EXACTLY SOLUBLE MODELS FOR SURFACE PARTITION OF LARGE CLUSTERS

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The surface partition of large clusters is studied analytically within a framework of the “Hills and Dales Model”. Three formulations are solved exactly by using the Laplace—Fourier transformation method. In the limit of small amplitude deformations, the “Hills and Dales Model” gives the upper and lower bounds for the surface entropy coefficient of large clusters. The found surface entropy coefficients are compared with those of large clusters within the 2- and 3-dimensional Ising models.

1. Introduction

The surface entropy of large clusters was introduced by Fisher in his droplet model (FDM) [1]. During last forty years, the FDM has been successfully used to analyze the condensation of a gaseous phase (droplets of all sizes) into a liquid. The systems analyzed with the FDM are numerous and involve the nuclear multifragmentation [2], nucleation of real fluids [3], the compressibility factor of real fluids [4], clusters within the Ising model [5], and percolation clusters [6].

Fisher postulated that the leading contribution to the surface entropy is proportional to the surface S, i.e. ωS (in dimensionless units) based on the study of the combinatorics of clusters. The coefficient ω is surface energy coefficient $\sigma_{\rm o}(T_{\rm c})$ per one constituent taken at the critical temperature $T_{\rm c}$. The surface entropy was studied recently in our paper [7]. There we developed the “Hills and Dales Model” (HDM) which is a statistical model of surface deformations that obeys the volume conservation of clusters under consideration. Using the novel mathematical method, the Laplace—Fourier transform [8], we were able to find the grand canonical surface partition (GCSP) of the HDM analytically. For vanishing deformations, we obtained the upper limit for the surface entropy coefficient ω of large clusters to be $\omega \approx 1.06009$ (in dimensionless units), i.e. about 6 % larger than that according to Fisher's postulate.

In the grand canonical formulation, the cluster volume is conserved on the average, but, in order to apply the HDM to small and finite clusters, it is necessary to consider a more strict form of the volume conservation. Therefore, in the present paper, we consider a specially constrained canonical formulation of the HDM and obtain the lower estimates for the surface entropy of finite and large clusters. For the limit of vanishing deformations, we also introduce the *semigrand canonical ensemble* which occupies an intermediate place between the grand canonical and canonical surface ensembles. With the help of the Laplace—Fourier transform technique [8], the constrained canonical surface partition (CCSP) and the semigrand canonical surface partition (SGCSP) are evaluated exactly for any volume of cluster. For large clusters, the leading contribution and its corrections are found analytically for the CCSP and SGCSP. The obtained values for the ω-coefficient are compared with the corresponding values for the 2- and 3-dimensional Ising models for different lattice geometries. It is shown that the values of ω within all the 2- and 3-dimensional Ising models lie between the supremum and infimum found by the HDM.

The paper is organized as follows. In Sect. 2, we formulate three ensembles for surface deformations within the HDM framework and solve them analytically by the Laplace—Fourier transform technique. Section 3 is devoted to the analysis of isochoric ensemble singularities and to the derivation of the upper estimates of the surface entropy coefficient. The lower estimates for the surface entropy coefficients are found and compared to the corresponding 2- and 3-dimensional Ising lattice values in Sect. 4. The conclusions are formulated in Sect. 5.

2. Hills and Dales Model

The HDM is a statistical model of surface deformations. We impose a necessary constraint that the deformations should conserve the total volume of the cluster of A-constituents. As in our previous paper [7], the main interest is focused on the deformations of vanishing amplitudes. This is sufficient to find both the absolute supremum and the absolute infimum for the ω-coefficient of the HDM. In this case, the shape of a deformation cannot be important for our consideration, so we can choose the regular one. For this reason, we consider cylindrical deformations of positive height $h_k > 0$ (hills) and negative height $-h_k$ (dales) with k-constituents at the base. For simplicity, it is assumed that the top (bottom) of a hill (dale) has the same shape as the surface of the original cluster of A-constituents. We also assume that (i) the statistical weight of deformations $\exp\left(-\sigma_o |\Delta S_k|/s_1/T\right)$ is given by the Boltzmann factor due to a change of the surface $|\Delta S_k|$ in units of the surface per constituent s_1; (ii) all hills of heights $h_k \le H_k$ (H_k is the maximal height of a hill with k-constituents at the base) have the same probability dh_k/H_k besides the statistical one; (iii) assumptions (i) and (ii) are valid for the dales.

These assumptions are not too restrictive and allow us to simplify the analysis and to find the one-particle statistical partition of a deformation of the k-constituent base as a convolution of two probabilities discussed above:

$$z_k^{\pm} \equiv \int\limits_0^{\pm H_k} \frac{dh_k}{\pm H_k}\, e^{-\frac{\sigma_o P_k |h_k|}{T s_1}} = T s_1 \frac{\left[1 - e^{-\frac{\sigma_o P_k H_k}{T s_1}}\right]}{\sigma_o P_k H_k}\,, \qquad (1)$$

where the upper (lower) sign corresponds to hills (dales). Here, P_k is the cylinder base perimeter. Our next step is to find the geometrical partition (degeneracy factor) or the number of ways to place the center of a given deformation on the surface of an A-constituent cluster which is occupied by the set of $\{n_l^{\pm} = 0, 1, 2, ...\}$ deformations of the l-constituents base.

