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On Some Variational Approximations in Two-Dimensional Classical Lattice Systems

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A new derivation is presented of some variational approximations for classical lattice systems that belong to the class of cluster-variation methods, among them the well-known Bethe–Peierls and Kramers–Wannier approximations. The limiting behavior of a hierarchical sequence of cluster-variation approximations, the so-called C hierarchy, is discussed. It is shown that this hierarchy provides a monotonically decreasing sequence of upper bounds f_n on the free energy per lattice site f and that $f_n \to f$ as $n \to \infty$. Our results are based on extension theorems for states given on subsets of the lattice, which might be of some independent interest, and on an application of transfer matrix concepts to the variational characterization of translation-invariant equilibrium states.

KEY WORDS: Classical lattice systems; variational principle; translation-invariant equilibrium states; cluster-variation method.

1. INTRODUCTION

If one wants to predict the macroscopic thermodynamic behavior of a system on the basis of a model for its microscopic structure, one is faced with the problem of calculating the free energy density of the system in equilibrium. A convenient approach to this problem is via the variational principle that states that the equilibrium free energy density is the minimum of the free energy functional over a certain set of thermodynamic states.

In practice it is necessary to introduce some kind of approximation to cast the variational problem into a manageable form. A well-established technique is to restrict the variation to a subset of all admissible states that may be characterized in terms of a finite number of parameters and to

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minimize by variation of these parameters. Evidently this technique results in an upper bound for the free energy density.

In this paper we are concerned with classical lattice models. The well-known mean-field approximation then falls into the above-mentioned category. It corresponds to restricting the variation to the set of all product states, i.e., states without correlations between the lattice sites.

An attempt to provide a systematic set of improvements on the mean-field approach by taking into account more and more correlations between sites is the cluster-variation method (CVM).[1] The CVM includes the well-known Bethe–Peierls approximation[2,3] and the Kramers–Wannier approximation.[4] The basic feature of the CVM is that it supplies approximate expressions for the configurational mean entropy (entropy per lattice site) in terms of the entropy expressions for finite clusters (sets of lattice points). For lattice models with finite-range interactions it is then possible to construct an approximate expression for the free energy functional which depends only on the restriction of the thermodynamic state to a certain finite set of clusters, the so-called basic clusters. This expression then is minimized by variation of the probability density functions corresponding to these basic clusters.[2] Thus it would seem that CVM approximations are of an entirely different character than the mean-field approximation.

Despite its limitations (e.g., it is known that critical exponents cannot be reproduced correctly by the CVM) the CVM seems to be to date the best available approximation method for calculating phase diagrams from molecular interactions.[6] It has been used extensively during the past decades (for some examples see Refs. 6–13). In contrast to the huge amount of applications, comparatively little effort has been spent on clarifying the exact nature of the approximations involved in the CVM and their influence on the results of the calculations.[14]

This paper is the third in a series reporting results of an investigation into the nature of the CVM.[15,16] It is our hope that a better understanding of the mathematical aspects of the CVM approximations will help to answer the major practical question in applications: namely, what correlations should be taken into account in setting up the approximation and what correlations may safely be neglected; in other words: how to select the basic cluster(s) so as to achieve maximum accuracy with a minimum of computational effort.

This paper is mainly concerned with the so-called "C hierarchy" of the CVM, introduced by Kikuchi and Brush.[17] This sequence of cluster-variation approximations for the square lattice in two dimensions, Z^2, is

[2] For a review of the cluster-variation method see Ref. 5.

interesting for several reasons. It is one of the more successful hierarchies of approximations for two-dimensional lattices, it includes the Kramers–Wannier approximation as its first element, and it provides some insight into the general structure of equilibrium states for finite-range interactions. Moreover our analysis of this C hierarchy leads to some results on the quasichemical or Bethe–Peierls approximation.

The organization of this paper is as follows. In Section 2 we introduce briefly the necessary background, define some notation, and, for completeness, review the relationship between the variational principle for the free energy per lattice site and the CVM.

In Section 3 a new derivation of the C hierarchy of CVM approximations is presented. Our approach enables us to prove that the C hierarchy provides a monotonically decreasing and converging sequence of upper bounds f_n^C for the free energy per site f. The structure of the approximations to the equilibrium state is discussed too.

In Section 4 we apply the techniques of Section 3 to the Bethe (or Bethe–Peierls or quasichemical) approximation. It is proved that for a large class of models on Z^2 this approximation provides an upper bound for the mean free energy f and that the Bethe mean free energy f_B is an improvement over the mean-field prediction f_{MF} in the sense that $f_{MF} \geqslant f_B \geqslant f$. The situation is different on the triangular lattice, however: in that case the first "sensible" improvement over the mean-field approximation seems to be the triangle approximation of the CVM.[10]

Section 5 contains a summary and some final remarks. It is pointed out how the exact results of the previous sections suggest various practical improvements or extensions of the CVM techniques. Also the relation between our approach to the approximations and previous formulations of the CVM is briefly discussed.

The results of Section 3 suggest the existence of an exact variational principle for the mean free energy which depends only upon the restriction of the thermodynamic states to a certain minimal subset of Z^2. Such a variational principle is established in an Appendix. The results of the Appendix sharpen a result given in Ref. 16.

2. CLUSTER EXPANSION OF THE ENTROPY AND THE CVM

Consider the square lattice in two dimensions Z^2. The origin $(0, 0) \in Z^2$ will be denoted by 0 and δ_1 and δ_2 will be the unit vectors $(1, 0)$ and $(0, 1)$, respectively. The δ_1 direction will often be called the horizontal direction and the δ_2 direction will be called the vertical one.

With each lattice point or site a of Z^2 we associate a variable ("spin") σ_a, which can take values in a finite set Ω_0. The configuration space for a

subset X of Z^2 is $\Omega_X = (\Omega_0)^X$ and the configuration space for the thermodynamic (infinite) system on the whole lattice is $\Omega = (\Omega_0)^{Z^2}$. If $X \cap Y = \varnothing$ and $\omega_X \in \Omega_X$, $\omega_Y \in \Omega_Y$, we denote by $\omega_X \times \omega_Y$ the configuration on $X \cup Y$ that coincides with ω_X on X and with ω_Y on Y. If ω is any configuration on X and $Y \subset X$ we denote the restriction of ω to Y by ω_Y.

Viewing Ω_0 as a discrete metric space observables of the system can be identified with real-valued elements of $C(\Omega)$ [respectively, $C(\Omega_X)$ in case of a finite subsystem], where $C(\Omega)$ denotes the continuous functions on Ω.

On Ω_0 we take as *a priori* measure the (unnormalized) counting measure μ_0. The product measures on Ω_X will be denote by μ_0 as well. Integration with respect to μ_0 will be denoted by the symbol $\langle \cdot \rangle_0$.

A (macro-) state of the system (any specific configuration may be viewed as a microstate) is a positive, linear, normalized functional on $C(\Omega)$. By restriction to $C(\Omega_X)$, X finite, it defines a density function $\rho[X]$ such that for all $f \in C(\Omega_X)$

$$\rho(f) = \langle f \cdot \rho[X] \rangle_0 \tag{1}$$

The set of density functions $\{\rho[X], X \subset Z^2, |X| < \infty\}$ (here $|X|$ denotes the number of sites in X) defined by a state ρ obeys the following: (i) normalization conditions: for all X,

$$\langle \rho[X] \rangle_0 = \sum_{\omega_X \in \Omega_X} \rho[X](\omega_X) = 1 \tag{2}$$

and (ii) compatibility conditions: for all Y and Z with $Y \cap Z = X \neq \varnothing$,

$$\sum_{\omega_{Y \setminus X}} \rho[Y](\omega_X \times \omega_{Y \setminus X}) = \sum_{\omega_{Z \setminus X}} \rho[Z](\omega_X \times \omega_{Z \setminus X}) \tag{3}$$

for all ω_X.

