# Proof of the Existence of the Cluster Free Energy 

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Received December 2, 1983; revised February 28, 1984


#### Abstract

Let $Z_{p}$ be the partition function over internal configurational degrees of freedom of a $p$-particle cluster. The existence of the limit, as $p \rightarrow \infty$, of $(\beta p)^{-1} \log Z_{p}$ is demonstrated for the square, triangle, hexagon, simple cubic, face-centered cubic, and body-centered cubic lattice gas models, and for continuous space models with potentials satisfying stability and tempering conditions. Pairwise, finite-range bonding is assumed throughout.


KEY WORDS: Clusters; metastability; connectedness.

## 1. INTRODUCTION

An object of paramount interest in the theory of metastability and nucleation is the physical cluster, an aggregate of molecules which may grow into a nucleus of the new, thermodynamically stable phase. ${ }^{(1-3)}$ Quantitative predictions of the nucleation rate are based on the Becker-Döring equations, ${ }^{(4)}$ which describe the kinetics of cluster growth. To evaluate the rate constants in these equations, or to determine the cluster size distribution in a vapor, one needs to know the free energy of formation as a function of cluster size.

It is commonly assumed that the asymptotic thermodynamic properties of clusters are well defined and identical to those of the new phase. While it would be surprising if either of these assumptions turned out to be incorrect, neither has been proved for any model. Important preliminary results in this regard are the upper and lower bounds on the cluster free energy established

[^0]by Penrose and Lebowitz ${ }^{(5)}$ for the square lattice gas. In this paper we prove the existence of the limiting cluster free energy density for various lattice gas models, and for continuous space models with potentials satisfying modified stability and tempering conditions.

Given a pairwise bonding criterion depending on the mutual potential energy of two particles (thus on their relative position but not on their momenta), a cluster is defined as a collection of particles connected by a network of bonds. We focus attention on the partition function of a single, isolated cluster. The partition function is evaluated over all internal configurational degrees of freedom, the cluster being centered in a volume large enough to accomodate all connected configurations, but otherwise unspecified. If the idealizations of pairwise, proximity bonding are made (and they usually are in nucleation theory ${ }^{(6)}$ ), the evaluation of such a partition function is all that is required for computing cluster free energies. The above idealizations are justified at the low temperatures and densities that obtain in many nucleation experiments, although one does not expect them to be adequate near the critical point.

The existence of the limiting cluster free energy density is not implied by the well-known results on the existence of the thermodynamic limit for systems of interacting particles. ${ }^{(7-9)}$ In the thermodynamic limit the number of particles $p$ and the volume of the system grow without bound, with the density (or average density) held fixed, while in the cluster problem considered here, the volume must grow as $p^{d}$ (or faster) in $d$ dimensions to encompass all connected configurations. Without the constraint of connectedness, the number of $p$-particle configurations would grow as $\exp$ (const $p^{d}$ ), and so for $d>1$ there would be no limiting free energy density. Through its restriction of configuration space, connectedness plays an essential role in the existence of the cluster free energy.

In Section 2 we extend the Penrose and Lebowitz lower bound on the free energy of clusters in the square lattice gas to other two- and threedimensional lattices. We then state a key lemma and prove the existence of the cluster free energy density for the lattice gas models. In Section 3 we consider continuous space models, and show that for a wide class of potentials, the conditions of the lemma are satisfied, proving the existence of the cluster free energy density for these models.