For the grand canonical surface partition (GCSP), the desired geometrical partition can be given in the excluded-volume approximation [7] as

$$\mathcal{G}_{\rm gc} = \left[S_A - \sum_{k=1}^{K_{\max}} k\,(n_k^{+} \;+\; n_k^{-})\, s_1\right] s_1^{-1}\,, \qquad (2)$$

where $s_1 k$ is the area occupied by a deformation of the k-constituent base ($k = 1, 2, ...$), S_A is the full surface of the cluster, and $K_{\max}(S_A)$ is the A-dependent size of the maximal allowed base on the cluster. It is clearly seen now that the first multiplier on the right-hand side (r.h.s.) of (2) corresponds to the available surface to place the center of each of $\{n_k^{\pm}\}$ deformations that exist on the cluster surface. It is necessary to impose the condition $\mathcal{G}_{\rm gc} \ge 0$ which ensures that the deformations do not overlap. Equation (2) is the van der Waals excluded-volume approximation usually used in statistical mechanics at low particle densities [9–13] and can be derived for objects of different sizes in the spirit of Ref. [14].

According to Eq. (1) the statistical partition for the hill with a k-constituent base matches that of the dale, i.e. $z_k^{+} = z_k^{-}$, and, therefore, the GCSP

$$Z_{\rm gc}(S_A) = \sum_{\{n_k^{\pm}=0\}}^{\infty} \left[\prod_{k=1}^{K_{\max}} \frac{\left[z_k^{+}\mathcal{G}_{\rm gc}\right]^{n_k^{+}}}{n_k^{+}!}\; \frac{\left[z_k^{-}\mathcal{G}_{\rm gc}\right]^{n_k^{-}}}{n_k^{-}!}\right] \Theta(s_1 \mathcal{G}_{\rm gc}) \qquad (3)$$

corresponds to the conserved (on the average) volume of the cluster because the probabilities of a hill and a dale of the same base are identical. The $\Theta(s_1\mathcal{G}_{\rm gc})$-function in (3) ensures that only the configurations with positive value of the free surface of a cluster are taken into account, but this makes the calculation of the GCSP very difficult.

For small and finite clusters, we have to impose a more strict constraint of the exact volume conservation of a cluster. This can be done in several ways, but here we consider a special version of the canonical ensemble assuming that the number of the hills n_k^{+} of the k-constituent base is always identical to the number of the corresponding dales, i.e. $n_k^{-} \equiv n_k^{+} \equiv n_k$. Then the canonical geometrical partition can be cast as follows

$$\mathcal{G}_c = \left[S_A - 2\sum_{k=1}^{K_{\max}} k\, n_k\, s_1\right] (2 s_1)^{-1}\,, \qquad (4)$$

where the factor of 2 in the denominator on the right-hand side (r.h.s.) of (4) accounts for the fact that it is necessary to place simultaneously the centers of two k-constituent base deformations (a hill and a dale) out of $2n_k$ on the surface of a cluster. Using the geometrical partition (4), one can obtain the partition function of the canonical ensemble by formally replacing $\mathcal{G}_{\rm gc} \to \mathcal{G}_c$ and inserting the Kronecker delta $\delta_{n_k^{+}, n_k^{-}}$ for each k-multiplier in (3). We consider, however, each pair of hills and dales of the same base as a single degree of freedom. Therefore, the number of ways to place each pair out of n_k distinguishable pairs [15] is still given by the canonical geometrical partition $\mathcal{G}_c$. Multiplying it with the probability of a pair of deformations $z_k^{+} z_k^{-}$ and repeating this for n_k pairs, we obtain the CCSP as follows

$$Z_{\rm cc}(S_A) = \sum_{\{n_k=0\}}^{\infty} \left[\prod_{k=1}^{K_{\max}} \frac{\left[z_k^+ z_k^- \mathcal{G}_c\right]^{n_k}}{n_k!} \right] \Theta(2s_1 \mathcal{G}_c)\,. \qquad (5)$$

It is clear that, by construction, this partition obeys the volume conservation more strictly than the GCSP. As in the case of the GCSP, the $\Theta(2s_1\mathcal{G}_c)$-function in the CCSP ensures that only the configurations with positive value of the free surface of a cluster are accounted for, but this constraint makes the calculation of partition (5) very difficult.