Since lattice translations induce translations on $C(\Omega)$ and on the set of states in a natural way, a state may be translation invariant, and the set of translation-invariant states will be denoted by I. We shall also frequently use the notion of "local translational invariance": a state ρ_X on $C(\Omega_X)$ is locally translation invariant (l.t.i.) if and only if

$$\rho_X(f) = \rho_X(\tau f)$$

for all $f \in C(\Omega_X)$ and all translations τ such that $\tau f \in C(\Omega_X)$. The set of locally translation-invariant states on $C(\Omega_X)$ will be denoted by I_X.

The Hamiltonian for a finite, nonempty subset Λ of Z^2 is

$$H[\Lambda] = \sum_{X \subset \Lambda} \Phi[X] \tag{4}$$

where $\Phi[X]$ is the potential function associated with the cluster (set of lattice points) X. The interaction Φ will be taken translation invariant and of finite range.

In the next section we shall impose some more restrictions on the interaction. Specifically we shall require some reflection symmetry: let Θ_l be the reflection in a line l in Z^2 such that the lattice (considering Z^2 as imbedded in R^2) is mapped onto itself by Θ_l. The induced transformations of Ω and of $C(\Omega)$ will be denoted by Θ_l as well. We shall call the interaction symmetric with respect to l if

$$\Phi[X](\omega_X) = \Phi[\Theta_l X](\Theta_l \omega_X) \tag{5}$$

for all clusters X and configurations ω_X.

Translation-invariant equilibrium states (we shall call them simply equilibrium states in this paper) may be characterized by the variational principle[18,19]

$$f = \min_{\rho \in I} \{\rho(e) - s(\rho)\} \tag{6}$$

Here e is an observable representing the mean energy (energy per site) in any translation-invariant state;

$$s(\rho) = \lim_{\Lambda \to Z^2} \frac{S_\rho[\Lambda]}{|\Lambda|} \tag{7}$$

is the mean entropy of the state ρ, and $\beta = (kT)^{-1}$ has been absorbed into the interaction. f is the mean free energy of the system in equilibrium.

The entropy of the finite subsystem in Λ in the state ρ, $S_\rho[\Lambda]$, is given by

$$S_\rho[\Lambda] = -\rho(\log \rho[\Lambda]) = -\langle \rho[\Lambda] \log \rho[\Lambda] \rangle_0 \tag{8}$$

$S_\rho[\Lambda]$ is an increasing, strongly subadditive function of Λ and a concave function of ρ. $s(\rho)$ is a w^*-upper semicontinuous and affine function of $\rho \in I$. Moreover one has for $\rho \in I$ that for most sets $X \subset Y$

$$S_\rho[Y] - S_\rho[X] \geqslant |Y \setminus X| \cdot s(\rho) \tag{9}$$

(cf. Ref. 18, p. 47; Ref. 19, p. 40). We shall make frequent use of Eq. (9) or its analog for infinite sublattices of Z^2.

In order to describe the cluster-variation formalism we introduce a cluster expansion for the mean entropy[20,21,15]: By defining the Möbius transform $q[\cdot]$ of $\log \rho[\cdot]$ via[22]

$$\begin{aligned} \log \rho[X] &= \sum_{Y \subset X} q[Y] \\ q[X] &= \sum_{Y \subset X} (-1)^{|X \setminus Y|} \log \rho[Y] \end{aligned} \tag{10}$$

we may express the mean entropy as the formal series[15]

$$\begin{aligned} s(\rho) &= -\sum_{X \ni 0} \frac{\rho(q[X])}{|X|} \\ &= -\sum_{X}^{*} \rho(q[X]) \\ &= -\sum_{X}^{*} \langle \rho[X]\, q[X] \rangle_0 \end{aligned} \tag{11}$$

Here $\sum^*$ denotes summation over all clusters X such that no two clusters are translates of one another; in other words each equivalence class with respect to translations contributes one term to the summation. Since the mean energy e may be chosen[18]

$$e = \sum_{X}^{*} \Phi[X] \tag{12}$$

the variational principle Eq. (6) may be written

$$f = \min_{\rho \in I} \phi(\rho) \tag{13}$$

with

$$\phi(\rho) = \sum_{X}^{*} \langle \rho[X](\Phi[X] + q[X]) \rangle_0 \tag{14}$$

Owing to the finite range of the interaction there is only a finite number of nonzero terms $\Phi[X]$; however, in general the number of nonzero terms $q[X]$ and $\langle \rho[X] \cdot q[X] \rangle_0$ will be infinite.

In the CVM formalism one introduces a first approximation by truncating the formal series for the mean entropy. Clusters X corresponding to terms retained in the summation are often called "preserved clusters." CVM approximations may be labeled by the set U of preserved clusters, and the object function for the minimization procedure is in the U approximation,

$$\phi_U(\rho) = \sum_{X : X \in U}^{*} \langle \rho[X](\Phi[X] + q[X]) \rangle_0 \tag{15}$$

Since $\phi_U(\rho)$ only depends on the (in general finite number of) density functions $\rho[X]$ for which $X \in U$ a second approximation is now introduced: the minimum of ϕ_U is determined, not by variation of $\rho \in I$, but by variation of the $\rho[X]$, $X \in U$, taking into account the normalization conditions [equation (2)] and the compatibility conditions [equation (3)] that link them, as well as the requirement of local translational invariance for each, but neglecting those compatibility conditions that involve clusters $\notin U$. This may constitute an additional approximation since whereas every $\rho \in I$ determines a compatible set $\{\rho[X], X \in U\}$ the converse may not necessarily be true. This means that the variation in the CVM may include nonphysical local states, i.e., sets of density functions $\rho[X]$, $X \in U$ that cannot be obtained from a thermodynamic state $\rho \in I$.

Neglecting all compatibility conditions that involve clusters $X \notin U$ is equivalent to the following "compatibility assumption": it is assumed that the set of density functions $\{\bar{\rho}[X], X \in U\}$ that results from the CVM minimization procedure admits an extension to a state $\bar{\rho} \in I$, in other words that the local states $\bar{\rho}_X$ on $C(\Omega_X)$ are compatible with a global state $\bar{\rho}$ on $C(\Omega)$. If the compatibility assumption is valid, then the neglect of the above-mentioned compatibility conditions is justified; if it is not valid, however, the CVM approximation results in a set of nonphysical density functions and the relevance of the approximation is doubtful.

An analysis of a specific CVM approximation thus involves an analysis of the following extension problem: given a set of density functions $\{\rho[X], X \in U\}$ that are normalized, compatible, and locally translation invariant, is there a corresponding translation-invariant state ρ on $C(\Omega)$?