## 2. EXISTENCE OF THE FREE ENERGY DENSITY FOR LATtICE GAS CLUSTERS

We consider a classical lattice gas, on lattice $L$, with a nearest-neighbor attractive potential $-\varepsilon(0<\varepsilon<\infty)$. Particles on neighboring sites are said to
be bound, and a cluster is a maximal connected set of occupied sites. The partition function over internal degrees of freedom of a $p$-particle cluster is

$$
\begin{equation*}
Z_{p}^{L}=\sum_{X \mid N(X)=p} \xi^{k(X)} \tag{1}
\end{equation*}
$$

where $\xi=e^{\beta \varepsilon}$ is the Boltzmann factor per bond $(\beta$ is the inverse temperature), and the sum includes exactly one representative from each class of translationally equivalent $p$-particle cluster configurations. $k(X)$ is the number of bonds in configuration $X$. In a recent paper ${ }^{(10)}$ we showed that

$$
\begin{equation*}
Z_{p+p^{\prime}}^{L} \geqslant Z_{p}^{L} Z_{p^{\prime}}^{L} \tag{2}
\end{equation*}
$$

We now derive an upper bound on $\log Z_{p}^{L}$. In a lattice with coordination number $q$, a p-particle cluster has fewer than $\frac{1}{2} q p$ bonds, and so

$$
\begin{equation*}
\log Z_{p}^{L} \leqslant \frac{1}{2} q p \beta \varepsilon+\log \sigma(L ; p) \tag{3}
\end{equation*}
$$

where $\sigma(L ; p)$ is the number of translationally nonequivalent $p$-particle cluster configurations in lattice $L$. Penrose and Lebowitz ${ }^{(5)}$ used the Peierls construction ${ }^{(11,12)}$ to show that for the square lattice, $\mathbb{Z}^{2}$,

$$
\begin{equation*}
\sigma\left(\mathbb{Z}^{2} ; p\right)<\frac{9}{8} 3^{2 p} \tag{4}
\end{equation*}
$$

The argument of Penrose and Lebowitz may be paraphrased as follows. There is a (one-to-many) correspondence between clusters in a lattice and contours (simple, closed walks) in the dual lattice. If the cluster is simply connected (contains no vacancies), then the contour is formed by placing an edge accross each boundary bond (each bond between an occupied site and an unoccupied site). If the cluster contains vacancies, some of its bonds are cut, so that a contour which traverses these bonds, as well as the original boundary bonds, encloses a site if and only if it is occupied. The contour of a $p$-particle cluster consists of at most $(q-2) p+2$ steps, so

$$
\begin{equation*}
\sigma(L ; p) \leqslant \sum_{N=3}^{(q-2) p+2} w_{N} \tag{5}
\end{equation*}
$$

where $w_{N}$ is the number of translationally nonequivalent simple, closed walks of $N$ steps in the dual lattice. Noting that for the square lattice $(q=4)$, $w_{N} \leqslant 3^{N-2}$ if $N$ is even, and $w_{N}=0$ for $N$ odd, we recover (4).

To obtain an upper bound on $\sigma(L ; p)$ for the hexagon lattice, we note that in the triangle lattice (dual to hexagon), $w_{N} \leqslant 3 \cdot 5^{N-2}$, since there are three possible arrangements of the two steps incident on the lowest, leftmost
corner, and since each of the remaining steps may be oriented in at most five ways. Thus

$$
\begin{equation*}
\sigma(\bigvee ; p) \leqslant 3 \sum_{n=3}^{p+2} 5^{n-2}<\frac{15}{4} 5^{p} \tag{6}
\end{equation*}
$$

A similar argument yields the upper bound

$$
\begin{equation*}
\sigma(A ; p) \leqslant \sum_{n=3}^{2 p+1} 2^{2 n-2}<\frac{4}{3} 4^{2 p} \tag{7}
\end{equation*}
$$

