INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS A: MATHEMATICAL AND GENERAL

J. Phys. A: Math. Gen. **38** (2005) 7461–7482 doi:10.1088/0305-4470/38/34/002

Cluster density functional theory for lattice models based on the theory of Möbius functions

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Received 18 April 2005, in final form 15 July 2005
Published 10 August 2005
Online at stacks.iop.org/JPhysA/38/7461

Abstract

Rosenfeld's fundamental-measure theory for lattice models is given a rigorous formulation in terms of the theory of Möbius functions of partially ordered sets. The free-energy density functional is expressed as an expansion in a finite set of lattice clusters. This set is endowed with a partial order, so that the coefficients of the cluster expansion are connected to its Möbius function. Because of this, it is rigorously proven that a unique such expansion exists for any lattice model. The low-density analysis of the free-energy functional motivates a redefinition of the basic clusters (zero-dimensional cavities) which guarantees a correct zero-density limit of the pair and triplet direct correlation functions. This new definition extends Rosenfeld's theory to lattice models with any kind of short-range interaction (repulsive or attractive, hard or soft, one or multicomponent ...). Finally, a proof is given that these functionals have a consistent dimensional reduction, i.e. the functional for dimension d' can be obtained from that for dimension d ($d' < d$) if the latter is evaluated at a density profile confined to a d'-dimensional subset.

PACS numbers: 61.20.Gy, 05.20.Jj, 05.50.+q, 02.10.Ox
Mathematics Subject Classification: 82B05, 82B20, 06A07

1. Introduction

Rosenfeld's fundamental-measure theory (FMT) is a singularity in the world of approximate density functional theories. While all approximate functionals are built aiming at incorporating as much information on the uniform phase as there is available (Evans 1992), FMT is constructed on purely geometrical arguments (Rosenfeld 1989). Because of this, typical density functional recipes provide simple functionals with great flexibility to incorporate data of very different nature on the thermodynamics and the structure of the fluids, while fundamental-measure (FM) functionals have a very rigid structure which rejects almost any

deviation from orthodoxy (Tarazona 2002, Cuesta *et al* 2002). In spite of this, FMT has been successfully applied to a wide variety of models, including soft interacting spheres (Schmidt 1999, Sweatman 2002), non-additive mixtures (Schmidt *et al* 2000, Schmidt 2001a, 2004), associating fluids (Yu and Wu 2002), nonspherical hard bodies (Rosenfeld 1994, Cuesta and Martínez-Ratón 1997a, 1997b, Schmidt 2001b, Martínez-Ratón 2004), fluids in porous media (Schmidt 2002a) and nanopores (González *et al* 1997). It has even been applied to nonequilibrium problems, such as random sequential adsorption (Schmidt 2002b).

The counterpart of this theory's rigidity is that FM functionals exhibit a set of very special properties not shared by any other approximate functional. To begin with, the structure of the fluids is *predicted* rather than input (as in the other theories). Furthermore, FMT is naturally formulated for multicomponent systems, while other theories have serious difficulties to pass from one-component fluids to even binary mixtures (Denton and Ashcroft 1991, Choudhury *et al* 2002). But perhaps the most striking and characteristic property which distinguishes FM functionals is *dimensional reduction*. This means that if a d-dimensional system is constrained to lie in a d'-dimensional subset (with $d' < d$) and we evaluate the d-dimensional FM functional at the density profile describing this d'-dimensional confinement, then we obtain as a result the FM functional for the d'-dimensional system (Rosenfeld *et al* 1996, 1997, Cuesta and Martínez-Ratón 1997a, 1997b). This extraordinary consistency of the theory (as a matter of fact at the origin of its rigidity (Cuesta *et al* 2002)) cannot be found in any other density functional theory, and is obviously a feature that exact functionals possess. Dimensional reduction can be extended down to zero-dimensional (0D) confinements (cavities holding no more than a particle), and as the theory has developed, it has become more and more clear that this latter fact can be reformulated as a constructing principle of any FM functional (Tarazona and Rosenfeld 1997, Cuesta and Martínez-Ratón 1997a, Tarazona 2000).