3. THE "*C* HIERARCHY" OF THE CVM

We introduce some notation for subsets of Z^2 which we shall use in the sequel:

$$L_n^{k,i} = \{z = (z_1, z_2) \in Z^2 \mid z_1 \in \{k, \ldots, k+n-1\}, z_2 = i\} \tag{16}$$

$$L^i = \{z = (z_1, z_2) \in Z^2 \mid z_2 = i\} \tag{17}$$

$$D_n^{k,i} = \{z = (z_1, z_2) \in Z^2 \mid z_1 \in \{k, \ldots, k+n-1\}, z_2 \in \{i, i+1\}\} \tag{18}$$

$$D^i = \{z = (z_1, z_2) \in Z^2 \mid z_2 \in \{i, i+1\}\} \tag{19}$$

(respectively, *l*ines and *d*ouble lines). In most cases the position of a specific set in the lattice will be irrelevant owing to translational invariance; in those cases we shall denote *any* translate of one of the sets defined above by the same symbol but omit the superscripts k, i.

As to the interaction Φ we shall require

$$\Phi[X] = 0 \qquad \text{unless} \quad X \subset D_2 \tag{20}$$

Here D_2, in accordance with equation (18) and the remark above, denotes any square of four nearest-neighbor pairs in Z^2. As a consequence of equation (20) the observable for the mean energy e may be taken to be an element of $C(\Omega_{D_2})$.

Moreover we shall require some reflection symmetry, as announced in the previous section: we shall require Φ to be symmetric with respect to the coordinate axes. Owing to the translational invariance of Φ this implies symmetry with respect to all lines parallel to these axes.

In the C hierarchy of the CVM the mean entropy of a translation-invariant state ρ is approximated by[17]

$$\begin{aligned} C_n(\rho) = &\{S_\rho[D_n] - S_\rho[D_{n-1}]\} \\ &- \{S_\rho[L_n] - S_\rho[L_{n-1}]\} \end{aligned} \tag{21}$$

The corresponding variational problem is

$$f_n^C = \min_{\rho \in I_{D_n}} \{\rho(e) - C_n(\rho)\} \tag{22}$$

Here the minimum is sought among the locally translation-invariant states on $C(\Omega_{D_n})$. This approximation may be derived from the general setup of the CVM by choosing as the set of preserved clusters D_n and all its subsets. In this section we present an alternative derivation of the approximation equation (22) that clarifies the nature of this approximation.

We start by restating some results that have been published previously.[16]

Definition I. For any state $\rho \in I$ or $\rho \in I_D$,

$$\text{(i)} \quad s_1(\rho) = \lim_{n \to \infty} \frac{S_\rho[L_n]}{n}$$

$$\text{(ii)} \quad s_2(\rho) = \lim_{n \to \infty} \frac{S_\rho[D_n]}{2n}$$

$$\text{(iii)} \quad b(\rho) = 2s_2(\rho) - s_1(\rho)$$

Lemma I.[16] For any $\rho \in I$ or I_D

$$\lim_{n \to \infty} C_n(\rho) = b(\rho)$$

Lemma II.[16] For any $\rho \in I_D$ there exists a sequence $(\hat{\rho}_n)$ in I such that

$$\text{(i)} \quad \lim_{n \to \infty} s(\hat{\rho}_n) = b(\rho)$$

$$\text{(ii)} \quad \lim_{n \to \infty} \hat{\rho}_n(e) = \rho(e)$$

$$\text{(iii)} \quad \lim_{n \to \infty} \hat{\rho}_n[A] = \rho[A]$$

for any finite $A \subset D$.

Lemma II implies the following extension theorem:

Theorem I. For any $\rho \in I_D$ there is a $\hat{\rho} \in I$ such that

(i) $\hat{\rho}$ is an extension of ρ

(ii) $s(\hat{\rho}) = b(\rho)$

Proof. Let $(\hat{\rho}_n)$ be the sequence in I corresponding to $\rho \in I_D$ as given by Lemma II. There is at least a subsequence of $(\hat{\rho}_n)$ that converges in w^* sense to a state $\hat{\rho} \in I$ (since I is w^* compact; alternatively use Proposition 1.4 of Ref. 18). That $\hat{\rho}$ is an extension of ρ is a consequence of Lemma II(iii) and the continuity of $\hat{\rho}$ and ρ. As to the entropy of $\hat{\rho}$, by the upper semicontinuity of s

$$s(\hat{\rho}) \geqslant \lim_{n \to \infty} s(\hat{\rho}_n) = b(\rho) \tag{23}$$

Since $\hat{\rho}$ is an extension of ρ we have $b(\hat{\rho}) = b(\rho)$, so

$$s(\hat{\rho}) \geqslant b(\hat{\rho}) \tag{24}$$

On the other hand, since for any state $\hat{\rho} \in I$

$$S_{\hat{\rho}}[D_n] - S_{\hat{\rho}}[L_n] \geqslant n \cdot s(\hat{\rho}) \tag{25}$$

[cf. Eq. (9)], it follows from Definition I that for any $\hat{\rho} \in I$

$$b(\hat{\rho}) \geqslant s(\hat{\rho}) \tag{26}$$

Combining Eqs. (24) and (26) establishes that $s(\hat{\rho}) = b(\hat{\rho}) = b(\rho)$, which completes the proof of Theorem I. ∎

For our next theorem we consider a special class of locally translation-invariant states on $C(\Omega_{D_n})$, namely, "symmetric" ones. Let Θ_i denote reflec-

tion in the line $x_2 = i + 1/2$ (here we view Z^2 as imbedded in R^2); so Θ_i interchanges the lower and the upper line of D^i. The induced transformations of configurations, observables and states will be denoted by Θ_i as well. Often we shall omit the location index i. The set of locally translation-invariant states on $C(\Omega_{D_n})$ that are invariant under Θ ("symmetric") will be denoted by $I^s_{D_n}$.

Crucial in our analysis of the C hierarchy is the following extension theorem.

Theorem II. For any $\rho \in I^s_{D_n}$ there is a state $\hat{\rho} \in I_D$ such that

(i) $\hat{\rho}$ is an extension of ρ

(ii) $b(\hat{\rho}) = S_{\hat{\rho}}[D_n] - S_{\hat{\rho}}[D_{n-1}] - s_1(\hat{\rho})$
$\geqslant C_n(\rho)$

Proof. Let $\rho \in I^s_{D_n}$ and consider the associated density functions $\rho[D_n]$ and $\rho[D_{n-1}]$. Using these functions we define density functions $\hat{\rho}[D_{n+k}]$ on sets D_{n+k} for $k = 1, 2, \ldots$ by

$$\hat{\rho}[D_{n+k}](\omega) = \frac{\prod_{l=0}^{k} \rho[D_n](\omega_{D_n^{i+l,j}})}{\prod_{l=1}^{k} \rho[D_{n-1}](\omega_{D_{n-1}^{i+l,j}})} \tag{27}$$

for all $\omega \in \Omega_{D^{i,j}_{n+k}}$, with i and j arbitrary. For configurations for which the denominator is zero the numerator is zero as well and we define the quotient to be zero. One easily verifies that $\hat{\rho}[D_{n+k}]$ is a well-defined, properly normalized density function and that the set of $\hat{\rho}[D_{n+k}]$ is compatible: $\hat{\rho}[D_{n+k+1}]$ may be reduced to $\hat{\rho}[D_{n+k}]$ by summation over the spins in $D_{n+k+1} \backslash D_{n+k}$. Thus the set of $\hat{\rho}[D_{n+k}]$ defines a state $\hat{\rho}$ on $C(\Omega_D)$. By construction $\hat{\rho}$ is invariant for translations in the δ_1 direction and it is an extension of ρ. To show that $\hat{\rho} \in I_D$ it remains to prove that its restrictions to the lower and the upper line coincide (are isomorphic). This now follows trivially from the fact that $\hat{\rho}$ inherits the reflection symmetry of ρ. Hence $\hat{\rho} \in I^s_D \subset I_D$.