for the triangle lattice.
The contour of a cluster in the simple cubic lattice may be constructed by placing a plaquette (unit square) across each boundary bond. As in the two-dimensional case, bonds within the cluster may be cut, so that the contour encloses a site iff it is occupied. Thus Eq. (5) applies if we interpret $w_{N}$ as the number of translationally nonequivalent, simple closed surfaces composed of $N$ plaquettes. To derive an upper bound on $w_{N}$, note first that there is one corner, $a$, where the orientations of three plaquettes are fixed. This corner may be taken as the one with coordinates $\left(x_{a}, y_{a}, z_{a}\right)$, where $x_{a}=\inf \left\{x_{j}\right\}, y_{a}=\inf \left\{y_{j} \mid x_{j}=x_{a}\right\}$, and $z_{a}=\inf \left\{z_{j} \mid x_{j}=x_{a}, y_{j}=y_{a}\right\}$, where $\left(x_{i}, y_{i}, z_{i}\right)$ are the coordinates of the $i$ th corner in the contour. An argument due to van der Waerden ${ }^{(13)}$ shows that the number of arrangements of the remaining $N-3$ plaquettes cannot exceed $3^{N-3}$. The rest of the surface may be constructed by attaching new plaquettes at free edges of plaquettes already present, and the order of placement can be fixed unambiguously. For example, we may attach the next plaquette at the free edge whose center has extreme coordinates (the same rule as was used to pick corner $a$ ). Each of the $N-3$ plaquettes not touching corner $a$ may be oriented in at most three ways, and so $w_{N} \leqslant 3^{N-3}$. Equation (5) then implies that

$$
\begin{equation*}
\sigma\left(\mathbb{Z}^{3} ; p\right) \leqslant \sum_{n=3}^{2 p+1} 3^{2 n-3}<\frac{3}{8} 3^{4 p} \tag{8}
\end{equation*}
$$

For the proof in Section 3 we shall also require upper bounds on the number of cluster configurations in lattices with longer-range bonding. A collection of occupied lattice sites which is connected if we allow bonds between first, second,..., up to $n$th neighbors will be called a cluster of type $n$. A simple extension of the Penrose-Lebowitz construction provides an upper bound on $\sigma^{(2)}\left(\mathbb{Z}^{2} ; p\right)$, the number of (translationally nonequivalent) type-2 cluster configurations in the square lattice (see Fig. 1). The contour of such a cluster may be constructed in the same manner as for nearest-neighbor (type1) clusters, as is illustrated below. The contour does not cross a second


Fig. 1. Construction of the contours of type-2 clusters in the square lattice.
neighbor bond, unless the bond has been cut so as to render the cluster simply connected, as in the example on the right side of Fig. 1.

The contour of a $p$-particle, type- 2 cluster consists of at most $4 p$ steps, and so from Eq. (5) we have

$$
\begin{equation*}
\sigma^{(2)}\left(\mathbb{Z}^{2} ; p\right) \leqslant \sum_{n=2}^{2 p} 3^{2 n-2}<\frac{3^{4 p}}{8} \tag{9}
\end{equation*}
$$

An analogous construction leads to the bound

$$
\begin{equation*}
\sigma^{(2)}\left(\mathbb{Z}^{3} ; p\right) \leqslant \sum_{n=3}^{3 p} 3^{2 n-3}<\frac{3^{6 p}}{24} \tag{10}
\end{equation*}
$$

for type-2 clusters in the simple cubic lattice. The right-hand side of Eq. (10) also serves as an upper bound on $\sigma$ (FCC; p), the number of (type-1) cluster configurations in the face-centered cubic lattice. This follows from the observation that if bonding in the simple cubic lattice is restricted to second neighbors only, we obtain the FCC bonding structure.

Deriving a bound on $\sigma^{(3)}\left(\mathbb{Z}^{3} ; p\right)$, the number of type- 3 cluster configurations in the simple cubic lattice, necessitates a modification of the procedure used so far. For if we employ the construction used above, we find that the contour of a type- 3 cluster may contain cubes which touch only at a corner. The Peierls argument does not seem to be useful in bounding the number of contours of the kind. Instead, we note that a type-3 cluster, if it is not already also a type- 2 cluster, may be converted into one by placing additional particles at certain unoccupied sites. This conversion, which establishes a connecting network of nearest- and/or next-nearest-neighbor bonds, requires the addition of at most $p-1$ new particles.

