barker[9] derived the CVM simply by considering conditional sums of the lattice configuration factor: Counting up microstates of parent cluster distribution which are consistent with a given macrostate of subcluster distribution should yield the lattice configuration factor with respect to the subcluster distribution. His treatment is mathematical rather than statistical when compared with the theories of Kikuchi and his coworkers. His mathematical approach to a series of clusters of different shapes has been refined into sophisticated theories by many authors,[10] and Morita[10, 11] has recently completed elegant mathematical formalism by the Möbius inversion method.

In this paper, we present an alternative statistical derivation of the CVM. We define a series of clusters so that every constituent of the series is the largest overlap cluster between two nearest neighboring clusters belonging to the next larger constituent. When we choose one type of clusters from the series as parent clusters to be distributed in the lattice, only smaller clusters involved in the series are treated as subclusters of the parent cluster. However, we explicitly treat only the largest overlap between parent clusters. We formulate the number of different ways of distributing parent clusters in the lattice so that they overlap each other at the largest overlap clusters in a physically correct manner, assuming that individual overlapping events are statistically independent of each other. A minor correction factor introduced for the deviation from the statistical independence is determined by reducibility conditions of the lattice configuration factor. Our method will be found to be much simpler and much clearer in statistical implications than Kikuchi's original combinatorial method and the pseudo-assembly method. As a consequence, our formulation will reveal a new statistical aspect of the CVM: The lattice system treated in the CVM is a Bethe tree of parent clusters.

This paper is organized as follows. The next section describes our formulation of the CVM. Following some preparatory subsections concerning definitions and internal consistency conditions, Subsections 2.6 and 2.7 show our original derivation of the lattice configuration factor. In the final Section 3, we reveal the new statistical aspect of the CVM by discussing about statistical implications of our formulation.

2. FORMULATION

2.1. Series of Clusters

Consider a lattice composed of N_1 lattice points which are equivalent to each other with respect to the translational and rotational symmetry of the lattice. However, the lattice points are classified into two types, A and B. They refer to A and B atoms in an alloy system, up and down spins in an Ising system, or occupied and empty sites in a lattice gas system. The number of the lattice points of A type is denoted as N_A.

We consider a cluster of some nearest neighboring lattice points forming a certain shape. The shape applies to congruence, i.e., is not affected by orientation. There exist many clusters of the same shape in the lattice. Every lattice point belongs to some different clusters of the same shape. In other words, the lattice is composed of many clusters of the same shape partially overlapping each other. The common part shared by two overlapping clusters is called the overlap cluster, whose size and shape depend on the separation and orientation of the two clusters. In a triangular lattice for instance, a triangular cluster composed of three nearest neighboring lattice points can overlap its neighboring triangular cluster at its angle or at its side.

The cluster that involves a single lattice point as the largest overlap cluster is a pair of nearest neighboring lattice points. The cluster that involves a two-point pair as the largest overlap cluster is, for example, a triangle composed of three nearest neighboring lattice points for a triangular lattice and a square composed of four nearest neighboring lattice points for a square lattice. In this way we can derive, but not uniquely, a series of clusters starting with a single lattice point. We call the ith cluster in the series the i-cluster. We regard a single lattice point as the smallest cluster, and we call it the 1-cluster. The 2-cluster is a pair of nearest neighboring lattice points, and so on. Note that the i-cluster is the largest overlap cluster shared by two $(i+1)$-clusters. Fig. 1 shows examples for a triangular lattice, a square one, and a simple cubic one. An alternative series of clusters for a triangular lattice is such that the 1- through 4-clusters are the same as in Fig. 1(a), but the 5-cluster is an equilateral triangle composed of six lattice points, the 6-cluster is a rhombus composed of two contacting 5-clusters, and so on.

We denote the number of i-clusters contained in the lattice as N_i. We have

$$N_i = a_i N_1 \tag{1}$$

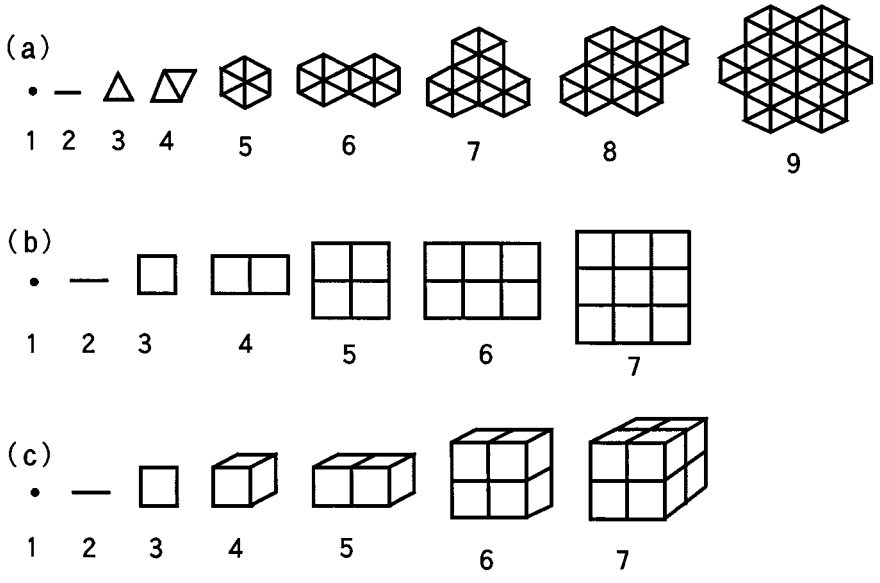

Fig. 1. Series of clusters. The i-cluster is the largest overlap cluster between two $(i+1)$-clusters. In these examples, the $(i+1)$-cluster is also the largest parent cluster for the i-subcluster: (a) triangular, (b) square, and (c) simple cubic lattices.