An additional problem in evaluating partitions (3) and (5) appears due to the explicit dependence S_A of the maximal base of deformations via $K_{\max}(S_A)$, because the standard method to deal with the excluded volume partitions, the usual Laplace transform [10–13] in S_A, cannot be applied in this case. However, as shown in [7], GCSP (3) can be solved analytically with the help of the Laplace—Fourier technique [8]. The latter employs the identity

$$G(S_A) = \int\limits_{-\infty}^{+\infty} d\xi \int\limits_{-\infty}^{+\infty} \frac{d\eta}{2\pi}\; e^{i\eta(S_A-\xi)}\; G(\xi) \qquad (6)$$

which is based on the Fourier representation of the Dirac δ-function. Similar to the GCSP, representation (6) allows us to decouple the additional S_A-dependence in $K_{\max}(S_A)$ of the CCSP and to reduce it to the exponential one which can be integrated by using the Laplace transformation [7, 8]

$$\begin{aligned}
\mathcal{Z}_{\rm cc}(\lambda) \;&\equiv \int\limits_0^{\infty} dS_A\; e^{-\lambda S_A}\; Z_{\rm cc}(S_A) = \\
&= \int\limits_0^{\infty} dS' \int\limits_{-\infty}^{+\infty} d\xi \int\limits_{-\infty}^{+\infty} \frac{d\eta}{2\pi}\; e^{i\eta(S'-\xi)-\lambda S'} \times \\
&\times \sum_{\{n_k=0\}}^{\infty} \left[\prod_{k=1}^{K_{\max}(\xi)} \frac{\left[z_k^+ z_k^- S' e^{2k\, s_1(i\eta-\lambda)}\right]^{n_k}}{n_k!\; (2\; s_1)^{n_k}} \right] \Theta(S') = \\
&= \int\limits_0^{\infty} dS' \int\limits_{-\infty}^{+\infty} d\xi \int\limits_{-\infty}^{+\infty} \frac{d\eta}{2\pi}\; e^{i\eta(S'-\xi)-\lambda S' + S' \mathcal{F}_{\rm cc}(\xi, \lambda - i\eta)}\,. \qquad (7)
\end{aligned}$$

After changing the integration variable $S_A \to S' = S_A - 2 \sum\limits_{k=1}^{K_{\max}(\xi)} k\, n_k\, s_1$, the constraint of the Θ-function has disappeared. Next all n_k were summed independently leading to the exponential function. Now the integration over S' in (7) can be done by giving the canonical isochoric partition

$$\mathcal{Z}_{\rm cc}(\lambda) = \int\limits_{-\infty}^{+\infty} d\xi \int\limits_{-\infty}^{+\infty} \frac{d\eta}{2\pi}\; \frac{e^{-i\eta\xi}}{\lambda - i\eta \;-\; \mathcal{F}_{\rm cc}(\xi, \lambda - i\eta)}\;, \qquad (8)$$

where the function $\mathcal{F}_{\rm cc}(\xi, \tilde{\lambda})$ is defined as

$$\mathcal{F}_{\rm cc}(\xi, \tilde{\lambda}) = \sum_{k=1}^{K_{\max}(\xi)} \frac{z_k^+\, z_k^-}{2\, s_1}\; e^{-2\,k\,s_1 \tilde{\lambda}}\,. \qquad (9)$$

Representation (8) is generic, and it is also valid for the GCSP, if the canonical function (9) is replaced by the grand canonical one

$$\mathcal{F}_{\rm gc}(\xi, \tilde{\lambda}) = \sum_{k=1}^{K_{\max}(\xi)} \left[\frac{z_k^+}{s_1} + \frac{z_k^-}{s_1} \right]\; e^{-k\,s_1 \tilde{\lambda}}\,. \qquad (10)$$

Before making the inverse Laplace transformation and studying the structure of singularities of functions (9) and (10), it is necessary to discuss one more ensemble for the surface deformations which will be called hereafter as the *semigrand canonical surface partition.*

This ensemble occupies an intermediate position between the constrained canonical and grand canonical formulations. It corresponds to the case where the hills and dales of the same base are considered to be indistinguishable. For that, we would like to explore the fact that the statistical probabilities of hills and dales of the same base are equal according to (1). Then, for the infinitesimally small amplitudes of deformations, the volume conservation constraint is fulfilled trivially. In the resent work, this ensemble will be used for the deformations of vanishing amplitudes only, but it may be used also for finite amplitudes of deformations, if the volume is not conserved.

Then the geometrical factor reads as

$$\mathcal{G}_{\rm sg} = \left[S_A - \sum_{k=1}^{K_{\max}} k\, n_k\, s_1 \right] s_1^{-1}\,, \qquad (11)$$

and the SGCSP has the form

$$Z_{\rm sg}(S_A) = \sum_{\{n_k=0\}}^{\infty} \left[\prod_{k=1}^{K_{\max}} \frac{\left[z_k^+ \mathcal{G}_{\rm sg}\right]^{n_k}}{n_k!} \right] \Theta(s_1 \mathcal{G}_{\rm sg})\,. \qquad (12)$$

It is easy to show that, using the Laplace—Fourier transformation technique [8], the SGCSP (12) can be transformed into the generic representation (8) for the function

$$\mathcal{F}_{\rm sg}(\xi,\tilde{\lambda}) = \sum_{k=1}^{K_{\max}(\xi)} \frac{z_k^+}{s_1}\, e^{-k\, s_1 \tilde{\lambda}} . \qquad (13)$$

By construction, Eqs. (12) and (13) are less fundamental than the corresponding grand canonical and constrained canonical functions.