The extension procedure defined by Eq. (27) will be referred to as Markov extension, for obvious reasons.

Now we shall calculate $b(\hat{\rho})$. From Eq. (27) we find

$$2s_2(\hat{\rho}) = \lim_{m \to \infty} \frac{(m-n+1)\, S_\rho[D_n] - (m-n)\, S_\rho[D_{n-1}]}{m}$$
$$= S_\rho[D_n] - S_\rho[D_{n-1}] \tag{28}$$

Furthermore,

$$\begin{aligned} s_1(\hat{\rho}) &\leqslant \{S_{\hat{\rho}}[L_n] - S_{\hat{\rho}}[L_{n-1}]\} \\ &= \{S_{\rho}[L_n] - S_{\rho}[L_{n-1}]\} \end{aligned} \tag{29}$$

[cf. equation (9)]. Hence,

$$\begin{aligned} b(\hat{\rho}) &= 2s_2(\hat{\rho}) - s_1(\hat{\rho}) \\ &= S_{\hat{\rho}}[D_n] - S_{\hat{\rho}}[D_{n-1}] - s_1(\hat{\rho}) \\ &\geqslant C_n(\rho) \end{aligned}$$

This completes the proof of Theorem II. ∎

By combining Theorems I and II we have the following:

Theorem III. For any $\rho \in I^s_{D_n}$ there is a $\hat{\rho} \in I$ such that

(i) $\hat{\rho}$ is an extension of ρ

(ii) $$\begin{aligned} s(\hat{\rho}) &= S_{\hat{\rho}}[D_n] - S_{\hat{\rho}}[D_{n-1}] - s_1(\hat{\rho}) \\ &\geqslant C_n(\rho) \end{aligned}$$

Let M_n denote the subset of I consisting of states that may be obtained from a state of $I^s_{D_n}$ by the construction outlined above ("Markov extensions"). A state $\rho \in M_n$ may thus be constructed from its restriction to $C(\Omega_{D_n})$, or, equivalently, from its density function $\rho[D_n]$. For the entropy of such a state

$$\begin{aligned} s(\rho) = b(\rho) &= 2s_2(\rho) - s_1(\rho) \\ &= S_{\rho}[D_n] - S_{\rho}[D_{n-1}] - s_1(\rho) \geqslant C_n(\rho) \end{aligned} \tag{30}$$

Notice that for all n

$$\varnothing \neq M_n \subset M_{n+1} \subset \cdots \subset I^s \subset I \tag{31}$$

Now we are in a position to introduce an approximation to the variational principle Eq. (6) in the "standard" fashion, namely, by restricting the variation to the subset M_n of I:

$$f^1_n = \min_{\rho \in M_n} \{\rho(e) - s(\rho)\} \tag{32}$$

Obviously,

$$f^1_n \geqslant f^1_{n+1} \geqslant \cdots \geqslant f \tag{33}$$

In order to be able to approximate the entropy of a state $\rho \in M_n$ by a finite computation $s(\rho)$ is replaced by the lower bound provided by Eq. (30); after this additional approximation we have the approximate variational principle

$$f_n^2 = \min_{\rho \in M_n} \{\rho(e) - C_n(\rho)\} \tag{34}$$

Then

$$f_n^2 \geqslant f_n^1 \tag{35}$$

Lemma III. If $e = \Theta e$ then

$$f_n^2 = f_n^C$$

Proof. By definition

$$f_n^C = \min_{\rho \in I_{D_n}} \{\rho(e) - C_n(\rho)\} \tag{36}$$

By the definition of M_n and Theorem III the definition Eq. (34) of f_n^2 is equivalent to

$$f_n^2 = \min_{\rho \in I_{D_n}^s} \{\rho(e) - C_n(\rho)\} \tag{37}$$

So

$$f_n^C \leqslant f_n^2 \tag{38}$$

Let now $\bar{\rho} \in I_{D_n}$ be such that

$$f_n^C = \bar{\rho}(e) - C_n(\bar{\rho}) \tag{39}$$

(Such a $\bar{\rho}$ exists since f_n^C is the minimum of a continuous function on a compact set.)

Let $\tilde{\rho}$ be the Markov extension of $\bar{\rho}$ to $C(\Omega_D)$ as defined by Eq. (27). By construction $\tilde{\rho}$ is invariant for translations in the δ_1 (horizontal) direction; however, if $\bar{\rho}$ is not symmetric it is not obvious that $\tilde{\rho}$ is symmetric or even locally translation invariant for translations in the δ_2 (vertical) direction. Thus define

$$\hat{\rho} = \tfrac{1}{2}(\tilde{\rho} + \Theta\tilde{\rho}) \tag{40}$$

Then $\hat{\rho} \in I_D^s$ and since $e = \Theta e$

$$\hat{\rho}(e) = \tilde{\rho}(e) = \bar{\rho}(e) \tag{41}$$

Since $s_2(\cdot)$ is affine

$$s_2(\hat{\rho}) = s_2(\tilde{\rho}) = \tfrac{1}{2}\{S_{\tilde{\rho}}[D_n] - S_{\tilde{\rho}}[D_{n-1}]\} \tag{42}$$

so [cf. equation (9)]

$$S_{\hat{\rho}}[D_n] - S_{\hat{\rho}}[D_{n-1}] \geqslant 2s_2(\hat{\rho}) = S_{\bar{\rho}}[D_n] - S_{\bar{\rho}}[D_{n-1}] \tag{43}$$

For any observable $f \in C(\Omega_{L_n^{k,i}})$, $i = 0, 1$ we have $\Theta_0 f \in C(\Omega_{L_n^{k,1-i}})$ and

$$\begin{aligned}\hat{\rho}(f) &= \tfrac{1}{2}\tilde{\rho}(f) + \tfrac{1}{2}\tilde{\rho}(\Theta_0 f) \\ &= \tfrac{1}{2}\bar{\rho}(f) + \tfrac{1}{2}\bar{\rho}(\Theta_0 f) \\ &= \bar{\rho}(f)\end{aligned}$$

since $\bar{\rho}$ is locally translation invariant on $D_n^{k,0}$; hence the restriction of $\hat{\rho}$ to $C(\Omega_{L_n})$ is equal to the restriction of $\bar{\rho}$ to $C(\Omega_{L_n})$; in other words,

$$\hat{\rho}[L_n] = \bar{\rho}[L_n] \tag{44}$$

Hence

$$S_{\hat{\rho}}[L_n] - S_{\hat{\rho}}[L_{n-1}] = S_{\bar{\rho}}[L_n] - S_{\bar{\rho}}[L_{n-1}] \tag{45}$$

Combining Eqs. (39), (41), (43), and (45) we find

$$f_n^C \geqslant \hat{\rho}(e) - C_n(\hat{\rho}) \geqslant f_n^2 \tag{46}$$

Equations (38) and (46) establish the lemma. ∎

We have thus shown that the C hierarchy of the CVM, which is usually derived via truncation of a formal cluster expansion for the mean entropy, may also be derived from the variational principle by restriction of the variation to a suitably chosen subset of the set of all translation-invariant states, followed by an approximation that serves to reduce the calculation of the entropy contribution to one that can be done in a finite computation.