where a_i is a positive integer. For example, if we adopt the series of clusters shown in Fig. 1(a) for a triangular lattice, the lattice contains N_1 lattice points $(a_1=1)$, $3N_1$ two-point pairs $(a_2=3)$, $2N_1$ triangles $(a_3=2)$, $3N_1$ rhombi $(a_4=3)$, and so on.

2.2. Cluster Configurations and Cluster Distributions

Clusters have some different configurations of A- and B-lattice-points, which cannot be transformed to each other by rotation or mirror image. For example, two-point pairs have three different configurations, AA, AB, and BB, and triangles have four different configurations, AAA, AAB, ABB, and BBB. The number of different configurations of the i-cluster is denoted as C_i. An i-cluster with the kth configuration is called an i_k-cluster. The number of different ways of placing an i_k-cluster in the lattice with each vertex at a fixed position is called the statistical weight or the number of different cluster orientations, w_{ik}. Fig. 2 shows some examples of different configurations of clusters in a triangular lattice and in a square one together with their statistical weights.

The number of i_k-clusters contained in the lattice is denoted as N_{ik}. For $i=1$, $N_{11}=N_1-N_A$ and $N_{12}=N_A$. We have

$$\sum_{k=1}^{C_i} N_{ik}=N_i \tag{2}$$

For later convenience, we define the fraction n_{ik} of i-clusters having the configuration k as

$$n_{ik}\equiv N_{ik}/N_i \tag{3}$$

The set of numbers, N_{i1}, N_{i2},..., N_{i,C_i} is abbreviated as $\{i\}$. The set $\{i\}$ represents a macrostate of the lattice with respect to the i-cluster distribution.

Our aim is to obtain (although approximately) the number of different ways, Z^i, of distributing $\{i\}$ clusters in the lattice, which is also called the lattice configuration factor with respect to the i-cluster distribution. When we distribute $\{i\}$ clusters at random at the N_i positions in the lattice, the number of different distributions, $G_i(\{i\})$, is given by

$$G_i(\{i\})\equiv N_i!\prod_{k=1}^{C_i}[w_{ik}^{N_{ik}}/N_{ik}!] \tag{4}$$

However, this number includes not only physically correct distributions but also incorrect ones: *Physically correct* means that every A- or B-lattice point of any i-cluster overlaps a lattice point of the same kind of any neighboring i-cluster. Only when $i=1$ (i.e., clusters are single lattice points), all of G_1 distributions are physically correct. We shall derive the probability of the distribution being physically correct when $\{i\}$ clusters are placed at random in the lattice. Then, Z^i is given by $G_i(\{i\})$ multiplied by this probability.

2.3. Subclusters and Their Internal Positions and Orientations

An i-cluster contains smaller clusters, i.e., 1- through $(i-1)$-clusters of the series. These are called subclusters of the parent i-cluster. Note that the subclusters do not necessarily include all the possible overlap clusters shared by two i-clusters. For example, when we take the 4-cluster (rectangle) as the parent cluster in a square lattice (see Fig. 1(b)), two 4-clusters can overlap each other at their long sides (linear chains of three nearest neighboring lattice points), but the long side is not taken as the subcluster of the 4-cluster since it does not appear in the cluster series.

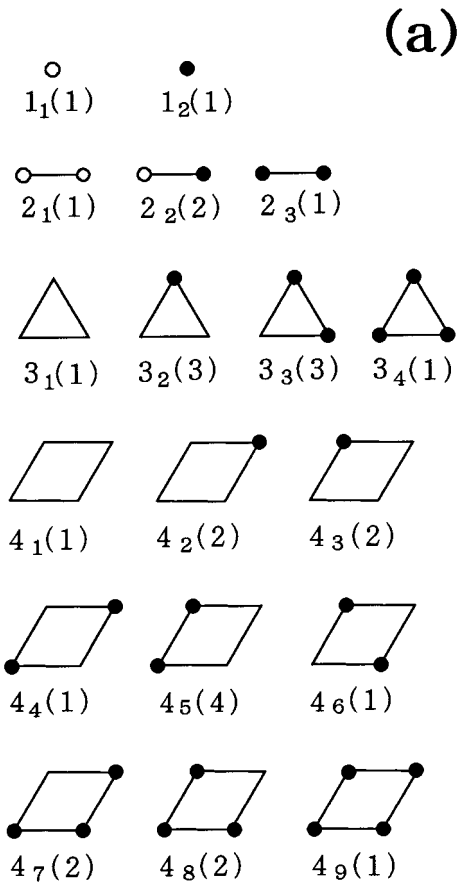

Fig. 2. Different configurations of clusters of the type 1 through 4 in a triangular lattice (a) and in a square lattice (b). Closed and open circles represent lattice points of A and B types, respectively, but open circles are omitted for 3- and 4-clusters. Statistical weights are given in parentheses.