3. Analysis of Singularities

To study the structure of singularities of the isochoric partition (8), it is necessary to make the inverse Laplace transformation ($\alpha \in \{\rm gc, sg, cc\}$):

$$Z_\alpha(S_A) \;=\; \int\limits_{\chi-i\infty}^{\chi+i\infty} \frac{d\lambda}{2\pi i}\; \mathcal{Z}_\alpha(\lambda)\; e^{\lambda\, S_A} =$$

$$\int\limits_{-\infty}^{+\infty} d\xi \int\limits_{-\infty}^{+\infty} \frac{d\eta}{2\pi} \int\limits_{\chi-i\infty}^{\chi+i\infty} \frac{d\lambda}{2\pi i}\; \frac{e^{\lambda\, S_A - i\eta\xi}}{i\lambda - i\eta \;-\; \mathcal{F}_\alpha(\xi, \lambda - i\eta)} =$$

$$= \int\limits_{-\infty}^{+\infty} d\xi \int\limits_{-\infty}^{+\infty} \frac{d\eta}{2\pi}\, e^{i\eta(S_A-\xi)} \sum_{\{\tilde{\lambda}_n\}} e^{\tilde{\lambda}_n\, S_A} \Big[1 - \frac{\partial \mathcal{F}_\alpha(\xi,\tilde{\lambda}_n)}{\partial \tilde{\lambda}_n}\Big]^{-1}, \qquad (14)$$

where the contour integral with respect to λ is reduced to the sum over the residues of all singular points $\lambda = \tilde{\lambda}_n + i\eta$ with $n = 0,1,2,..$, since this contour in the complex λ-plane obeys the inequality $\chi > \max({\rm Re}\{\tilde{\lambda}_n\})$. Now all integrations in (14) can be done, and all three surface partitions ($\alpha \in \{\rm gc, sg, cc\}$) can be written as

$$Z_\alpha(S_A) \;=\; \sum_{\{\tilde{\lambda}_n\}} e^{\tilde{\lambda}_n\, S_A} \Big[1 - \frac{\partial \mathcal{F}_\alpha(S_A,\tilde{\lambda}_n)}{\partial \tilde{\lambda}_n}\Big]^{-1}, \qquad (15)$$

i.e. the double integral in (14) simply reduces to the substitution $\xi \to S_A$ in the sum over singularities. This remarkable answer for all three surface partitions is a partial example of the general theorem on the Laplace-Fourier transformation properties proved in [8].

The simple poles in (14) are defined by the condition $\tilde{\lambda}_n \;=\; \mathcal{F}_\alpha(S_A, \tilde{\lambda}_n)$, and the latter can be cast for each ensemble as a system of two coupled transcendental equations

$$R_n^\alpha = \sum_{k=1}^{K_{\max}(S_A)} \phi_k^\alpha\; e^{-k\, R_n^\alpha} \cos(I_n^\alpha k)\,, \qquad (16)$$

$$I_n^\alpha = - \sum_{k=1}^{K_{\max}(S_A)} \phi_k^\alpha\; e^{-k\, R_n^\alpha} \sin(I_n^\alpha k) \qquad (17)$$

for dimensionless variables defined as $R_n^\alpha = s_1{\rm Re}(\tilde{\lambda}_n)$ and $I_n^\alpha = s_1{\rm Im}(\tilde{\lambda}_n)$ for the GCSP and SGCSP, and as $R_n^c = 2s_1{\rm Re}(\tilde{\lambda}_n)$ and $I_n^c = 2s_1{\rm Im}(\tilde{\lambda}_n)$ for the CCSP. Here, the function ϕ_k^α is given by the expression

$$\phi_k^\alpha = \left\{ \begin{array}{ll} z_k^+ + z_k^-\,, & \mbox{for } \alpha = {\rm gc}\,, \\ z_k^+ z_k^-\,, & \mbox{for } \alpha = {\rm cc}\,, \\ z_k^+\,, & \mbox{for } \alpha = {\rm sg}\,. \end{array} \right. \qquad (18)$$

To this point, Eqs. (16), (17) and (18) are general and can be used for particular models which specify the height of hills and the depth of dales. But it is possible to give both the upper and lower estimates for all three partition functions of large clusters, and even to estimate corrections for finite and small clusters. For the upper estimate, let us consider the real root ($R_0^\alpha; I_0^\alpha = 0$) of these equations. It is sufficient to consider the limit $K_{\max}(S_A) \to \infty$, because the r.h.s. of (16) for $I_n^\alpha = I_0^\alpha = 0$ is a monotonously increasing function of $K_{\max}(S_A)$. Since $z_k^+ = z_k^-$ are the monotonously decreasing functions of H_k, the maximal value of the r.h.s. of (16) corresponds to the limit of infinitesimally small amplitudes of deformations, $H_k \to 0 \Rightarrow z_k^+ = z_k^- = 1$. Under these conditions, Eq. (17) for $I_n^\alpha = I_0^\alpha = 0$ becomes an identity, and Eq. (16) acquires the form