The condition that the energy observable must be symmetric ($e = \Theta e$) is not a restriction; for symmetric Φ, e may always be written in a symmetric form.

Notice, by the way, that we have four equivalent expressions for the C approximation:

$$f_n^C = \min_{\rho \in M_n} \{\rho(e) - C_n(\rho)\} \tag{47a}$$

$$f_n^C = \min_{\rho \in I_{D_n}^s} \{\rho(e) - C_n(\rho)\} \tag{47b}$$

$$f_n^C = \min_{\rho \in I_{D_n}} \{\rho(e) - C_n(\rho)\} \tag{47c}$$

$$f_n^C = \min_{\rho \in I} \{\rho(e) - C_n(\rho)\} \tag{47d}$$

Equation (47d) follows from the fact that on the one hand $M_n \subset I$ and on the other hand the restriction to $C(\Omega_{D_n})$ of a state $\rho \in I$ belongs to I_{D_n}.

The alternative derivation of the approximations of the C hierarchy presented above serves to prove the following:

Theorem IV.

$$\text{(i)} \quad f_n^C \geqslant f$$

$$\text{(ii)} \quad f_n^C \geqslant f_{n+1}^C \qquad \text{for all} \quad n \geqslant 2$$

$$\text{(iii)} \quad \lim_{n \to \infty} f_n^C = f$$

Proof. Proposition (i) follows from Eqs. (33), (35), and Lemma III. As to proposition (ii),

$$f_n^C = \min_{\rho \in M_n} \{\rho(e) - C_n(\rho)\}$$

$$\geqslant \min_{\rho \in M_n} \{\rho(e) - C_{n+1}(\rho)\} + \min_{\rho \in M_n} \{C_{n+1}(\rho) - C_n(\rho)\}$$

Since $M_n \subset M_{n+1}$ the first term is

$$\geqslant \min_{\rho \in M_{n+1}} \{\rho(e) - C_{n+1}(\rho)\} = f_{n+1}^C$$

As to the second term, for $\rho \in M_n$

$$S_\rho[D_{n+1}] - S_\rho[D_n] = S_\rho[D_n] - S_\rho[D_{n-1}]$$

[cf. equation (27)]. Hence for $\rho \in M_n$

$$C_{n+1}(\rho) - C_n(\rho)$$

$$= \{S_\rho[L_n] - S_\rho[L_{n-1}]\} - \{S_\rho[L_{n+1}] - S_\rho[L_n]\}$$

which is $\geqslant 0$ by the strong subadditivity of the entropy. Consequently,

$$f_n^C \geqslant f_{n+1}^C$$

As to proposition (iii), from (i) and (ii) it follows that

$$f_\infty^C = \lim_{n \to \infty} f_n^C$$

exists and that

$$f_{\infty}^{C} \geqslant f \tag{48}$$

Now let $\bar{\rho} \in I$ be an equilibrium state, thus

$$f = \bar{\rho}(e) - s(\bar{\rho}) \tag{49}$$

Then

$$\begin{aligned} f_{\infty}^{C} &= \lim_{n \to \infty} \min_{\rho \in I_{D_n}} \{\rho(e) - C_n(\rho)\} \\ &\leqslant \lim_{n \to \infty} \{\bar{\rho}(e) - C_n(\bar{\rho})\} \\ &= \bar{\rho}(e) - b(\bar{\rho}) \quad \text{(Lemma I)} \\ &= \bar{\rho}(e) - s(\bar{\rho}) \quad \text{(Ref. 16, Corollary I)} \\ &= f \end{aligned} \tag{50}$$

Equations (48) and (50) establish $f_{\infty}^{C} = f$, which completes the proof of the theorem. ∎

The above theorem establishes that the C hierarchy of the CVM provides a monotonically decreasing and converging sequence of upper bounds f_n^C on the mean free energy f.

Our next theorem shows how the states that minimize the approximate expression for the free energy functional in the C-hierarchy approximate equilibrium states.

Theorem V. Let $\bar{\rho}_n \in I_{D_n}^s$ be such that $f_n^C = \bar{\rho}_n(e) - C_n(\bar{\rho}_n)$. Let $\hat{\rho}_n \in I$ be the extension of $\bar{\rho}_n$ as given by Theorem III. Let $\bar{\rho}$ be a w^*-limit point of $(\hat{\rho}_n)$. Then $\bar{\rho}$ is an equilibrium state.

Proof. Obviously $\bar{\rho} \in I$. Now

$$\bar{\rho}(e) = \lim_{n \to \infty} \hat{\rho}_n(e) = \lim_{n \to \infty} \bar{\rho}_n(e)$$

and

$$s(\bar{\rho}) \geqslant \lim_{n \to \infty} s(\hat{\rho}_n) \geqslant \lim_{n \to \infty} C_n(\bar{\rho}_n)$$

Hence

$$\begin{aligned} f &= \min_{\rho \in I} \{\rho(e) - s(\rho)\} \\ &\leqslant \bar{\rho}(e) - s(\bar{\rho}) \\ &\leqslant \lim_{n \to \infty} \{\bar{\rho}_n(e) - C_n(\bar{\rho}_n)\} \\ &= \lim_{n \to \infty} f_n^C \\ &= f \end{aligned}$$

Hence $f = \bar{\rho}(e) - s(\bar{\rho})$, so $\bar{\rho}$ is an equilibrium state.

Corollary I. If there is a unique equilibrium state $\bar{\rho}$, then

$$\hat{\rho}_n \xrightarrow{w^*} \bar{\rho}$$

(with $\hat{\rho}_n$ as in Theorem V).

Proof. The above statement follows trivially from Theorem V and the fact that I is w^* compact. ∎

4. THE BETHE APPROXIMATION

In this section we use the results of Section 3 to throw some light on the Bethe approximation. The Bethe or Bethe–Peierls or quasichemical approach tries to take into account nearest-neighbor correlations and may be derived from the general setup of the CVM by choosing as preserved clusters only single points and nearest-neighbor pairs.

In this section we shall use the notation n for any nearest-neighbor pair of sites in the lattice Z^2 and p for any single site.

We have the following extension theorem.

Theorem VI. Let ρ be a state on $C(\Omega_n)$ such that (i) ρ is locally translation invariant, (ii) ρ is symmetric, i.e., invariant for the transformation that interchanges the two sites of n.