We call a j-cluster contained in an i-cluster a $j^{(i)}$-subcluster. We denote the number of $j^{(i)}$-subclusters contained in one parent i-cluster as b_j^i. When lattice points contained in a parent cluster are not all equivalent in the sense that they cannot all be transformed to each other by rotation or mirror image, b_j^i positions for $j^{(i)}$-subclusters in the parent i-cluster are not always all equivalent in the same sense. We denote the number of these constitutionally non-equivalent internal positions for a $j^{(i)}$-subcluster as P_j^i,

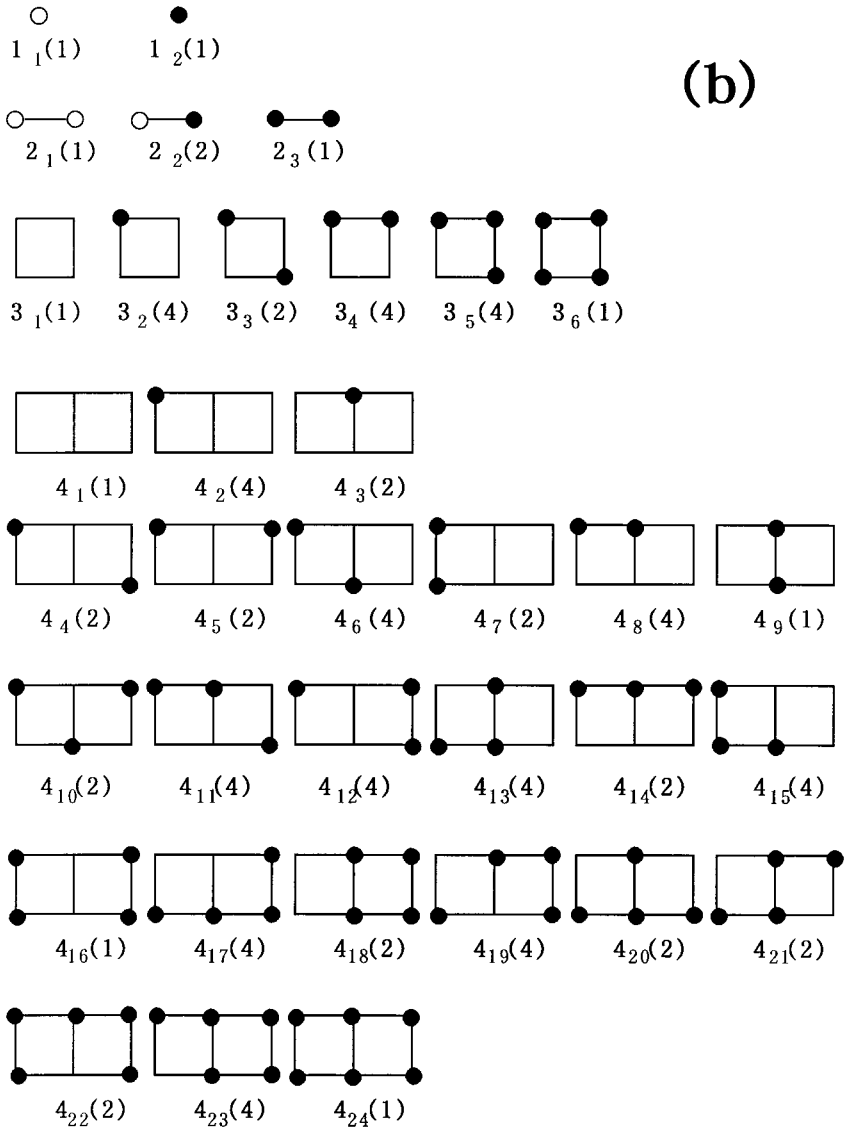

Fig. 2. (*Continued*)

and the number of the non-equivalent internal positions of the sth kind in a parent i-cluster as b_j^{is}. We have

$$\sum_{s=1}^{P_j^i} b_j^{is} = b_j^i \tag{5}$$

We call the $j^{(i)}$-subcluster located at an internal position of the sth kind the $j^{(is)}$-subcluster.

For example, consider the 4-cluster (rhombus) in a triangular lattice (see Fig. 2(a)). It has four $1^{(4)}$-subclusters (single lattice points, $b_1^4 = 4$) at two kinds of non-equivalent internal positions ($P_1^4 = 2$), i.e., two acute and two obtuse angle sites ($b_1^{41} = b_1^{42} = 2$), and five $2^{(4)}$-subclusters (two-point pairs, $b_2^4 = 5$) at two kinds of non-equivalent internal positions ($P_2^4 = 2$), i.e., four sides ($b_2^{41} = 4$) and one short diagonal site ($b_2^{42} = 1$) of the rhombus. But, the two $3^{(4)}$-subclusters (triangles, $b_3^4 = 2$) occupy equivalent internal positions ($P_3^4 = 1$).