FMT was first formulated as a continuum theory, simply because the overwhelming majority of applications of density functional theory is to continuum systems (for important exceptions see Robledo (1980), Robledo and Varea (1981), Percus (1982), Nieswand *et al* (1993), Buschle *et al* (2000a, 2000b), Prestipino and Griaquinta (2003), Heinrichs *et al* (2004)). But in a series of recent works the authors have extended the theory to lattice models and formulated a lattice fundamental-measure theory (LFMT) (Lafuente and Cuesta 2002, 2003, 2004, Lafuente 2004). The extension has been most revealing about the structure of FM functionals. In fact, LFMT, whose roots are very close to the continuum theory (Lafuente and Cuesta 2002), has its most natural formulation as a cluster theory, becoming a kind of density functional version of Kikuchi's cluster variation method (Kikuchi 1951) in Morita's formulation (Morita 1994). In its latest formulation, LFMT can be constructed, for hard-particle models, out of the exact free-energy functional of a given set of 0D cavities (Lafuente and Cuesta 2004, Lafuente 2004). The final result is of an extraordinary simplicity, given the fact that it provides the exact functional of many one-dimensional systems, and a good approximation for higher dimensional ones (typically a Bethe approximation for lattice gases with nearest-neighbour exclusion). On the other hand, LFMT exhibits consistency under mappings more general than lower dimensional confinements of the system (such as, for instance, the application of infinite external fields on some subset of the lattice nodes, which exclude the presence of particles; an illustration of one such mapping can be seen in Lafuente and Cuesta (2003), where it was used to derive the FM functional for a nearest neighbour-excluding lattice gas in a body-centred cubic lattice from the one in a simple cubic lattice). We will come back to this point in section 7.

In this work, we will present a formulation of LFMT based on a powerful combinatorial tool known as the *theory of Möbius functions* (Rota 1964, Aigner 1979, Stanley 1999), whose natural context are *incidence algebras* defined on partially ordered sets (or *posets*). This

formalism will allow us to rigorously prove a list of results about LFMT. Thus, after revisiting LFTM as reported in Lafuente and Cuesta (2004) and analysing its structure (section 2), we will prove (section 3) that given a lattice model as well as a basic set of clusters $\mathcal{W}_{\max}$ (to be precisely defined later) of the lattice, there exists a unique linear combination of the free-energy density functionals on subclusters of $\mathcal{W}_{\max}$ which yields the exact free energy when evaluated at 0D density profiles (i.e., density profiles with support one of those clusters). A special choice of $\mathcal{W}_{\max}$ gives rise to LFMT (section 4), but the cluster expansion has a wider range of applications (section 5). We study the low-density limit of the cluster expansion and redefine the clusters of LFMT (0D cavities) in such a way that for any model it is guaranteed that the zero-density limits of the pair and triplet direct correlation functions are exact (section 6), and suggest how to extend the definition in order to systematically incorporate higher order direct correlation functions. Finally, the behaviour of the cluster expansion under the action of certain mappings between lattice models is analysed, a consequence of which is the proof that LFMT is closed under dimensional reduction and other more general mappings (section 7).

The basic result used in the above proofs is a theorem about the Möbius function which is stated and proven in the appendix. The appendix also contains an important result of the theory of Möbius functions (the cross-cut theorem) that yields some simplifications in the calculation of the Möbius functions of a given poset. We conclude this paper with a discussion about some of the consequences of this reformulation of LFMT (the simplicity of its application being perhaps one of the most remarkable because of its practical consequences) as well as some open questions related to this theory.

2. Lattice fundamental-measure recipe reviewed: a multicomponent example

In this section, we will review the procedure recently proposed by the authors (Lafuente and Cuesta 2004) to construct a FM functional for any hard-core lattice model. As in the latest versions for continuum models (Tarazona and Rosenfeld 1997, Tarazona 2000), the constructive principle is based on the exact dimensional crossover to 0D cavities. In brief, the aim of this procedure is to build the simplest functional (under certain assumptions) which, applied to 0D cavities, produces the exact result.