Then there exists a state $\hat{\rho} \in I$ such that (i) $\hat{\rho}$ is an extension of ρ, in the sense that its restriction to *each* nearest-neighbor pair coincides with ρ, and

$$\text{(ii)} \quad s(\hat{\rho}) \geqslant 2S_\rho[n] - 3S_\rho[p]$$

Proof. The proof is again by construction: denote any square consisting of four nearest-neighbor pairs in Z^2 by D_2 [in accordance with Eq. (18)]. For any square D_2 we number the sites from 1 to 4 counted

counterclockwise, taking the one in the lower left corner (the first one in lexicographical order) as number one for definiteness. A subset of D_2 will be denoted by enumeration of the corresponding numbers. As a first step we extend ρ to triangles: define density functions for triangles $\{124\}$ and $\{234\}$ by

$$\begin{aligned}\tilde{\rho}[124](\sigma_1, \sigma_2, \sigma_4) &= \frac{\rho[n](\sigma_1, \sigma_2) \cdot \rho[n](\sigma_1, \sigma_4)}{\rho[p](\sigma_1)} \\ \tilde{\rho}[234](\sigma_2, \sigma_3, \sigma_4) &= \frac{\rho[n](\sigma_2, \sigma_3) \cdot \rho[n](\sigma_3, \sigma_4)}{\rho[p](\sigma_3)}\end{aligned} \tag{51}$$

(As always, the quotient is defined as zero for configurations for which the denominator is zero.) Equation (51) defines an extension of ρ to these triangles, as may be checked by trivial computation. Now notice that

$$\begin{aligned}&\sum_{\sigma_1} \tilde{\rho}[124](\sigma_1, \sigma_2, \sigma_4) \\ &\quad = \sum_{\sigma_1} \frac{\rho[n](\sigma_1, \sigma_2) \cdot \rho[n](\sigma_1, \sigma_4)}{\rho[p](\sigma_1)} \\ &\quad = \sum_{\sigma_1} \frac{\rho[n](\sigma_2, \sigma_1) \cdot \rho[n](\sigma_1, \sigma_4)}{\rho[p](\sigma_1)} \\ &\quad = \sum_{\sigma_3} \frac{\rho[n](\sigma_2, \sigma_3) \cdot \rho[n](\sigma_3, \sigma_4)}{\rho[p](\sigma_3)} \\ &\quad = \sum_{\sigma_3} \tilde{\rho}[234](\sigma_2, \sigma_3, \sigma_4)\end{aligned} \tag{52}$$

where we used the symmetry of ρ.

Hence the two density functions defined by Eq. (51) are compatible and we may define a further extension of ρ to $C(\Omega_{D_2})$ via (in obvious shorthand notation)

$$\tilde{\rho}[1234] = \frac{\tilde{\rho}[124] \cdot \tilde{\rho}[234]}{\tilde{\rho}[24]} \tag{53}$$

where $\tilde{\rho}[24]$ is unambiguously defined, as established by Eq. (52). This defines a state $\tilde{\rho}$ on $C(\Omega_{D_2})$ which has ρ its restriction to each nearest-neighbor pair contained in D_2 and is thus an element of I_{D_2}. Now we want to use Theorem III. Recalling the definition of the reflection Θ from Section 3, we define

$$\hat{\rho} = \tfrac{1}{2}(\tilde{\rho} + \Theta\tilde{\rho}) \tag{54}$$

($\Theta\tilde{\rho}$ may be constructed in the same way as $\tilde{\rho}$, starting with extension to triangles $\{123\}$ and $\{134\}$.)

Owing to the presupposed symmetry of ρ, $\hat{\rho}$, too, has the property that its restriction to each nearest-neighbor pair in D_2 coincides with ρ. Since $\hat{\rho} \in I^s_{D_2}$, Theorem III now guarantees the existence of a further, translation-invariant extension to the whole of Z^2.

As to the entropy of $\hat{\rho}$, Theorem III states

$$s(\hat{\rho}) = S_{\hat{\rho}}[D_2] - S_{\hat{\rho}}[D_1] - s_1(\hat{\rho}) \tag{55}$$

Now

$$\begin{aligned} S_{\hat{\rho}}[D_2] &\geqslant \tfrac{1}{2}S_{\tilde{\rho}}[D_2] + \tfrac{1}{2}S_{\Theta\tilde{\rho}}[D_2] \\ &= S_{\tilde{\rho}}[D_2] \\ &= S_{\tilde{\rho}}[124] + S_{\tilde{\rho}}[234] - S_{\tilde{\rho}}[24] \\ &= 4S_{\rho}[n] - 2S_{\rho}[p] - S_{\hat{\rho}}[24] \end{aligned} \tag{56}$$

From the subadditivity of $S_{\tilde{\rho}}[\cdot]$

$$S_{\tilde{\rho}}[24] \leqslant S_{\tilde{\rho}}[2] + S_{\tilde{\rho}}[4] = 2S_{\rho}[p] \tag{57}$$

So

$$S_{\hat{\rho}}[D_2] \geqslant 4S_{\rho}[n] - 4S_{\rho}[p] \tag{58}$$

Noting that D_1 is a nearest-neighbor pair n and that

$$s_1(\hat{\rho}) \leqslant S_{\hat{\rho}}[n] - S_{\hat{\rho}}[p] = S_{\rho}[n] - S_{\rho}[p] \tag{59}$$

[cf. Eq. (9)], we find from Eqs. (55)–(59)

$$s(\hat{\rho}) \geqslant 2S_{\rho}[n] - 3S_{\rho}[p]$$

which completes the proof of this theorem. ∎

Remark. Note that the above theorem pertains to an isotropic situation where horizontal and vertical nearest-neighbor pairs are equivalent. For the anisotropic case the above construction only works if $|\Omega_0| = 2$, i.e., for the lattice gas or spin-$\frac{1}{2}$ models. Otherwise the compatibility expressed by Eq. (52) does not hold (as can be shown by explicit calculation).

Let us now call states in I that may be obtained from a density function for a pair of nearest-neighbor sites in the manner of Theorem VI

"Bethe states." The set of Bethe states will be denoted by B. From the proof of Theorem VI it follows that [cf. Eq. (31)]

$$B \subset M_2 \tag{60}$$

We introduce an approximation to the variational principle Eq. (6) by restricting the variation to the set of Bethe states and replacing the entropy contribution to the free energy by the lower bound provided by Theorem VI:

$$f_B = \min_{\rho \in B} \{\rho(e) - 2S_\rho[n] + 3S_\rho[p]\} \tag{61}$$

Then

$$f_B \geqslant f \tag{62}$$

In the case of symmetric, isotropic nearest-neighbor interactions the variational problem Eq. (61) corresponds precisely to the Bethe–Peierls approximation on Z^2. Equation (61) essentially provides the generalization of the traditional Bethe approximation to models with an interaction that extends beyond nearest neighbor. However, there is the limitation that $e \in C(\Omega_{D_2})$ [cf. Eq. (20)].

The first approximation of the C hierarchy, based on the square D_2 as basic cluster, is exactly the Kramers–Wannier approximation.[4.17] Equations (58) and (60) imply

$$f_B \geqslant f_{KW} \tag{63}$$

On the other hand, since Bethe states include product states (mean-field states) and for a mean-field state $S_\rho[n] = 2S_\rho[p]$, one finds

$$f_{MF} \geqslant f_B \tag{64}$$

So we established that, as far as the prediction of the mean free energy is concerned, on Z^2 the Kramers–Wannier approach is indeed an improvement over the Bethe approach, which in turn improves upon the mean field result:

$$f_{MF} \geqslant f_B \geqslant f_{KW} \geqslant f \tag{65}$$

The situation is different on the triangular lattice, however. On the one hand all our results for the square lattice have an analog for the triangular lattice, since the latter may be mapped onto Z^2 via the introduction of a suitable nonorthogonal coordinate system. On the other hand, as a consequence of the different topological structure of the triangular lattice, the

Bethe approximation on Z^2 does not correspond to the Bethe approximation on the triangular lattice under this mapping. Owing to the existence in the latter of triangles consisting of three translationally distinct nearest-neighbor pairs, the Bethe approximation on the triangular lattice is an example of a CVM approximation for which the compatibility assumption may be violated: given locally translation-invariant and compatible density functions for nearest-neighbor pairs, there may not exist a translation-invariant extension to the entire lattice. An example is provided by the spin-$\frac{1}{2}$ Ising antiferromagnet: at very strong interaction the equilibrium state as predicted by the Bethe approximation has probability tending to one of finding opposite spins on each nearest-neighbor pair; obviously this state has no extension to triangles (the frustration effect), let alone to the entire lattice. For such models the Bethe approximation is thus an "unphysical" one; it would seem that in those cases the first sensible step beyond mean field is the Kikuchi triangle approximation,[10] in which nearest-neighbor triangles, the nearest-neighbor pairs and single sites are taken as preserved clusters. That approximation may be based upon an extension theorem similar in spirit to the above theorems, thus showing its "sensibility."