It is noted that the largest subcluster, i.e., the $(i-1)^{(i)}$-subcluster, has in general only one kind of internal positions, i.e., $P_{i-1}^i = 1$. Because, if not, two largest subclusters at different kinds of internal positions can be combined to form a larger overlap cluster.

A j_q-cluster has w_{jq} orientations in the lattice, which are equivalent in the sense that they can be transformed to each other by rotation or mirror image. However, all of the w_{jq} cluster orientations are not always equivalent in the parent i-cluster in the same sense. These constitutionally non-equivalent orientations of subclusters, which we call the non-equivalent internal orientations, arise from the constitutional non-equivalency of the lattice sites in the parent cluster. We call a $j^{(is)}$-subcluster having the qth configuration a $j_q^{(is)}$-subcluster. We denote the number of non-equivalent internal orientations of a $j_q^{(is)}$-subcluster as O_{jq}^{is}, and the statistical weight of the rth kind of non-equivalent internal orientations as w_{jqr}^{is}. We have for any s

$$\sum_{r=1}^{O_{jq}^{is}} w_{jqr}^{is} = w_{jq} \tag{6}$$

For example, a 3_2-cluster of a triangular lattice (an ABB triangle) has three equivalent cluster orientations in the lattice ($w_{32} = 3$), but when it is contained by a parent 4-cluster (a rhombus), the three orientations are not all equivalent: The $3_2^{(4)}$-subcluster has a single kind of internal positions ($P_3^4 = 1$), but its orientations are classified according as the A-point is located either at an acute angle ($w_{321}^4 = 1$) or at an obtuse angle ($w_{322}^4 = 2$)

of the rhombus, i.e., $O_{32}^4 = 2$. It is found that the two kinds of sites, i.e., acute and obtuse angle sites of the rhombus, give rise to non-equivalent internal orientations of the subclusters.

2.4. Numbers of Subclusters and Overlap Multiplicity

The total number of $j^{(i)}$-subclusters contained in all the i-clusters is $b_j^i N_i$. Since the total number of j-clusters contained in the lattice is N_j, the lattice is covered c_j^i times by the $j^{(i)}$-subclusters, where

$$c_j^i \equiv b_j^i N_i / N_j = a_i b_j^i / a_j \tag{7}$$

In other words, c_j^i nearest neighboring i-clusters share a common $j^{(i)}$-subcluster. The parameter c_j^i is called the overlap multiplicity of i-clusters with respect to $j^{(i)}$-subclusters.

In a triangular lattice, for example, the number N_4 of 4-clusters (rhombi) in the lattice is $3N_1$ ($a_4 = 3$) and each of them contains two $3^{(4)}$-subclusters (triangles), i.e., $b_3^4 = 2$. The total number of the $3^{(4)}$-subclusters is then $b_3^4 N_4 = 6N_1$, while the number N_3 of 3-clusters in the lattice is $2N_1$ ($a_3 = 2$). Accordingly, the number of times c_3^4 that the $3^{(4)}$-subclusters cover the lattice is $6N_1/2N_1 = 3$. It is also easy to find that a $3^{(4)}$-subcluster is shared by three 4-clusters. Tables I and II list values of a_i, b_j^i, and c_j^i for some small clusters of the cluster series in a triangular lattice and in a square one, respectively.

We call a $j_q^{(is)}$-subcluster with an internal orientation of the rth kind a $j_{qr}^{(is)}$-subcluster. We denote the numbers of $j_q^{(i)}$-, $j_q^{(is)}$-, and $j_{qr}^{(is)}$-subclusters contained in one parent i_k-cluster as m_{jq}^{ik}, m_{jq}^{iks}, and m_{jqr}^{iks}, respectively. The total numbers of $j_q^{(i)}$-, $j_q^{(is)}$- and $j_{qr}^{(is)}$-subclusters contained in all the

Table I. Parameters for a Triangular Lattice[a]

i-cluster	a_i	b_1^i	b_2^i	b_3^i	b_4^i	b_5^i	c_1^i	c_2^i	c_3^i	c_4^i	c_5^i	x_1^i	x_2^i	x_3^i	x_4^i	x_5^i
1	1	1	0	0	0	0	1	0	0	0	0	1	0	0	0	0
2	3	2	1	0	0	0	6	1	0	0	0	−5	1	0	0	0
3	2	3	3	1	0	0	6	2	1	0	0	1	−1	1	0	0
4	3	4	5	2	1	0	12	5	3	1	0	1	0	−2	1	0
5	2	6	9	4	3	1	12	6	4	2	1	1	0	0	−1	1

[a] Clusters of the type 1 through 4 are illustrated in Fig. 1 but the 5-cluster is the equilateral triangular cluster composed of six lattice points.