The first hypothesis of the recipe, based on the exact functional for one-dimensional hard rods (Lafuente and Cuesta 2002) and the common pattern shared by all lattice FM functionals studied by the authors (Lafuente and Cuesta 2002, 2003), is that the excess (over ideal) free-energy functional of an arbitrary hard-core multicomponent system in a lattice $\mathcal{L}$ has the form (in units of kT, the Boltzmann constant times the temperature)

$$\mathcal{F}_{\mathrm{FM}}^{\mathrm{ex}}[\boldsymbol{\rho}] = \sum_{\boldsymbol{s}\in\mathcal{L}} \sum_{k\in\mathcal{I}} a_k \Phi_0(n^{(k)}(\boldsymbol{s})), \qquad n^{(k)}(\boldsymbol{s}) \equiv \sum_{i=1}^{p} \sum_{\boldsymbol{t}\in\mathcal{C}_i^{(k)}(\boldsymbol{s})} \rho_i(\boldsymbol{t}), \tag{2.1}$$

where $\mathcal{I}$ is a set of indices which label the different weighted densities $n^{(k)}(\boldsymbol{s})$; a_k are integer coefficients which depend on the specific model; $\Phi_0(\eta) \equiv \eta + (1-\eta)\ln(1-\eta)$ is the excess free energy of a cavity admitting no more than one particle in which $0 \leqslant \eta \leqslant 1$ is the average occupancy; $\boldsymbol{\rho}(\boldsymbol{s}) = (\rho_1(\boldsymbol{s}), \ldots, \rho_p(\boldsymbol{s}))$ denotes the vector of one-particle density functions of the different p species and $\boldsymbol{\mathcal{C}}^{(k)}(\boldsymbol{s}) \equiv \left(\mathcal{C}_1^{(k)}(\boldsymbol{s}), \ldots, \mathcal{C}_p^{(k)}(\boldsymbol{s})\right)$ are vectors formed by subsets of lattice nodes, i.e. $\mathcal{C}_i^{(k)}(\boldsymbol{s}) \subset \mathcal{L}$ (i is a species subindex). Hereafter, $\boldsymbol{\mathcal{C}}^{(k)}(\boldsymbol{s})$ will be referred to as (multicomponent) *cluster* or *cavity*. The kind of cavities involved in (2.1) are 1-*particle*

cavities because they are such that if a particle of species i occupies a node of $\mathcal{C}_i^{(k)}(\boldsymbol{s})$, it excludes all nodes of $\mathcal{C}_j^{(k)}(\boldsymbol{s})$ to particles of species j, for any $j = 1, \ldots, p$ (including i).[1]

Associated to cavities are 0*D density profiles*. If $\boldsymbol{\mathcal{C}} = (\mathcal{C}_1, \ldots, \mathcal{C}_p)$ denotes a cavity, a 0D density profile associated with $\boldsymbol{\mathcal{C}}$ is a density vector, denoted $\boldsymbol{\rho}_{\boldsymbol{\mathcal{C}}}(\boldsymbol{s}) = (\rho_{\mathcal{C}_1}(\boldsymbol{s}), \ldots, \rho_{\mathcal{C}_p}(\boldsymbol{s}))$, with support $\boldsymbol{\mathcal{C}}$ (i.e., $\rho_{\mathcal{C}_i}$ has support $\mathcal{C}_i$). Note that for such $\boldsymbol{\rho}_{\boldsymbol{\mathcal{C}}}(\boldsymbol{s})$, the corresponding weighted density $\sum_{i=1}^{p} \sum_{\boldsymbol{t}\in\mathcal{C}_i} \rho_{\mathcal{C}_i}(\boldsymbol{t})$ is the average occupancy of the lattice subset defined by $\bigcup_{i=1}^{p} \mathcal{C}_i^{(k)}(\boldsymbol{s})$.

Given a specific model, the functional in (2.1) will be completely determined by the coefficients $\{a_k\}$ and the family of subsets $\left\{\mathcal{C}_i^{(k)}(\boldsymbol{s})\right\}$. As it was shown in a previous work (Lafuente and Cuesta 2004), these unknowns can be uniquely determined by just imposing that the approximate functional (2.1) recovers the exact limit for *any* 0D density profile of any 1-particle cavity. This condition can be expressed as

$$\mathcal{F}_{\mathrm{FM}}^{\mathrm{ex}}[\boldsymbol{\rho}_{\boldsymbol{\mathcal{C}}}] = \Phi_0(\eta) \qquad \text{for } \textit{any } 1\text{-particle cavity } \boldsymbol{\mathcal{C}}, \tag{2.2}$$