5. SUMMARY AND FINAL REMARKS

In this paper we have presented a new derivation of some interesting approximations of the cluster-variation method. The derivation employs an extension theorem to define a subset of the set of all translation-invariant states consisting of states that are obtained from their restriction to a finite part of the lattice, the so-called basic cluster. In the variational principle the variation is then restricted to this subset of admissible states and the entropy of these states is estimated in terms of a lower bound. We used this technique to make some statements about the C hierarchy of the CVM and about the (isotropic) Bethe approximation; however, there are other CVM approximations that may be treated by the same techniques, notably the so-called W hierarchy.[17]

In this paper we restricted our attention to classical lattice models on Z^2 with a translation-invariant interaction that is fairly short-ranged [cf. Eq. (20)] and that is symmetric with respect to reflections in the coordinate axes. These restrictions were motivated by the fact that our primary interest in this investigation was the C hierarchy of the CVM.

Besides satisfying intellectual curiosity our results may have some practical relevance. For instance our derivation of the Bethe approximation shows how to apply this approximation to models in which the interaction

is not limited to nearest-neighbor pairs. Also CVM approximations may be improved by using a better (larger) lower bound on the entropy contribution, which is often easily obtained: again taking the Bethe approximation as an example, one might use Eq. (56) instead of Eq. (58) to provide the bound on the entropy, since the extension to relevant clusters larger than the basic cluster (the nearest-neighbor pair in this case) is known and may be calculated explicity. Thus it may be possible to improve upon a CVM approximation without going to a larger basic cluster.

We want to stress the importance of the compatibility assumption (cf. Section 2) in all approximations of the cluster-variation type, since its influence or even its existence is not always realized. This importance is reflected in the fact that our results are all based on extension theorems that serve to show the validity of the compatibility assumption for the specific approximation at hand. On occasions some CVM approximations have been reported to produce nonsensical results[23,24] instead of improving on an approximation based on a smaller basic cluster (or on a smaller number of basic clusters); this may well be related to violation of the compatibility assumption.

We want to end with a brief discussion of the relation between our approach to cluster-variation approximations and previous formulations of these techniques. It may be helpful to consider the mean field approximation first. It is well known that the mean field results may be derived in a number of ways. Among the possible approaches are the following:

(1) *The effective field approach:* The interactions between sites are replaced by an effective external field, i.e., the Hamiltonian is modified on the basis of physical intuition.

(2) *The combinatorial approach:* In the evaluation of the partition function or "sum over states" the combinatorial factor (i.e., the number of configurations with specified energy) is calculated under the assumption that there are no correlations between different lattice sites.

(3) *The variational approach:* In the variational principle, the variation is restricted to the set of product states.

Cluster-variation and related approximations resulted from attempts to improve on the mean field approximation. Such attempts have been based, among others, on the effective field concept and on the combinatorial approach, but not, to our knowledge, on the variational approach, in spite of the fact that from such a point of view it is most obvious how to improve the approximation: the variation should be restricted to a larger set of states than just the product states. Actually the work reported in this paper resulted from an attempt to derive the Bethe results along these lines. Thus, from the conceptual point of view, our

approach to the approximations differs completely from the effective field or combinatorial approach.

On the more technical level, however, all three formulations are (of course) intimately connected. The original formulation of the CVM[1,25,26] was based on the combinatorial approach. Kikuchi derived his approximate expressions for the combinatorial factor using the following mental picture: one considers an "assembly"[25] of finite lattices, each having a specific spin configuration; one now adds one site to each lattice and tries to assign values to the additional spin variables in such a way that a number of previously selected basic probabilities (e.g., the probability of finding any specific configuration on a pair of nearest-neighbor sites) are unchanged by this step in the construction of the lattice. Obviously, what happens here is an attack on what we called the extension problem (cf. Section 2): one tries to construct a thermodynamic translation-invariant state having prescribed restrictions to the chosen basic clusters. In a way the extension theorems that lie at the heart of our formulation are the formalized mathematical counterpart of this early "assembly method"[1,25,26] for constructing approximate expressions for the combinatorial factor.

Later, Morita reformulated the CVM using the Möbius transformation formalism (cf. Section 2)[20]; the Möbius transformation serves to automate the often complicated counting procedures that are inherent in the assembly method. Morita also showed (e.g., see Refs. 27, 28, and references therein) that the CVM may be formulated in terms of effective fields and effective interactions.

[The above remarks on the CVM pertain to the Bethe approximation as well, since the Bethe approximation may be regarded as one of the CVM approximations (e.g., see Ref. 26).]

Further insight in the connections between our formulation and the traditional ones may be obtained through a paper by Woodbury[29]; Woodbury showed that a number of cluster-variation approximations may be derived from general properties of the entropy functionals $S_\rho[A]$; the notions of conditional entropy, Markov process and strong subadditivity are already implicitly present in his formulation.

From the practical point of view of course all methods for deriving CVM formulas are equivalent: eventually, all derivations result in the same set of equations to be solved. From the theoretical point of view, however, each formulation has its specific advantages. The effective field approach is probably most useful if one is interested in statements about the predicted magnetization, since it provides consistency relations for that quantity. The combinatorial approach is handy to treat series expansions of the partition function.[26] For investigation of the free energy prediction the variational approach as developed in this work is a convenient starting point.

Moreover, an important advantage of our formulation is the fact that it is truly thermodynamic (infinite system) from the start; consequently possible problems associated with the taking of the thermodynamic limit are avoided.

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APPENDIX: A "MINIMAL" RESTRICTED VARIATIONAL PRINCIPLE

In a previous paper[16] we established the existence of an exact variational principle for the mean free energy f which involved variation over I_D instead of I and hence involved only restrictions to $C(\Omega_D)$ of thermodynamic states. This result was based in essence on the existence of a transfer matrix, translated into the language of the variational characterization of equilibrium states. The existence of a transfer matrix is intimately related to the possibility of "Markovian extension." Since in the analysis of the C hierarchy the Markovian extension procedure was used in two orthogonal directions subsequently ("double Markovian extension") it is an interesting and in a way a natural question to ask whether there exists an exact variational principle for the mean free energy based on such double Markovian extension. Such a variational principle will be presented in this appendix.

Consider the following subsets of Z^2 (recalling the definition of $L_n^{k,i}$ [Eq. (16)]:

$$H_n^{k,i} = L_n^{k-n+1,i} \cup L_n^{k,i+1} \tag{A1}$$

$$S_n^{k,i} = H_n^{k,i} \cup H_n^{k+1,i} \tag{A2}$$

$$S^{k,i} = \bigcup_{n=1}^{\infty} S_n^{k,i} \tag{A3}$$

Often we shall omit the location indices k, i to indicate *any* translate of one of the sets defined above. Thus S_n consists of any square D_2 with two "arms," $(n-1)$ points long each.