Table II. Parameters for a Square Lattice[a]

i-cluster	a_i	b_1^i	b_2^i	b_3^i	b_4^i	b_5^i	c_1^i	c_2^i	c_3^i	c_4^i	c_5^i	x_1^i	x_2^i	x_3^i	x_4^i	x_5^i
1	1	1	0	0	0	0	1	0	0	0	0	1	0	0	0	0
2	2	2	1	0	0	0	4	1	0	0	0	−3	1	0	0	0
3	1	4	4	1	0	0	4	2	1	0	0	1	−1	1	0	0
4	2	6	7	2	1	0	12	7	4	1	0	1	0	−3	1	0
5	1	9	12	4	4	1	9	6	4	2	1	0	0	1	−1	1

[a] The clusters are illustrated in Fig. 1.

i-clusters are denoted as M_{jq}^i, M_{jq}^{is} and M_{jqr}^{is}, respectively. These are defined by

$$M_{jq}^i \equiv \sum_{k=1}^{C_i} m_{jq}^{ik} N_{ik} \tag{8.1}$$

$$M_{jq}^{is} \equiv \sum_{k=1}^{C_i} m_{jq}^{iks} N_{ik} \tag{8.2}$$

$$M_{jqr}^{is} \equiv \sum_{k=1}^{C_i} m_{jqr}^{iks} N_{ik} \tag{8.3}$$

We have

$$\sum_{r=1}^{O_{jq}^{is}} M_{jqr}^{is} = M_{jq}^{is}, \qquad \sum_{s=1}^{P_j^i} M_{jq}^{is} = M_{jq}^i, \qquad \text{and} \qquad \sum_{q=1}^{C_j^i} M_{jq}^i = b_j^i N_i \tag{9}$$

The definition of the overlap multiplicity gives us a relation:

$$M_{jq}^i = c_j^i N_{jq} \tag{10}$$

2.5. Internal Consistency of the Parent Cluster

We have to note that the $j_q^{(i)}$-subcluster has no preference to any of the non-equivalent internal positions in the parent i-cluster. In other words, any parent i-cluster distribution $\{i\}$ is not always physically correct, but only the sets $\{i\}$ that realize a uniform distribution of $j_q^{(i)}$-subclusters over different internal positions are physically allowed. This requirement can be expressed as follows.

$$\frac{M_{jq}^{i1}}{b_j^{i1}} = \frac{M_{jq}^{i2}}{b_j^{i2}} = \cdots = \frac{M_{jq}^{iP}}{b_j^{iP}} \qquad \left(= \frac{M_{jq}^i}{b_j^i} \right) \tag{11}$$

where the superscripts P are abbreviations of P_j^i. We have similar sets of equations for each of C_j different subcluster configurations q. These equations are called the internal consistency conditions of the i-cluster with respect to the statistical uniformity among the non-equivalent internal positions of the $j^{(i)}$-subcluster.

We have to recall also that the j_q-cluster has no preferential orientation in the lattice. $j_q^{(is)}$-clusters have to be distributed uniformly over different internal orientations. The requirement can be expressed as

$$\frac{M_{jq1}^{is}}{w_{jq1}^{is}} = \frac{M_{jq2}^{is}}{w_{jq2}^{is}} = \cdots = \frac{M_{jqO}^{is}}{w_{jqO}^{is}} \qquad \left(= \frac{M_{jq}^{is}}{w_{jq}} \right) \tag{12}$$

for every kind s of the non-equivalent internal positions and for every configuration q of the $j^{(i)}$-subcluster. The subscripts O are abbreviations of O_{jq}^{is}. These equations are called the internal consistency conditions of the i-cluster with respect to the statistical uniformity among the non-equivalent internal orientations of $j^{(i)}$-subclusters.

2.6. Physically Correct Overlap Between i-Clusters

Suppose that we distribute $\{i\}$-clusters at random in the lattice. The distribution may be either physically correct or incorrect. The number of different distributions including physically incorrect ones is $G_i(\{i\})$. Consider an i-cluster at any specified position in the lattice, which we call the central i-cluster. It contains $b_j^i j^{(i)}$-subclusters, each of which is shared by $c_j^i - 1$ neighboring i-clusters. We focus our attention to one of the $j^{(i)}$-subclusters. Suppose the $j^{(i)}$-subcluster is of the $j_{qr}^{(is)}$-type. In order that one neighboring i-cluster overlaps the central one at the $j_q^{(i)}$-subcluster in concern in a physically correct manner, the neighboring i-cluster should contain a $j_q^{(i)}$-subcluster at the proper internal position of the s'th kind with the proper internal orientation of the r'th kind. Note that s' and r' are not necessarily identical to s and r, respectively, but that they are particular ones depending on s and r, respectively.

For example, Fig. 3 shows a 4_2-cluster ($ABBB$ rhombus, $PQRS$) in a triangular lattice which shares its $3_2^{(4)}$-subcluster (ABB triangle, PQR) with two neighboring 4-clusters, $XQRP$ and $YRPQ$. The internal orientation of the $3_2^{(4)}$-subcluster in $PQRS$ is equivalent to that in $XQRP$, but not equivalent to that in $YRPQ$.