with $\eta = \sum_{i=1}^{p} \sum_{\boldsymbol{t}\in\mathcal{C}_i} \rho_{\mathcal{C}_i}(\boldsymbol{t})$, the average occupancy of the cavity $\boldsymbol{\mathcal{C}}$. From this, it is easy to note that if the functional $\mathcal{F}_{\mathrm{FM}}^{\mathrm{ex}}[\boldsymbol{\rho}]$ satisfies condition (2.2) for the 0D density profile $\boldsymbol{\rho}_{\boldsymbol{\mathcal{C}}}(\boldsymbol{s})$, then this condition immediately holds for *any* 0D density profile $\boldsymbol{\rho}_{\boldsymbol{\mathcal{C}}'}(\boldsymbol{s})$ such that $\boldsymbol{\mathcal{C}}' \subset \boldsymbol{\mathcal{C}}$ (inclusion here must be understood componentwise), simply because $\boldsymbol{\rho}_{\boldsymbol{\mathcal{C}}'}$ is nothing but a particular choice of $\boldsymbol{\rho}_{\boldsymbol{\mathcal{C}}}$. Therefore, we can focus on the set of *maximal cavities* (Lafuente and Cuesta 2002), which are cavities not contained in any other cavity. Thus condition (2.2) holds if and only if it holds for the set of maximal cavities, i.e. we can replace (2.2) by

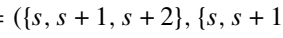
$$\mathcal{F}_{\mathrm{FM}}^{\mathrm{ex}}[\boldsymbol{\rho}_{\boldsymbol{\mathcal{C}}}] = \Phi_0(\eta) \qquad \text{for } \textit{any maximal } 1\text{-particle cavity } \boldsymbol{\mathcal{C}}. \tag{2.3}$$

In Lafuente and Cuesta (2004) we showed (and this will be proved in full detail in this paper) that once we have determined the set of maximal cavities—which only depends on the geometry of the interaction—condition (2.3) completely determines the functional (2.1) for the given system. In other words, this condition uniquely fixes both sets $\{a_k\}$ and $\left\{\mathcal{C}_i^{(k)}(\boldsymbol{s})\right\}$.

As an illustration of the procedure, we refer the reader to Lafuente and Cuesta (2004) for some specific examples of one-component models. Here, in order to review the recipe and to extend the collection of examples, we will apply the procedure to a one-dimensional binary hard-rod mixture.

Let us consider a system in the one-dimensional lattice $\mathbb{Z}$ with two species of hard rods: the largest of length $\sigma_{\mathrm{L}} = 3$ and the smallest $\sigma_{\mathrm{S}} = 2$ (both in lattice spacing units). This model represents one example of the non-additive case which was exactly solved in Lafuente and Cuesta (2002).

The first step of the procedure amounts to determining the set of maximal 1-particle cavities of the system. As in previous examples, we will use a diagrammatic notation for cavities $\boldsymbol{\mathcal{C}} = (\mathcal{C}_{\mathrm{L}}, \mathcal{C}_{\mathrm{S}})$. Remember that each $\mathcal{C}_i$ ($i = \mathrm{L}, \mathrm{S}$) is a subset of lattice nodes and thus we can associate $\mathcal{C}_i$ with the labelled graph whose vertices are the lattice nodes in $\mathcal{C}_i \subset \mathcal{L}$ and whose edges are the bonds linking nearest neighbours in the embedding lattice $\mathcal{L}$. In this representation we could have, for instance,

$\boldsymbol{\mathcal{C}} = (\{s, s+1, s+2\}, \{s, s+1\}) =$  $(s \in \mathbb{Z})$, (2.4)

[1] In previous works, this kind of object has been referred to as '0D cavity', but as we will redefine this concept later on, this new nomenclature is preferred.

where we have merged the two components of $\mathcal{C}$ in the last diagram using different colours to denote each species (white for the large and black for the small)[2]. With this notation, the set of maximal 1-particle cavities for the hard-rod binary mixture is

$$\mathcal{W}_{\max} = \{ \overset{s}{\bullet}\!\!-\!\!\bullet\!\!-\!\!\circ,\ \overset{s}{\circ}\!\!-\!\!\bullet\!\!-\!\!\bullet \mid s \in \mathbb{Z}\}. \tag{2.5}$$