We shall proceed to prove that the entropy $s(\bar{\rho})$ of an equilibrium state $\bar{\rho}$ may be calculated from its restriction to $C(\Omega_S)$, and that essentially the state may be constructed from this restriction. This then sharpens the result of Theorem I and Corollary I of Ref. 16 where a similar proposition was proved for restriction to the infinitely long double line D.

Definition AI. For any $\rho \in I_S$

$$K_n(\rho) = \{S_\rho[S_n] - S_\rho[H_n]\} - \{S_\rho[L_n] - S_\rho[L_{n-1}]\} \tag{A4}$$

Theorem AI. For any $\rho \in I_S$ there exists a state $\hat{\rho} \in I_D$ such that

(i) $\hat{\rho}$ is an extension of ρ

(ii) $b(\hat{\rho}) = \lim_{n \to \infty} K_n(\rho) \equiv k(\rho)$

Proof. Let ρ_n denote the restriction of $\rho \in I_S$ to $C(\Omega_{S_n})$. Define, for each n, a sequence of density functions for $k = 1, 2, \ldots$ by

$$\hat{\rho}_n\left[\bigcup_{i=-k}^{k} S_n^{l+i,j}\right](\omega) = \frac{\prod_{i=-k}^{k} \rho[S_n](\omega_{S_n^{l+i,j}})}{\prod_{i=-k+1}^{k} \rho[H_n](\omega_{H_n^{l+i,j}})} \tag{A5}$$

for all $\omega \in C(\Omega_{\bigcup_{i=-k}^{k} S_n^{l+i,j}})$, with l and j arbitrary, again defining the quotient as zero for configurations for which the denominator is zero. This sequence of density functions defines a state $\hat{\rho}_n$ on $C(\Omega_D)$. $\hat{\rho}_n$ is, by construction, an extension of ρ_n and translation invariant for horizontal translations. Moreover, from the local translational invariance of ρ_n it follows that $\hat{\rho}_n$ is locally translation invariant in the δ_2 (vertical) direction for all $f \in C(\Omega_{L_n})$.

At least a subsequence of $(\hat{\rho}_n)$ converges w^* to a state $\hat{\rho}$; obviously $\hat{\rho}$ is an extension of ρ and an element of I_D [consider local observables first and extend by continuity to the whole of $C(\Omega_D)$].

As to the entropy of $\hat{\rho}$, we have

$$\begin{aligned} 2s_2(\hat{\rho}_n) &= S_\rho[S_n] - S_\rho[H_n] \\ &= S_{\hat{\rho}}[S_n] - S_{\hat{\rho}}[H_n] \geqslant 2s_2(\hat{\rho}) \end{aligned} \tag{A6}$$

where the equality follows from Eq. (A5) and the inequality from Eq. (9). Taking $n \to \infty$ and using the upper-semicontinuity of $s_2(\cdot)$

$$2s_2(\hat{\rho}) \geqslant \lim_{n \to \infty} \{S_\rho[S_n] - S_\rho[H_n]\} \geqslant 2s_2(\hat{\rho}) \tag{A7}$$

Since furthermore

$$s_1(\hat{\rho}) = \lim_{n\to\infty} \{S_{\hat{\rho}}[L_n] - S_{\hat{\rho}}[L_{n-1}]\}$$
$$= \lim_{n\to\infty} \{S_{\rho}[L_n] - S_{\rho}[L_{n-1}]\} \tag{A8}$$

we find

$$b(\hat{\rho}) = 2s_2(\hat{\rho}) - s_1(\hat{\rho}) = \lim_{n\to\infty} K_n(\rho) \equiv k(\rho)$$

and we have also established the existence of this limit. ▮

Theorem AII. (i) There is the following variational principle for the mean free energy f:

$$f = \min_{\rho\in I_S} \{\rho(e) - k(\rho)\} \tag{A9}$$

(ii) The restriction to $C(\Omega_S)$ of an equilibrium state satisfies Eq. (A9), and each state in I_S that satisfies Eq. (A9) is the restriction to $C(\Omega_S)$ of an equilibrium state.

Proof. From Eq. (9) we have for any $\rho \in I_D$

$$2s_2(\rho) \leqslant S_\rho[S_n] - S_\rho[H_n] \tag{A10}$$

Since

$$s_1(\rho) = \lim_{n\to\infty} \{S_\rho[L_n] - S_\rho[L_{n-1}]\} \tag{A11}$$

it follows that

$$b(\rho) \leqslant k(\rho), \qquad \text{for any} \quad \rho \in I_D \tag{A12}$$

Since [cf. Eq. (26)]

$$s(\rho) \leqslant b(\rho), \qquad \text{for any} \quad \rho \in I \tag{A13}$$

we have

$$s(\rho) \leqslant k(\rho), \qquad \text{for any} \quad \rho \in I \tag{A14}$$

Now let $\bar{\rho} \in I$ be an equilibrium state, thus

$$f = \bar{\rho}(e) - s(\bar{\rho}) \tag{A15}$$

Let $\bar{\rho}_s$ be the restriction of $\bar{\rho}$ to $C(\Omega_S)$. Then

$$\begin{aligned}\min_{\rho \in I_S} \{\rho(e) - k(\rho)\} &\leqslant \bar{\rho}_s(e) - k(\bar{\rho}_s) \\ &= \bar{\rho}(e) - k(\bar{\rho}) \\ &\leqslant \bar{\rho}(e) - s(\bar{\rho}) \\ &= f \end{aligned} \tag{A16}$$

On the other hand, let now $\bar{\rho}_s \in I_S$ be such that

$$\min_{\rho \in I_S} \{\rho(e) - k(\rho)\} = \bar{\rho}_s(e) - k(\bar{\rho}_s) \tag{A17}$$

Denote by $\hat{\rho}$ the extension of $\bar{\rho}_s$ to $C(\Omega)$ for which

$$k(\bar{\rho}_s) = s(\hat{\rho}) \tag{A18}$$

The existence of such a $\hat{\rho} \in I$ is guaranteed by combination of Theorems AI and I. Then

$$\begin{aligned}\min_{\rho \in I_S} \{\rho(e) - k(\rho)\} &= \bar{\rho}_s(e) - k(\bar{\rho}_s) \\ &= \hat{\rho}(e) - s(\hat{\rho}) \\ &\geqslant f \end{aligned} \tag{A19}$$

Equations (A16) and (A19) prove proposition (i). Proposition (ii) then follows from the same equations. ∎

In those cases where coexisting equilibrium states may be distinguished by their expectation values for some observable in $C(\Omega_S)$ (e.g., "magnetization") the above theorem implies that all equilibrium states have the "double Markovian structure" indicated by the proofs of Theorems AI and I, and that they are completely determined by their restriction to S. Since the possibility of restriction to D is the consequence of the existence of a (row-to-row) transfer matrix, Theorem AII then seems to indicate the existence of some stochastic operator that performs a similar role in the orthogonal direction for restrictions of equilibrium states to D.

One may base a sequence of variational approximations on the variational principle Eq. (A9). Such approximations would be related to the approximation scheme proposed by Surda.[30]

Although Theorems AI and AII have been formulated and proved for the square lattice and an interaction that obeys Eq. (20), there are obvious analogs for d-dimensional models with general translation-invariant, finite-range interactions.

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