$$R_0^\alpha = B^\alpha \sum_{k=1}^{\infty} e^{-k\, R_0^\alpha} = B^\alpha \left[e^{R_0^\alpha} - 1 \right]^{-1} , \qquad (19)$$

where $B^{\rm gc} = 2$ and $B^{\rm cc} = B^{\rm sg} = 1$. Therefore, the real roots of (16) and the corresponding surface entropy coefficients $\omega_{\rm U}^\alpha$ are as follows:

$$R_0^\alpha = \left\{ \begin{array}{ll} \omega_{\rm U}^{\rm gc} = \max\{\omega^{\rm gc}\} \approx 1.060090\,, & \alpha = {\rm gc}\,, \\ 2\omega_{\rm U}^{\rm cc} = 2\max\{\omega^{\rm cc}\} \approx 0.806466\,, & \alpha = {\rm cc} \\ \omega_{\rm U}^{\rm sg} = \max\{\omega^{\rm sg}\} \approx 0.806466\,, & \alpha = {\rm sg}\,. \end{array} \right. \qquad (20)$$

Results (20) correspond to the upper estimate for the surface partitions because, for $I_n^\alpha \neq 0$ defined by (17), the inequality $\cos(I_n^\alpha k) \leq 1$ cannot become the equality for all values of k simultaneously. Then it follows that the real root of (16) obeys the inequality $R_0^\alpha > R_{n>0}^\alpha$. The last result means that, in the limit of infinite cluster $S_A \to \infty$, all surface partitions (15) are represented by the farthest right singularity among all simple poles $\{\tilde{\lambda}_n\}$,

$$\max\{Z_\alpha(S_A)\} \rightarrow \frac{e^{\omega_{\rm U}^\alpha \frac{S_A}{s_1}}}{1+\frac{R_0^\alpha(R_0^\alpha+B^\alpha)}{B^\alpha}} = g_\alpha\, e^{\omega_{\rm U}^\alpha \frac{S_A}{s_1}}\,, \qquad (21)$$

where the geometrical degeneracy prefactor g_α is defined as follows: $g_{\rm gc} \approx 0.38139$ and $g_{\rm cc} = g_{\rm sg} \approx 0.407025$. Thus, the geometrical factor of the leading term for all three models is practically the same.

Remarkably, result (21) is model independent. This is a consequence of the limit of vanishing deformations, in which all model specific parameters vanish. The second remarkable fact is that Eq. (21) is valid for any self-non-intersecting surfaces of cluster. This is so because both *the shape and dimensionality of the cluster under consideration do not enter into our equations explicitly.* For our analysis of the HDM surface partitions, it was sufficient to require that the cluster surface together with deformations is a regular surface without self-intersections. Therefore, for vanishing deformations, the latter means that Eq. (21) should be valid for any self-non-intersecting surfaces.

For large, but finite clusters, it is necessary to take into account not only the farthest right singularity $\tilde{\lambda}_0$ in (15), but all other roots with positive real part $R^\alpha_{n>0} > 0$. The analysis presented in Appendix A shows that, besides the opposite signs, there are two branches of solutions, $I_n^{\alpha\,+}$ and $I_n^{\alpha\,-}$, for the same $n \geq 1$ value:

$$|I_n^{\alpha\,\pm}| \approx 2\pi n \pm \frac{B^\alpha}{2\pi n}\,, \qquad (22)$$

$$R_n^\alpha \approx \frac{(B^\alpha)^2}{8\pi^2 n^2}\,. \qquad (23)$$

The exact solutions $(R_n^\alpha; I_n^{\alpha\,\pm})$ for $n \geq 1$ which have the largest real part are shown in Fig. 1 together with the curve parametrized by functions $I_x^{\alpha\,+}$ and R_x^α taken from Eqs. (22) and (23), respectively. It is clear from Eq. (23) and Fig. 1 that the largest real part $R_1^{\rm gc} \approx 0.0582$ for the GCSP is about 18 times smaller than $R_0^{\rm gc}$, whereas, for the CCSP and SGCSP, the real part $R_1^{\rm cc} = R_1^{\rm sg}$ of the first rightest complex root of Eqs. (16) and (17) is about 63.6 times smaller than $R_0^{\rm cc} = R_0^{\rm sg}$. Therefore, for a cluster of a few constituents, the correction to the leading term (21) is exponentially small for all considered partitions. Using approximations (22) and (23), one can estimate the upper limit of the $(R_n^\alpha; I_n^{\alpha\,\pm})$ root contribution into Eq. (15) for $n > 2$ as

$$\left| e^{\tilde{\lambda}_n S_A} \left[1 - \frac{\partial \mathcal{F}_\alpha(S_A,\tilde{\lambda}_n)}{\partial \tilde{\lambda}_n}\right]^{-1} \right| \leq e^{\frac{(B^\alpha)^2 S_A}{8\pi^2 n^2 s_1}} \,/\, (2\pi^2 n^2)\,. \quad (24)$$