Let $r(Y)$ be the minimum number of particles needed to convert a type3 cluster, $Y$, into a type-2 cluster. Each type- 3 cluster of $p$ particles may be obtained by removing $r(Y)$ particles from a certain $(p+r(Y))$-particle, type-

2 cluster. The number of $p$-particle, type- 3 clusters corresponding to a given $t$-particle, type-2 cluster cannot exceed $\binom{t}{p}$, and so

$$
\begin{equation*}
\sigma^{(3)}\left(\mathbb{Z}^{3} ; p\right) \leqslant \sum_{r=0}^{p-1}\binom{p+r}{p} \sigma^{(2)}\left(\mathbb{Z}^{3} ; p+r\right)<\frac{p}{24} 2^{2 p} 3^{12 p-6} \tag{11}
\end{equation*}
$$

The right-hand side of Eq. (11) is also an upper bound on $\sigma(B C C ; p)$, the number of $p$-particle, type-1 cluster configurations in the body-centered cubic lattice, for if, in the simple cubic lattice, we restrict bonding to third neighbors only, we obtain the $B C C$ bonding structure.

In all the cases examined, we have seen that $\sigma(L ; p)<e^{c p}$, where $c$ is a finite constant which depends on the lattice. From Eq. (3) we therefore conclude that there is a finite constant $C(L)$ such that $(\beta p)^{-1} \log Z_{p}^{L}<C(L)$ for all $p$.

Lemma. Let $Y_{p}(p=1,2,3, \ldots)$ be a sequence with $Y_{1}>0$, and satisfying

$$
\begin{equation*}
Y_{p+p^{\prime}} \geqslant Y_{p} Y_{p^{\prime}} \tag{12}
\end{equation*}
$$

and let

$$
\begin{equation*}
y_{p} \equiv p^{-1} \log Y_{p}<C<\infty \tag{13}
\end{equation*}
$$

for all $p$. Then $y_{p}$ converges to a limit.
The proof is given in the Appendix. For the square, triangle, hexagon, simple cubic, FCC, and BCC lattices, the sequence $Z_{p}^{L}$ has been shown to satisfy the conditions of the lemma. Hence for these lattices we have the following:

Theorem. Let $Z_{p}^{L}$ be the partition function over internal degrees of freedom of a p-particle cluster in a lattice gas with nearest-neighbor attractive interactions. Then the limiting free energy density

$$
\begin{equation*}
\kappa \equiv \lim _{p \rightarrow \infty}-(\beta p)^{-1} \log Z_{p}^{L} \tag{14}
\end{equation*}
$$

exists for all temperatures.

## 3. PROOF FOR STABLE, TEMPERED POTENTIALS

In this section we prove the existence of the limiting cluster free energy density for continuous space models in two and three dimensions, where the potential $U$ and the cluster definition satisfy the following conditions:
(i) There is a constant $\varepsilon_{0}>-\infty$ such that for all cluster configurations $X=\left\{\mathbf{x}_{1}, \ldots, \mathbf{x}_{p}\right\}$,

$$
\begin{equation*}
U\left(\mathbf{x}_{1}, \ldots, \mathbf{x}_{p}\right) \geqslant \varepsilon_{0} p \tag{15}
\end{equation*}
$$

(ii) If $X=\left\{\mathbf{x}_{1}, \ldots, \mathbf{x}_{p}\right\}$ and $X^{\prime}=\left\{\mathbf{x}_{1}^{\prime}, \ldots, \mathbf{x}_{p^{\prime}}^{\prime}\right\}$ are cluster configurations, then there is a region $A \subset \mathbb{R}^{d}$ with volume $\mu(A) \geqslant a>0$, such that for $\mathbf{x} \in A$, $X^{\prime \prime}=\left\{\mathbf{x}_{1}, \ldots, \mathbf{x}_{p}, \mathbf{x}_{1}^{\prime}+\mathbf{x}, \ldots, \mathbf{x}_{p^{\prime}}^{\prime}+\mathbf{x}\right\}$ is a cluster configuration and

$$
\begin{equation*}
U\left(X^{\prime \prime}\right) \leqslant U(X)+U\left(X^{\prime}\right) \tag{16}
\end{equation*}
$$

(iii) Particles $i$ and $j$ are bound iff $0<r_{0} \leqslant\left|\mathbf{x}_{i}-\mathbf{x}_{j}\right| \leqslant \lambda<\infty$. A cluster configuration must have a connecting network of pairwise bonds.