Since we want the functional in (2.1) to recover the exact limit for all 0D density profiles with support any cavity in $\mathcal{W}_{\max}$, there must be, in (2.1), a contribution for each $\mathcal{C} \in \mathcal{W}_{\max}$. Note that the weighted density $n^{(k)}(s)$ can be identified with the associated cavity $\mathcal{C}^{(k)}(s) = \left(\mathcal{C}_1^{(k)}(s), \ldots, \mathcal{C}_p^{(k)}(s)\right)$ and the latter with the corresponding diagram. Therefore, we can write this contribution to (2.1) as

$$\sum_{s\in\mathbb{Z}} [a(\bullet\!\!-\!\!\bullet\!\!-\!\!\circ)\Phi_0(\overset{s}{\bullet}\!\!-\!\!\bullet\!\!-\!\!\circ) + a(\circ\!\!-\!\!\bullet\!\!-\!\!\bullet)\Phi_0(\overset{s}{\circ}\!\!-\!\!\bullet\!\!-\!\!\bullet)]. \tag{2.6}$$

Furthermore, if we want the functional to yield the exact 0D limit, coefficients $a(\bullet\!\!-\!\!\bullet\!\!-\!\!\circ)$ and $a(\circ\!\!-\!\!\bullet\!\!-\!\!\bullet)$ must be equal to 1 (this is always true for the coefficients associated with maximal cavities). This first analysis provides us with an initial guess for the functional of the system, namely

$$\mathcal{F}^{\mathrm{ex}}_{\mathrm{FM1}}[\rho] = \sum_{s\in\mathbb{Z}} [\Phi_0(\overset{s}{\bullet}\!\!-\!\!\bullet\!\!-\!\!\circ) + \Phi_0(\overset{s}{\circ}\!\!-\!\!\bullet\!\!-\!\!\bullet)]. \tag{2.7}$$

If this were the final functional, then it should satisfy condition (2.3). If we take, for instance, $\rho_{\mathcal{C}}$, with $\mathcal{C} = \bullet\!\!-\overset{t}{\bullet}\!\!-\!\!\circ$, as a test 0D density profile and evaluate the functional $\mathcal{F}^{\mathrm{ex}}_{\mathrm{FM1}}$, we obtain

$$\begin{aligned}\mathcal{F}^{\mathrm{ex}}_{\mathrm{FM1}}[\rho_{\bullet\!\!-\overset{t}{\bullet}\!\!-\!\!\circ}] = {} & \Phi_0(\bullet\!\!-\overset{t}{\bullet}\!\!-\!\!\circ) + \Phi_0(\circ\!\!-\overset{t}{\bullet}\!\!-\!\!\circ) + \Phi_0(\bullet\!\!-\overset{t}{\circ}) + \Phi_0(\overset{t}{\bullet}\!\!-\!\!\circ) \\ & + \Phi_0(\bullet\!\!-\overset{t}{\bullet}) + \Phi_0(\overset{t}{\circ}\!\!-\!\!\circ) + \Phi_0(\bullet_{t-1}) + \Phi_0(\circ_{t-1}) + 2\Phi_0(\circ_{t+1}),\end{aligned} \tag{2.8}$$

where we have used that $\Phi_0(0) = 0$ and that $\rho_{\mathcal{C}}$ is zero outside $\mathcal{C} = \bullet\!\!-\overset{t}{\bullet}\!\!-\!\!\circ$ (which implies, for instance, that $\Phi_0(\bullet\!\!-\!\!\bullet\!\!-\overset{t}{\circ}) = \Phi_0(\bullet\!\!-\overset{t}{\circ})$). Now, if we check (2.8) with condition (2.3), we note that apart from the exact contribution $\Phi_0(\bullet\!\!-\overset{t}{\bullet}\!\!-\!\!\circ)$ we also have a few spurious terms. Therefore, the initial guess (2.7) needs to be modified in order to eliminate these terms. Taking into account the general form (2.1) and the procedure explained in Lafuente and Cuesta (2004), we propose as a second guess

$$\mathcal{F}^{\mathrm{ex}}_{\mathrm{FM2}}[\rho] = \sum_{s\in\mathbb{Z}} [\Phi_0(\overset{s}{\bullet}\!\!-\!\!\bullet\!\!-\!\!\circ) + \Phi_0(\overset{s}{\circ}\!\!-\!\!\bullet\!\!-\!\!\bullet) + a(\circ\!\!-\!\!\bullet\!\!-\!\!\circ)\Phi_0(\circ\!\!-\overset{s}{\bullet}\!\!-\!\!\circ)], \tag{2.9}$$

where the coefficient $a(\circ\!\!-\!\!\bullet\!\!-\!\!\circ)$ has to be determined. The only way to remove the term $\Phi_0(\circ\!\!-\overset{t}{\bullet}\!\!-\!\!\circ)$ in (2.8) is to set $a(\circ\!\!-\!\!\bullet\!\!-\!\!\circ) = -1$; thus