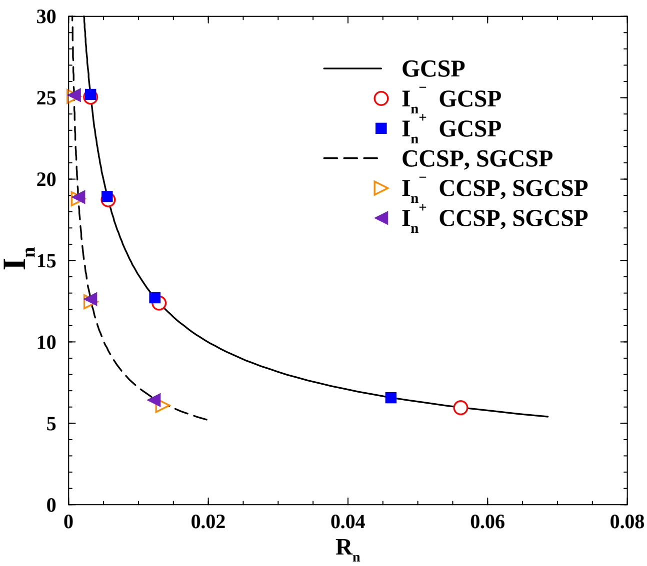

Fig. 1. The first quadrant of the complex plane $s_1\tilde{\lambda}_n \equiv R_n + iI_n$ shows the roots of the system of equations (16) and (17). The symbols represent the two branches I_n^- and I_n^+ of the roots for the upper estimate of three surface partitions. The curve is defined by the approximation given by (22) and (23) (see text for more details)

This result shows that, for all three considered partitions, the total contribution of all complex poles in (15) is negligibly small compared to the leading term (21) for a cluster of a few constituents or more.

4. Surface Entropy Coefficients

To complete our analysis of the limit of vanishing deformations, we would like to find the lower estimate for the GCSP, CCSP and SGCSP for large clusters. This estimate corresponds to the absence of all other deformations except for those of the smallest base. In other words, one has to substitute $K_{\max}(S_A) = 1$ in all corresponding expressions. Then Eqs. (16) and (17) become, respectively,

$$R_n^\alpha = \phi_1^\alpha\, e^{-R_n^\alpha} \cos(I_n^\alpha)\,, \qquad (25)$$

$$I_n^\alpha = -\phi_1^\alpha\, e^{-R_n^\alpha} \sin(I_n^\alpha)\,. \qquad (26)$$

Similar to the previous consideration, the leading term of the lower estimate for the surface partitions (15) is given by the real root $(R_0^\alpha; I_0^\alpha = 0)$ of system (25), (26):

$$R_0^\alpha = \begin{cases} \omega_{\rm L}^{\rm gc} = \min\{\omega^{\rm gc}\} \approx 0.852606\,, & \alpha = {\rm gc}, \\ 2\omega_{\rm L}^{\rm cc} = 2\min\{\omega^{\rm cc}\} \approx 0.567143\,, & \alpha = {\rm cc}, \\ \omega_{\rm L}^{\rm sg} = \min\{\omega^{\rm sg}\} \approx 0.567143\,, & \alpha = {\rm sg}. \end{cases} \qquad (27)$$

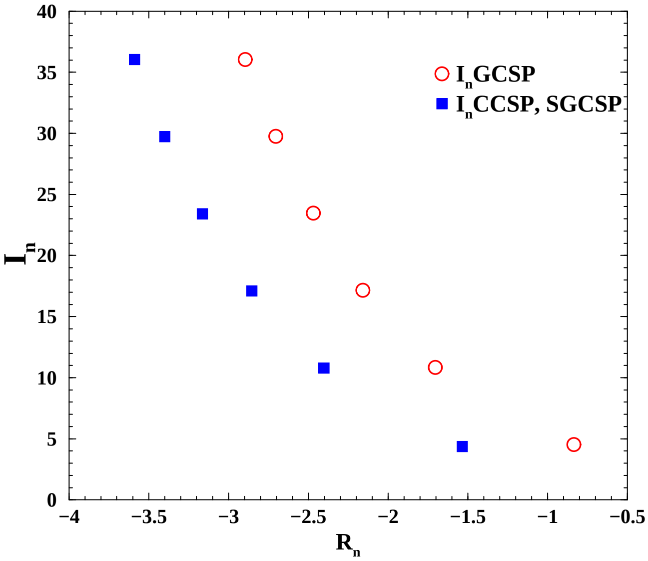

Fig. 2. The second quadrant of the complex plane $s_1\tilde{\lambda}_n \equiv R_n + iI_n$ shows the complex roots of the system of equations (25) and (26) with the largest real parts. The circles and squares represent the roots for the lower estimate of the GCSP and CCSP(SGCSP), respectively

Again, as in the case of upper estimates, one can show that the real root $(R_0^\alpha; I_0^\alpha = 0)$ approximates well the lower estimate for the partition function for a system of a few constituents. In fact, each of three surface partitions has only a single root with positive real part which coincides with $(R_0^\alpha; I_0^\alpha = 0)$. In Fig. 2, a few complex roots of Eqs. (25) and (26) with the largest real parts are shown. Since all these roots have negative real part, they generate an exponentially small contribution to the lower estimate of the surface partition for a system of a few constituents.