Conditions (i) and (ii) are analogous to the stability and strong tempering conditions encountered in proofs of the thermodynamic limit. Condition (iii) simply expresses finite range bonding-a cutoff on the potential is not required. The presence of a connecting network of pairwise bonds is a necessary condition for a cluster; supplementary restrictions on the potential energy may also be imposed.

Examples of pairwise potentials and bond definitions satisfying conditions (i)-(iii) are: (1) the hard-core square well potential

$$
V(r)=\left\{\begin{array}{cc}
+\infty, & r<r_{0}  \tag{17}\\
-\varepsilon, & r_{0} \leqslant r \leqslant \lambda \\
0, & r>\lambda
\end{array}\right.
$$

with particles $i$ and $j$ bound iff $r_{0} \leqslant\left|\mathbf{x}_{i}-\mathbf{x}_{j}\right| \leqslant \lambda$, and, (2) the Lennard-Jones potential

$$
\begin{equation*}
V(r)=4 \varepsilon\left[\left(\frac{r_{0}}{r}\right)^{12}-\left(\frac{r_{0}}{r}\right)^{6}\right] \tag{18}
\end{equation*}
$$

with particles $i$ and $j$ bound iff $V\left(\left|\mathbf{x}_{i}-\mathbf{x}_{j}\right|\right) \leqslant \alpha<0(\alpha>\varepsilon)$. These potentials satisfy conditions (see Refs. 8 and 9) which ensure the validity of Eq. (15) for all configurations. That the HCSW model satisfies condition (ii) may be seen as follows. Let $x_{R}=\sup _{\mathrm{x}_{i} \in X} x_{i}$ and let $x_{L}^{\prime}=\inf _{\mathrm{x}_{i}^{\prime} \in X^{\prime}} x_{i}^{\prime}$, where $x_{i}$ is the $x$ component of $\mathbf{x}_{i}$. Let $A$ be the set of points $\mathbf{x}$ such that $x \geqslant x_{R}+r_{0}$ and $\left|\mathbf{x}-\mathbf{x}_{R}\right| \leqslant \lambda$. Clearly $\mu(A)$ is the same for all $X$, and $\mu(A)>0$. If we translate $X^{\prime}$ rigidly so that $\mathbf{x}_{L}^{\prime} \in A$, we generate, from $X$ and $X^{\prime}$ a new cluster $X^{\prime \prime}$ with $k\left(X^{\prime \prime}\right) \geqslant k(X)+k\left(X^{\prime}\right)+1$. Similarly, for the Lennard-Jones model we may take $x_{R}$ and $x_{L}^{\prime}$ as above, and $A$ as the set of points $\mathbf{x}$ satisfying $x \geqslant x_{R}+r_{1}$ and $\left|\mathbf{x}-\mathbf{x}_{R}\right| \leqslant r_{2}$, where $r_{1}<r_{2}$ and $V\left(r_{1}\right)=V\left(r_{2}\right)=\alpha$.