$$\mathcal{F}^{\mathrm{ex}}_{\mathrm{FM2}}[\rho_{\bullet\!\!-\overset{t}{\bullet}\!\!-\!\!\circ}] = \Phi_0(\bullet\!\!-\overset{t}{\bullet}\!\!-\!\!\circ) + \Phi_0(\bullet\!\!-\overset{t}{\bullet}) + \Phi_0(\overset{t}{\bullet}\!\!-\!\!\circ) + \Phi_0(\bullet_{t-1}) + \Phi_0(\circ_{t+1}). \tag{2.10}$$

We can now iterate the procedure to remove the term $\Phi_0(\bullet\!\!-\!\!\bullet)$. The next guess is

$$\mathcal{F}^{\mathrm{ex}}_{\mathrm{FM3}}[\rho] = \sum_{s\in\mathbb{Z}} [\Phi_0(\overset{s}{\bullet}\!\!-\!\!\bullet\!\!-\!\!\circ) + \Phi_0(\overset{s}{\circ}\!\!-\!\!\bullet\!\!-\!\!\bullet) - \Phi_0(\circ\!\!-\overset{s}{\bullet}\!\!-\!\!\circ) + a(\bullet\!\!-\!\!\bullet)\Phi_0(\overset{s}{\bullet}\!\!-\!\!\bullet)]. \tag{2.11}$$

Choosing $a(\bullet\!\!-\!\!\bullet) = -1$, we get $\mathcal{F}^{\mathrm{ex}}_{\mathrm{FM3}}[\rho_{\bullet\!\!-\overset{t}{\bullet}\!\!-\!\!\circ}] = \Phi_0(\bullet\!\!-\overset{t}{\bullet}\!\!-\!\!\circ)$, and the exact limit is recovered for any 0D density profile with support any maximal 1-particle cavity in the set $\{\overset{s}{\bullet}\!\!-\!\!\bullet\!\!-\!\!\circ \mid s \in \mathbb{Z}\} \subset \mathcal{W}_{\max}$. By symmetry, it is easy to verify that the functional (2.11) needs no additional terms to fully satisfy (2.3); therefore the functional

$$\mathcal{F}^{\mathrm{ex}}_{\mathrm{FM}}[\rho] = \sum_{s\in\mathbb{Z}} [\Phi_0(\overset{s}{\bullet}\!\!-\!\!\bullet\!\!-\!\!\circ) + \Phi_0(\overset{s}{\circ}\!\!-\!\!\bullet\!\!-\!\!\bullet) - \Phi_0(\circ\!\!-\overset{s}{\bullet}\!\!-\!\!\circ) - \Phi_0(\overset{s}{\bullet}\!\!-\!\!\bullet)] \tag{2.12}$$

[2] Note that all nodes of the graph carry their corresponding label; the fact that we write a single label s on one of the nodes of the graph simply aims at avoiding clumsy notation.

is exact when evaluated at any 0D density profile. Moreover, this functional coincides with the *exact* one, obtained by Lafuente and Cuesta (2002) through a different, more involved method.

As it was discussed in Lafuente and Cuesta (2004), there are two remarkable features in this procedure. First of all, if we start off the iteration with the terms associated with maximal 1-particle cavities, then all clusters defining the weighted densities $n^{(k)}(\boldsymbol{s})$ in (2.1) are also 1-particle cavities of the system. Not only that, they are *intersections of the maximal cavities*. Secondly, once we adopt the first ansatz $\mathcal{F}^{\rm ex}_{\rm FM1}[\rho]$, there is a unique functional which fulfils condition (2.3) (in other words, the specific scheme one follows in order to remove the spurious terms is irrelevant). This last statement will be rigorously proved in the next section.