The ω-coefficients for the upper and lower estimates of all three surface partitions are summarized in the Table 1. A comparison with the corresponding coefficient for liquids should be made with care, because various contributions to the surface tension, i.e., the eigen surface tension of a liquid drop, the geometrical degeneracy factor (surface partition), and the part induced by the interaction between clusters, are not exactly known. Therefore, even the linear temperature dependence of the surface tension $\sigma(T) = \sigma_o (T_c - T)/T_c$ due to Fisher [1] applied to a nuclear liquid ($\sigma_o \approx 18$ MeV; $T_c \approx 18$ MeV [9]) may be used to estimate the ω-coefficient, if both the eigen surface tension and the interaction-induced one are non-increasing functions of temperature. Under these assumptions, one can get the following inequality for a nuclear liquid:

$$\omega_{\rm nucl} \ \le \ 1 \ < \ \omega_{\rm U}^{\rm gc} \ = \ 1.060090\,. \tag{28}$$

That is, the upper estimate for the GCSP provides, indeed, the upper limit for the surface partition of the nuclear matter.

A similar analysis for real liquids is difficult because of a complicated temperature dependence of the surface tension. Therefore, we would like to compare the ω-coefficients from Table 1 with the ω-coefficients for the large spin clusters of various 2- and 3-dimensional Ising models which are listed in Tables 2 and 3, respectively [16]. Such a comparison can be made because the surface entropy of large spin clusters on the Ising lattices are similar to the considered surface partitions (15) [16].

The ω-coefficient for the d-dimensional Ising model is defined as the energy $2J$ required to flip a given spin interacting with its q-neighbors to the opposite direction per $(d-1)$-dimensional surface divided by the value of critical temperature

$$\omega_{\rm lat} = \frac{q\ J}{T_c\, d}\,. \tag{29}$$

Here, q is the coordination number for the lattice, and J denotes the coupling constant of the model. A comparison of Tables 1– 3 shows that all lattice $\omega_{\rm lat}$-coefficients, indeed, lie between the upper estimates for the constrained canonical and grand canonical surface

T a b l e 1. The maximal and minimal values of the ω-coefficient for three statistical partitions of the HDM

Partition	$\max\{\omega^\alpha\}$	$\min\{\omega^\alpha\}$
GCSP	1.060090	0.852606
SGCSP	0.806466	0.567143
CCSP	0.403233	0.283572

T a b l e 2. The values of the $\omega_{\rm lat}$-coefficient for various 2-dimensional Ising models. For more details see the text

Lattice type	$\omega_{\rm lat} = \frac{\sigma}{T_c}$
Honeycomb	0.987718
Kagome	0.933132
Square	0.881374
Triangular	0.823960
Diamond	0.739640

T a b l e 3. The values of the $\omega_{\rm lat}$-coefficient for various 3-dimensional Ising models

Lattice type	$\omega_{\rm lat} = \frac{\sigma}{T_c}$
Simple cubic	0.44342
Body-centered cubic	0.41989
Face-centered cubic	0.40840

partitions,

$$0.403233 \;=\; \omega_{\rm U}^{\rm cc} \;<\; \omega_{\rm lat} \;<\; \omega_{\rm U}^{\rm gc} \;=\; 1.060090\,, \qquad (30)$$

i.e. $\omega_{\rm U}^{\rm cc}$ and $\omega_{\rm U}^{\rm gc}$ are the infimum and supremum for 2- and 3-dimensional Ising models, respectively.

The HDM partitions do not have an explicit dependence on the surface dimension, but a comparison of the HDM and Ising model ω-coefficients shows that the HDM ensembles seem to possess some sort of internal dimension: the GCSP is close to honeycomb, kagome, or square lattices, whereas the SGCSP is similar to triangular and diamond lattices, and the $\max\{\omega\}$ of the CCSP is closer to the 3-dimensional Ising models. In some cases, the agreement with the lattice data is remarkable – $\omega_{\rm L}^{\rm gc}$ coincides with the arithmetical average of the ω-coefficients for square and triangular lattices up to a fifth digit, but, in most cases, the values agree within a few per cent. The latter is not surprising, because the HDM estimates the surface entropy of a single cluster, whereas, on the lattice, the spin clusters do interact with each other and this, of course, changes the surface tension and, consequently, affects the value of critical temperature. It is remarkable that the so oversimplified estimates of the surface partitions for a single large cluster reasonably approximate the ω-coefficients for 2- and 3-dimensional Ising models.