The cluster partition function over internal configurational degrees of freedom is

$$
\begin{equation*}
Z_{p}=\frac{1}{p!} \int_{\mathscr{E}} d \mathbf{x}_{2} \cdots d \mathbf{x}_{p} e^{-\beta U(X)} \tag{19}
\end{equation*}
$$

where $\mathscr{C}$ indicates the restriction to connected configurations. Particle 1 is fixed at the origin. We now show that $Z_{p}$ satisfies condition (13). From Eq. (15) we have that

$$
\begin{equation*}
Z_{p} \leqslant \frac{e^{-\beta \epsilon_{0} p}}{p!} \int_{\mathscr{C}} d \mathbf{x}_{2} \cdots d \mathbf{x}_{p} \equiv e^{-\beta \varepsilon_{0} p} I_{p} \tag{20}
\end{equation*}
$$

To place an upper bound on $I_{p}$, we establish a correspondence between cluster configurations and configurations of particles in a square or cubic lattice. Mark off such a lattice (of the same dimensionality, $d$, as the model under consideration), with spacing $\lambda$. Let $s$ of the cells be occupied by particles belonging to cluster configuration $X=\left\{\mathbf{x}_{1}, \ldots, \mathbf{x}_{p}\right\}$. In the lattice which is dual to the one just constructed, we take a site to be occupied iff its corresponding cell contains at least one particle in $X$. Suppose $\mathbf{x}_{i}, \mathbf{x}_{j} \in X$ are the coordinates of a bound pair. If particles $i$ and $j$ are not in the same cell, then, in two dimensions, they occupy cells with either an edge or a corner in common, corresponding to nearest or next-nearest neighbors in the dual lattice. Similarly, in three dimensions, a pair of bound particles are either in the same cell, or they occupy cells sharing a face, an edge, or a corner, corresponding to a pair of nearest-neighbor, next-nearest-neighbor, or thirdneighbor sites in the dual lattice. Thus the set of occupied sites corresponding to cluster configuration $X$ is a cluster of type $d$ (as defined in Section 2), in $d$ dimensions ( $d=2$ or 3 ).

We may classify configurations in $I_{p}$ according to their corresponding lattice cluster, and write

$$
\begin{equation*}
I_{p}=\sum_{s=1}^{p} \sum_{Y \mid N(Y)=s}\left(\sum_{n_{1}=1}^{p-s+1} \cdots \sum_{n_{s}=1}^{p-s+1}\right)^{*}\left(\prod_{i=1}^{s} \frac{1}{n_{i}!}\right) I_{p}^{(s)}\left(Y ; n_{1} \cdots n_{s}\right) \tag{21}
\end{equation*}
$$

where the second sum is over (translationally nonequivalent) $s$-particle type $d$ cluster configurations, $n_{i}$ is the number of particles in the $i$ th occupied cell, and $*$ indicates the restriction: $\sum_{i} n_{i}=p . I_{p}^{(s)}\left(Y ; n_{1}, \ldots, n_{s}\right)$ is the integral over all connected configurations $\left\{\mathbf{x}_{1}, \ldots, \mathbf{x}_{p}\right\}$, where $\mathbf{x}_{1}=0$ and $\mathbf{x}_{1}, \ldots, \mathbf{x}_{n}$ are restricted to the first occupied cell, $\mathbf{x}_{n_{1}+1}, \ldots, \mathbf{x}_{n_{1}+n_{2}}$ to the second, etc. Since

$$
\left(\prod_{i=1}^{s} \frac{1}{n_{i}!}\right) I_{p}^{(s)}\left(Y ; n_{1}, \ldots, n_{s}\right) \leqslant \lambda^{d(p-1)}
$$

we have

$$
\begin{equation*}
I_{p} \leqslant \lambda^{d(p-1)} \sum_{s=1}^{p}\binom{p-1}{s-1} \sigma^{(d)}\left(\mathbb{Z}^{d} ; s\right) \tag{22}
\end{equation*}
$$

In Section 2 it was shown that for $d=2$ or 3 ,

$$
\sigma^{(d)}\left(\mathbb{Z}^{d} ; s\right)<e^{C(d) s}
$$

Thus

$$
\begin{equation*}
I_{p} \leqslant \lambda^{d(p-1)} e^{C(d) p}\left(1+e^{-C(d)}\right)^{p-1} \tag{23}
\end{equation*}
$$