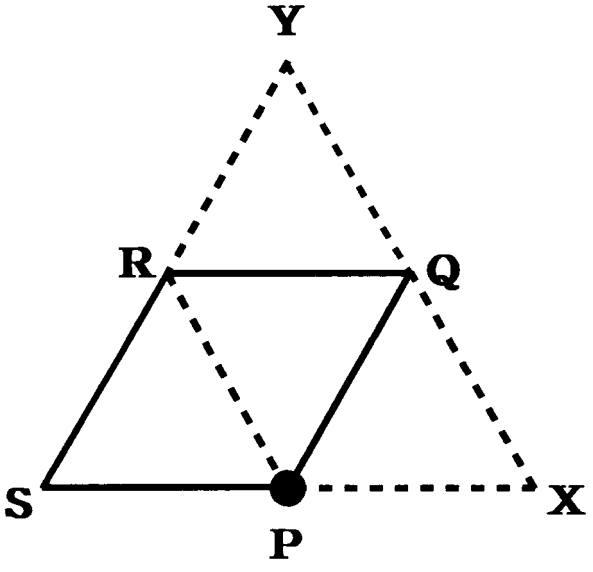

Fig. 3. Overlap of three rhombi clusters in a triangular lattice. The rhombus *PQRS* is a 4_2-cluster which has a *A*-lattice-point (marked by a closed circle) at *P* and *B*-lattice-points at *Q*, *R*, and *S*. Its $3_2^{(4)}$-subcluster *PQR* (*ABB*-type triangle) is shared by neighboring rhombi *XQRP* and *YRPQ*. The subcluster *PQR* has an internal orientation in *XQRP*, equivalent to that in *PQRS*, but does not in *YRPQ*.

The probability $u_{jqr'}^{is'}$ of the neighboring *i*-cluster having such a physically correct $j_{qr}^{(is')}$-subcluster with the proper internal orientation at the proper internal position is given by

$$u_{jqr'}^{is'} = \sum_k n_{ik} m_{jqr'}^{iks'} / b_j^{is'} w_{jqr'}^{is'} \tag{13}$$

The factor n_{ik} represents the probability of the parent *i*-cluster having the *k*th configuration, $m_{jqr'}^{iks'}/b_j^{is'}$ the probability of the parent i_k-cluster containing a $j_{qr'}^{(is')}$-subcluster at the proper internal position of the s'th kind, and $1/w_{jqr'}^{is'}$ the probability of the $j_{qr'}^{(is')}$-subcluster having the proper internal orientation of the r'th kind. Note that the probabilities, $m_{jqr'}^{iks'}/b_j^{is'}$ and $1/w_{jqr'}^{is'}$, are valid under the internal consistency conditions, Eqs. (11) and (12), respectively. The summation over the configurations *k* of the parent cluster gives the probability in question. We can rewrite Eq. (13) as

$$u_{jqr'}^{is'} = M_{jqr'}^{is'} / N_i b_j^{is'} w_{jqr'}^{is'} = n_{jq} / w_{jq} \tag{14}$$

The first equality has been derived by using Eq. (8.3), and the second derived by using the internal consistency conditions, Eqs. (11) and (12). Since the final expression does not depend on the internal position s' and the internal orientation r', we omit the superscript s' and subscript r' for u hereafter.

We assume that every couple of neighboring i-clusters overlap each other statistically independently of any other couple. Then, the probability of the central i-cluster sharing its $j_q^{(i)}$-subcluster with $c_j^i - 1$ neighboring i-clusters in a physically correct manner is given by u_{jq}^i raised to the power of $c_j^i - 1$. Further raising the power M_{jq}^i times gives the probability of all the $M_{jq}^i j_q^{(i)}$-subclusters being shared by c_j^i i-clusters each in a physically correct manner, but this probability counts every overlapping between two i-clusters c_j^i times. Then, the probability v_j^i of all the i-clusters overlapping each other at $j^{(i)}$-subclusters in a physically correct manner is given by

$$\begin{aligned} v_j^i &= \prod_{q=1}^{C_j} u_{jq}^{i\ (c_j^i - 1)\, M_{jq}^i / c_j^i} \\ &= \prod_{q=1}^{C_j} (n_{jq}/w_{jq})^{(c_j^i - 1)\, N_{jq}} \\ &= G_j(\{j\})^{1 - c_j^i} \end{aligned} \tag{15}$$

The first expression is derived by substituting Eq. (10) and the last one is obtained within the Stirling approximation.

However, successive overlapping of parent clusters in the lattice generates multiple loops of overlapping clusters to violate the assumption of statistical independence of individual overlapping. Then, to obtain the probability p_j^i of all the i-clusters overlapping correctly at the $j^{(i)}$-subclusters in the lattice, we introduce a correction factor y_j^i in such a manner as

$$p_j^i = v_j^i y_j^i \tag{16}$$

We assume that y_j^i is a minor factor which can be represented as a function of $\{j-1\}$. Then, the number of different i-cluster distributions which are physically correct with respect to overlapping at $j^{(i)}$-subclusters ($1 \leqq j < i$), or more simply, the lattice configuration factor $Z_j^i(\{i\})$, can be expressed as

$$Z_j^i(\{i\}) = G_i(\{i\}) \times p_j^i = G_i(\{i\})\, G_j(\{j\})^{1 - c_j^i}\, y_j^i(\{j-1\}) \tag{17}$$

It is an explicit form of $Z_{i-1}^i(\{i\})$ that we want to obtain, since Z_{i-1}^i is expected to be the most precise configuration factor among Z^i's.