It would be interesting to check whether the lower estimate of the CCSP, $\omega_{\rm L}^{\rm cc} \approx 0.283572$, is an infimum for the Ising lattices of higher dimensions $d > 3$. If this is the case, then we can give an upper limit for the critical temperature of those lattices, by using Eq. (29), as

$$\frac{T_{\rm c}}{J} \;\le\; \frac{q}{\omega_{\rm L}^{\rm cc}\, d} \;\approx\; 3.5264\,\frac{q}{d}\,. \qquad (31)$$

On the other hand, the lower estimate for the critical temperature of Ising lattices, $\frac{T_{\rm c}}{J} \;\ge\; \frac{q}{\omega_{\rm U}^{\rm gc}\, d}$, is provided by the supremum of the ω-coefficients of surface partitions.

5. Conclusions

We have formulated the grand canonical and constrained canonical partitions of surface deformations in the framework of the HDM. Both partitions conserve the volume of a deformed cluster and take into account all surface deformations with non-negative value of the free surface of this cluster. The grand canonical surface partition conserves the cluster volume on the average, whereas it is conserved exactly in the constrained canonical formulation. These partitions are solved exactly for an arbitrary (finite or infinite) size of the largest deformation by the Laplace—Fourier transformation technique, and the general analytical expression (15) for these partitions in terms of the set of isochoric ensemble singularities is derived.

Similarly, we have introduced and solved a special ensemble, a semigrand canonical partition, which obeys all constraints discussed above in the limit of vanishing deformations and occupies an intermediate place between the grand canonical and constrained canonical ensembles.

Then we considered the limit of vanishing deformations for all three surface partitions, and obtained the upper and lower estimates for the surface entropy for each of these partitions. The comparison of the obtained ω-coefficients for surface partitions with the corresponding coefficients for the large spin clusters of 2- and 3-dimensional Ising models shows that the upper estimate of the GCSP is a supremum, whereas the upper estimate of the CCSP is an infimum for the considered lattices. The question of the Ising lattice ω-coefficients for higher dimensions is discussed.

The developed formalism is rather general and, therefore, may be applied to the surface deformations of any kind of clusters, if the underlying mechanism of the surface deformations is given.

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APPENDIX A

For large, but finite clusters, it is necessary to take into account not only the farthest right singularity $\tilde\lambda_0$ in (15), but all other roots of Eqs. (16) and (17) which have positive real part $R^\alpha_{n>0} > 0$. In this case for each $R^\alpha_{n>0}$, there are two roots $\pm I^\alpha_n$ of (17), because the partition function (15) is real by definition. The roots of Eqs. (16) and (17) with the largest real part are insensitive to the large values of $K_{\max}(S_A)$, therefore, it is sufficient to keep $K_{\max}(S_A) \to \infty$. Then, in the limit of the vanishing amplitude of deformations, Eqs. (16) and (17) can be, respectively, rewritten as

$$\frac{B^\alpha R^\alpha_n}{(R^\alpha_n)^2 + (I^\alpha_n)^2} \;=\; e^{R^\alpha_n} \cos(I^\alpha_n) - 1\,, \qquad (A1)$$

$$\frac{B^\alpha I^\alpha_n}{(R^\alpha_n)^2 + (I^\alpha_n)^2} = -\; e^{R^\alpha_n} \sin(I^\alpha_n)\,. \qquad (A2)$$

After some algebra, the system of (A1) and (A2) can be reduced to a single equation for R^α_n

$$\cos\;\left[\frac{B^\alpha(B^\alpha+2R^\alpha_n)}{e^{2R^\alpha_n}-1} - (R^\alpha_n)^2\right]^{1/2} \;=$$

$$= \cosh R^\alpha_n - \frac{B^\alpha}{B^\alpha + 2\;R^\alpha_n}\sinh R^\alpha_n\,, \qquad (A3)$$

and the quadrature $I_n^\alpha = \sqrt{\frac{B^\alpha(B^\alpha+2R_n^\alpha)}{e^{2R_n^\alpha}-1} - (R_n^\alpha)^2}$. The analysis shows that, besides the opposite signs, there are two branches of solutions, $I_n^{\alpha\,+}$ and $I_n^{\alpha\,-}$, for the same $n \geq 1$ value. Expanding both sides of (A3) for $R_n^\alpha \ll 1$ and keeping the leading terms, one obtains (22) and (23). In Fig. 1, this approximation is compared with a few exact solutions $(R_n; I_n^{\pm})$ for $n \geq 1$ which have the largest real part.

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ТОЧНО РОЗВ'ЯЗУВАНІ МОДЕЛІ ДЛЯ ФУНКЦІЙ РОЗПОДІЛУ ПОВЕРХОНЬ ВЕЛИКИХ КЛАСТЕРІВ

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Р е з ю м е

В рамках "моделі пагорбів та долин" аналітично досліджуються функції розподілу поверхонь великих кластерів. Три формулювання моделі розв'язані точно із застосуванням методу перетворення Лапласа—Фур'є. У границі деформацій малих амплітуд "модель пагорбів та долин" визначає верхню та нижню межі для коефіцієнта поверхневої ентропії великих кластерів. Проведено порівняння знайдених коефіцієнтів поверхневої ентропії з коефіцієнтами поверхневої ентропії великих кластерів в дво- та тривимірній моделі Ізінга.