We conclude that for models satisfying conditions (i) and (ii), there is a finite constant $C^{\prime}$ such that

$$
\begin{equation*}
p^{-1} \log Z_{p}<-\beta \varepsilon_{0}+d|\log \lambda|+C^{\prime} \tag{24}
\end{equation*}
$$

It remains to show that $Z_{p}$ satisfies condition (12). Consider a pair of configurations ( $X, X^{\prime}$ ) occurring in the product

$$
\begin{equation*}
Z_{p} Z_{p^{\prime}}=\frac{1}{p^{\prime} p^{\prime}!} \int_{\mathscr{C}} d \mathbf{x}_{2} \cdots d \mathbf{x}_{p} \int_{\mathscr{C}} d \mathbf{x}_{2}^{\prime} \cdots d \mathbf{x}_{p^{\prime}}^{\prime} e^{-\beta\left[U(X)+U\left(X^{\prime}\right)\right]} \tag{25}
\end{equation*}
$$

Condition (ii) assures the existence of a region $A$ about $X$, such that if $X^{\prime}$ is translated rigidly so that $\mathbf{x}_{1}^{\prime} \in A$, we obtain a $p+p^{\prime}$-particle cluster. The contribution to $Z_{p+p^{\prime}}$ due to clusters formed from the pair $\left(X, X^{\prime}\right)$ is, by (16), at least $a e^{-\beta\left[U(X)+U\left(X^{\prime}\right)\right]}$, hence

$$
\begin{equation*}
a Z_{p+p^{\prime}} \geqslant\left(a Z_{p}\right)\left(a Z_{p^{\prime}}\right) \tag{26}
\end{equation*}
$$

Thus $a Z_{p}$ satisfies the conditions of the lemma, which proves the existence of

$$
\begin{equation*}
\lim _{p \rightarrow \infty} p^{-1} \log Z_{p}=\lim _{p \rightarrow \infty} p^{-1} \log \left(a Z_{p}\right) \tag{27}
\end{equation*}
$$

Thus the theorem of Section 2 applies if $Z_{p}$ is the internal configurational partition function of a $p$-particle cluster for a continuous space model ( $d=2$ or 3 ) satisfying conditions (i)-(iii).

Our result shows that for a large class of potentials and bonding definitions, the physical cluster approach to the theory of condensation introduced by Bijl, ${ }^{(14)}$ Band, ${ }^{(15)}$ and Frenkel ${ }^{(16)}$ is self-consistent.

## ACKNOWLEDGMENT

The authors thank the referee for proposing a derivation of Eq. (9) which is simpler than our original argument.

## APPENDIX. PROOF OF LEMMA ${ }^{4}$

Let $Y_{p}$ be a sequence satisfying conditions (12) and (13). First note that

$$
\begin{equation*}
y_{2^{n}}=2^{-n} \log Y_{2^{n}} \geqslant 2^{-(n-1)} \log Y_{2^{n-1}}=y_{2^{n-1}} \tag{A1}
\end{equation*}
$$

which, together with (13), implies that $\lim _{n \rightarrow \infty} y_{2^{n}}=y$. Now consider $y_{n}$ for $2^{m-1}<n<2^{m}$, i.e., $n=2^{m-1}+j$, where $j$ has the binary expansion

$$
\begin{equation*}
j=\sum_{n=0}^{m-2} a_{n} 2^{n} \tag{A2}
\end{equation*}
$$

with each $a_{n}$ either zero or one. By repeated application of (12) we have

$$
\begin{equation*}
\log Y_{2^{m-1}+j} \geqslant \log Y_{2^{m-1}}+\sum_{n=0}^{m-2} a_{n} \log Y_{2^{n}} \tag{A3}
\end{equation*}
$$