2.7. Reducibility of the Configuration Factor

The lattice configuration factor must satisfy the reducibility condition: Summing up $Z_j^i(\{i\})$ over possible distributions $\{i\}$ that are consistent with a given distribution of smaller j-clusters, $\{j\}$, should yield $Z_{j-1}^j(\{j\})$, i.e.,

$$\sum_{\{i\}}^{(i,j)'} Z_j^i(\{i\}) = Z_{j-1}^j(\{j\}) \tag{18}$$

The constraints (i, j) on the summation consist of Eq. (2) the normalization condition of $\{i\}$, Eq. (10) the distribution $\{j\}$, and Eqs. (11) and (12) the internal consistency conditions with respect to the $j^{(i)}$-subcluster. We substitute Eq. (17) into Eq. (18) and then replace the summation with the maximum term only, where we denote $\{i\}$ as $\{i^*\}_j$, which are functions of $\{j\}$:

$$\sum_{\{i\}}^{(i,j)'} Z_j^i(\{i\}) = G_i(\{i^*\}_j)\, G_j(\{j\})^{1-c_j^i}\, y_j^i(\{j-1\}) \tag{19}$$

We can see from Eq. (17) that the leading factor in the right hand side of Eq. (18) is $G_j(\{j\})$. Then we can express $G_i(\{i^*\}_j)$ as

$$G_i(\{i^*\}_j) = G_j(\{j\})^{c_j^i} f_j^i(\{j-1\}) \tag{20}$$

where the minor factor f_j^i is a function of $\{j-1\}$.

Next, we consider the reduction of $Z_j^i(\{i\})$ for a given $\{j-1\}$. We have

$$\begin{aligned}\sum_{\{i\}}^{(i,j-1)'} Z_j^i(\{i\}) &= G_i(\{i^*\}_{j-1})\, G_j(\{j^*\}_{j-1})^{1-c_j^i}\, y_j^i(\{j-1\}) \\ &= G_{j-1}(\{j-1\})^{c_{j-1}^i + (1-c_j^i)\, c_{j-1}^j}\, y_j^i(\{j-1\}) \\ &\quad \times \text{function of } \{j-2\}\end{aligned} \tag{21}$$

The last expression is obtained by applying Eq. (20) to the first two factors in the second expression and should be identical to

$$Z_{j-2}^{j-1}(\{j-1\}) = G_{j-1}(\{j-1\})\, G_{j-2}(\{j-2\})^{1-c_{j-2}^{j-1}}\, y_{j-2}^{j-1}(\{j-3\}) \tag{22}$$

Comparing the leading factors of these two expressions, we see that $y_j^i(\{j-1\})$ should contain a factor $G_{j-1}(\{j-1\})$ raised to a power

$1-c^i_{j-1}-(1-c^i_j)\,c^j_{j-1}$. In this way, we can prove via a recursive procedure that $y^i_j(\{j-1\})$ is factorized into $G_k(\{k\})$'s with $1 \leqq k \leqq j-1$.

Now we consider the configuration factor $Z^i_{i-1}(\{i\})$. According to the above discussion, we represent its correction factor y^i_{i-1} as

$$y^i_{i-1}(\{i-2\}) = \prod_{k=1}^{i-2} G_k(\{k\})^{x^i_k} \tag{23}$$

When we define $x^i_i \equiv 1$ and $x^i_{i-1} \equiv 1 - c^i_{i-1}$ for convenience, we have from Eqs. (17) and (23)

$$Z^i_{i-1}(\{i\}) = \prod_{k=1}^{i} G_k(\{k\})^{x^i_k} \tag{24}$$

Summing up Z^i_{i-1} for a given j-cluster distribution $\{j\}$, we have

$$\sum_{\{i\}}^{(i,j)'} z^i_{i-1}(\{i\}) = \prod_{k=1}^{j} G_k(\{k\})^{x^i_k} \prod_{k=j+1}^{i} G_k(\{k^*\}_j)^{x^i_k} \tag{25}$$

Applying Eq. (20), we obtain the leading factor $G_j(\{j\})$ raised to the power of $\sum_{k=j}^{i} x^i_k c^k_j$. Letting this to be equal to the leading factor $G_j(\{j\})$ in $Z^j_{j-1}(\{j\})$, we arrive at the goal:

$$\sum_{k=j}^{i} x^i_k c^k_j = 1 \tag{26}$$

Since Eq. (26) turns out to be to $x^i_i = 1$ when $j = i$ and to $x^i_{i-1} = 1 - c^i_{i-1}$ when $j = i-1$, it holds for any j with $1 \leqq j \leqq i$ and determines uniquely $\{x^i\}$. When we define $x^i_k = 0$ for $k > i$, we have a matrix representation:

$$\begin{pmatrix} 1 & & & \\ & 1 & & 0 \\ & & \ddots & \\ x^i_k & & & 1 \end{pmatrix} \begin{pmatrix} 1 & & & \\ & 1 & & 0 \\ & & \ddots & \\ c^k_j & & & 1 \end{pmatrix} = \begin{pmatrix} 1 & & & \\ & 1 & & 0 \\ & & \ddots & \\ 1 & & & 1 \end{pmatrix} \tag{27}$$

Equation (26) can alternatively be interpreted as follows: If we regard the lattice as being covered by parent clusters of the type 1 through i and x^i_j-fold by parent j-clusters (note that x^i_j may also be negative) respectively, then Eq. (26) implies that for respective j, the aggregate of the parent j-clusters and j-subclusters contained in larger parent clusters cover the lattice only one fold.