Dividing by $2^{m-1}+j$, this becomes

$$
\begin{align*}
y_{2^{m-1}+j} \geqslant & \frac{2^{m-1}}{2^{m-1}+j} y_{2^{m-1}}+\sum_{n=0}^{m-2} a_{n} \frac{2^{n}}{2^{m-1}+j} y_{2^{n}} \\
= & y_{2^{m-1}}-\sum_{n=0}^{m-2} a_{n} \frac{2^{n}}{2^{m-1}+j}\left(y_{2^{m-1}}-y_{2^{n}}\right) \\
\geqslant & y_{2^{m-1}}-\frac{1}{2^{m-1}} \sum_{n=0}^{[m / 2-1]} 2^{n}\left(y_{2^{m-1}}-y_{2^{n}}\right) \\
& -\frac{1}{2^{m-1}} \sum_{n=[m / 2]}^{m-2} 2^{n}\left(y_{2^{m-1}}-y_{2^{n}}\right) \tag{A4}
\end{align*}
$$

where square brackets indicate the largest integer of their argument. In the first sum, the summand cannot exceed $2^{m / 2-1}\left(y-y_{1}\right)$. The second sum is less than $2^{m-1}\left(y_{2^{m-1}}-y_{2[m / 2]}\right)$. Thus

$$
\begin{equation*}
y_{2^{m-1}+j} \geqslant y_{2^{m-1}}-\frac{m\left(y-y_{1}\right)}{2^{m / 2+1}}-\left(y_{2^{m-1}}-y_{2[m / 2]}\right) \tag{A5}
\end{equation*}
$$

for $1 \leqslant j<2^{m-1}$. Now consider, for $j$ again given by (A2), $y_{2^{m-j}}$. By (12) we have that

$$
\begin{equation*}
\log Y_{2^{m}-j} \leqslant \log Y_{2^{m}}-\sum_{n=0}^{m-2} a_{n} \log Y_{2^{n}} \tag{A6}
\end{equation*}
$$

[^1]so that
\[

$$
\begin{equation*}
y_{2^{m}-j} \leqslant y_{2^{m}}+\sum_{n=0}^{m-2} a_{n} \frac{2^{n}}{2^{m}-j}\left(y_{2^{m}}-y_{2^{n}}\right) \tag{A7}
\end{equation*}
$$

\]

Using the same rearrangements and estimates as in the derivation of (A5), we then find

$$
\begin{equation*}
y_{2^{m}-j} \leqslant y_{2 m}+\frac{m\left(y-y_{1}\right)}{2^{m / 2+1}}+\left(y_{2^{m}}-y_{2[m / 2]}\right) \tag{A8}
\end{equation*}
$$

for $1 \leqslant j<2^{m-1}$. Thus for $2^{m-1}<n<2^{m}$,

$$
\begin{align*}
y_{2^{m-1}} & -\frac{m\left(y-y_{1}\right)}{2^{m / 2+1}}-\left(y_{2^{m-1}}-y_{2^{[m / 2]}}\right) \\
& \leqslant y_{n} \leqslant y_{2^{m}}+\frac{m\left(y-y_{1}\right)}{2^{m / 2+1}}+\left(y_{2^{m}}-y_{2[m / 2]}\right) \tag{A9}
\end{align*}
$$

which implies

$$
\begin{equation*}
\left|y_{n}-y_{2^{m}}\right| \leqslant \frac{m\left(y-y_{1}\right)}{2^{m / 2+1}}+y_{2^{m}}-y_{2[m / 2]} \tag{A10}
\end{equation*}
$$

Let $m(n)=2^{\left[1-\log _{2} n\right]}$. Then we have from (A10) that

$$
\begin{equation*}
\left|y_{n}-y\right| \leqslant\left|y_{n}-y_{2^{m(n)}}\right|+\left|y-y_{2^{m(n)}}\right| \xrightarrow[n \rightarrow \infty]{ } 0 \tag{A11}
\end{equation*}
$$

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[^1]:    ${ }^{4}$ Professor M. E. Fisher has recently pointed out to us that this lemma is a special case of a result in the theory of subadditive functions. (See Theorem 6.6 .1 of Ref. 17.)