3. CONCLUDING REMARKS

In summary, the lattice configuration factor $Z^{i}_{i-1}(\{i\})$ is given by Eq. (24) with $\{x^{i}\}$ defined by Eq. (26): The i-cluster distribution $\{i\}$ must meet Eq. (2), the normalization condition of $\{i\}$, as well as the internal consistency conditions Eq. (12) with respect to the $(i-1)^{(i)}$-subcluster. The distribution of smaller clusters, $\{j\}$ with any $j<i$, is defined as functions of $\{i\}$ by Eq. (10) combined with Eq. (8.1). It should be pointed out that the cluster series in which the j-cluster is the largest overlap cluster between nearest neighboring $(j+1)$-clusters enables us to establish a consistent relation between lattice configuration factors through reducibility conditions.

Tables I and II include x's obtained for small parent clusters in a triangular lattice and in a square one, respectively. They indicate that the configuration factors $Z^{i}_{i-1}(\{i\})$ for $i=1, 2$, and 3 correspond to the Bragg–Williams, the quasichemical, and the Kikuchi approximation, respectively.[1] These approximations require no internal consistency conditions.

It should be mentioned that Hijmans and de Boer[6] have already derived a set of equations equivalent to Eq. (26) by the pseudo-assembly method, although their definition of cluster series is different from ours. Morita[10, 11] also has recently derived a formula equivalent to Eq. (26) in an elegant mathematical manner by applying the Möbius inversion method to a generalized cluster series. However, our method of deriving Eq. (26) discloses much simpler and much clearer statistical implications of the

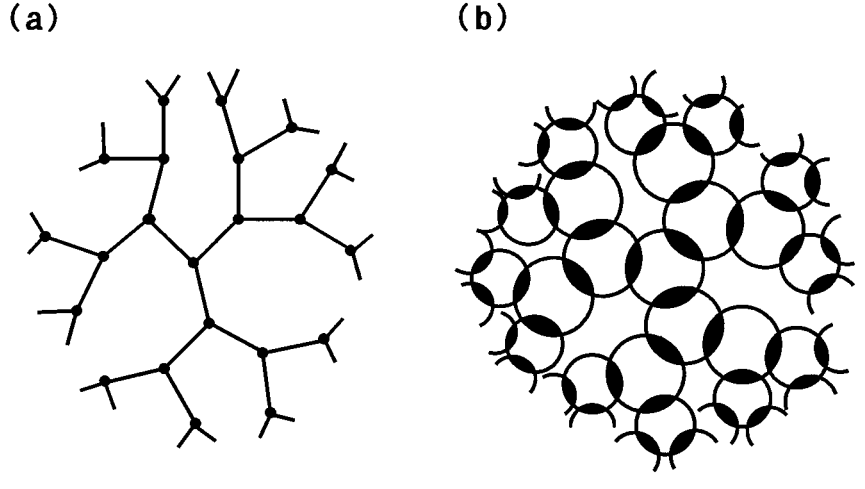

Fig. 4. Bethe trees with the coordination number 3: (a) Every lattice point is connected with three other lattice points, but there exists no loop of bonds; (b) The lattice points and bonds are replaced with parent clusters (circles) and overlap subclusters (painted parts), respectively.

CVM than ever. The essential assumption employed in our formulation is that individual overlapping events among parent clusters are statistically independent of each other. This implies that we disorganize all of interaction loops among parent clusters to transform the lattice into a kind of Bethe tree where the constituent entities are parent clusters instead of lattice points: Every parent cluster is connected with nearest neighboring parent clusters by sharing the largest subclusters, but the connection between the parent clusters does not form any loop. Figure 4 illustrates a Bethe tree of parent clusters. This is a new statistical aspect of the CVM, which has been revealed by this work.

REFERENCES

1. C. Domb, *Adv. Phys.* **9**:149 (1960).
2. M. E. Fisher, *Rep. Prog. Phys.* **30**:615 (1967).
3. R. Kikuchi, *Phys. Rev.* **81**:988 (1951).
4. R. Kikuchi, *J. Chem. Phys.* **19**:1230 (1951).
5. M. Kurata, R. Kikuchi, and T. Watari, *J. Chem. Phys.* **21**:434 (1953).
6. H. Hijmans and J. de Boer, *Physica* **21**:471 (1955).
7. H. Hijmans and J. de Boer, *Physica* **21**:485 (1955).
8. H. Hijmans and J. de Boer, *Physica* **21**:499 (1955).
9. J. A. Barker, *Proc. Roy. Soc. A* **216**:45 (1953).
10. See a recent review: T. Morita, *Prog. Theor. Phys. Suppl.* **115**:27 (1994).
11. T. Morita, *J. Stat. Phys.* **59**:819 (1990).