3. A cluster expansion for the free-energy density functional

If we analyse the general expression (2.1), we note that the FM excess free-energy functional is built from 1-particle cavity contributions. Furthermore, if we considered the system defined only in one of these cavities, say $\boldsymbol{\mathcal{C}}^{(k)}(\boldsymbol{s}) = \left(\mathcal{C}_1^{(k)}(\boldsymbol{s}), \ldots, \mathcal{C}_p^{(k)}(\boldsymbol{s})\right)$, the exact excess free energy would be $\Phi_0(n^{(k)}(\boldsymbol{s}))$. Therefore, if $\mathcal{F}^{\rm ex}_{\mathcal{C}}[\rho]$ denotes the exact excess functional of a system defined in cavity $\boldsymbol{\mathcal{C}}$, we can rewrite (2.1) as[3]

$$\mathcal{F}^{\rm ex}_{\rm FM}[\rho] = \sum_{\mathcal{C}\ \text{cavity}} a(\boldsymbol{\mathcal{C}})\mathcal{F}^{\rm ex}_{\mathcal{C}}[\rho]. \tag{3.1}$$

Using this cluster notation, the exact excess free-energy functional of the system under consideration can be denoted as $\mathcal{F}^{\rm ex}_{\mathcal{L}}[\rho]$, where $\boldsymbol{\mathcal{L}}$ is the (multicomponent) cluster $(\mathcal{L}, \ldots, \mathcal{L})$. Now taking into account that, in general, functional (3.1) is approximate, the exact one can be written as

$$\mathcal{F}^{\rm ex}_{\mathcal{L}}[\rho] \equiv \Psi[\rho] + \sum_{\mathcal{C}\ \text{cavity}} a(\boldsymbol{\mathcal{C}})\mathcal{F}^{\rm ex}_{\mathcal{C}}[\rho], \tag{3.2}$$

where $\Psi[\rho]$ is the error of the FM approximation (obviously an unknown functional).

As mentioned in the previous section, the coefficients $a(\boldsymbol{\mathcal{C}})$ are uniquely determined by condition (2.3), which, as we have shown, can be implemented through an iterative procedure determined by the inclusion relations of the intersections of the cavities involved (considered as subsets of lattice nodes). In this section, we will show that expression (3.2) is a particular case of *Möbius inversion formula*, a major result of the *theory of partially ordered sets*. The reader is strongly advised to consult the specialized literature on this subject (Rota 1964, Aigner 1979, Stanley 1999). Here, we will just introduce the necessary mathematical background to provide a comprehensible and rigorous foundation to LFMT.

3.1. Möbius inversion formula in a nutshell

A *partially ordered set* or *poset* is a set together with a partial order relation denoted by $\leqslant$, i.e. a binary relation satisfying reflexivity, antisymmetry and transitivity. An example of poset is the set of clusters involved in functional (2.12):

$$\mathcal{W} \equiv \{ {}^{s}\bullet\!\!-\!\!\bullet\!\!-\!\!\circ,\ {}^{s}\circ\!\!-\!\!\bullet\!\!-\!\!\bullet,\ \circ\!\!-\!\!{}^{s}\bullet\!\!-\!\!\circ,\ {}^{s}\bullet\!\!-\!\!\bullet \mid s \in \mathbb{Z}\},$$

with the order defined by $\boldsymbol{\mathcal{C}} \leqslant \boldsymbol{\mathcal{C}}'$ if and only if $\boldsymbol{\mathcal{C}} \subset \boldsymbol{\mathcal{C}}'$. In general, the order is partial because there may be pairs of elements which are not comparable. In our example neither ${}^{s}\bullet\!\!-\!\!\bullet\!\!-\!\!\circ \leqslant \circ\!\!-\!\!{}^{s}\bullet\!\!-\!\!\circ$ nor $\circ\!\!-\!\!{}^{s}\bullet\!\!-\!\!\circ \leqslant {}^{s}\bullet\!\!-\!\!\bullet\!\!-\!\!\circ$ are true, while, for instance, ${}^{s}\circ\!\!-\!\!\bullet\!\!-\!\!\circ \leqslant {}^{s}\bullet\!\!-\!\!\bullet\!\!-\!\!\circ$.

[3] This rewriting of (2.1) is particularly interesting because the fact that any explicit reference to the function $\Phi_0(\eta)$ has disappeared makes it applicable to cavities more general than the 1-particle cavities considered so far. More on this in sections 5 and 6.

Let $\mathcal{W}$ be a poset. For any $x, y \in \mathcal{W}$ such that $x \leqslant y$, we define the *interval* $[x, y] \equiv \{z \in \mathcal{W} | x \leqslant z \leqslant y\}$. A poset is said to be *locally finite* if all its intervals are finite. The set of all intervals of poset $\mathcal{W}$ will be denoted by $\mathrm{Int}(\mathcal{W})$.

Given a locally finite poset $\mathcal{W}$, the *incidence algebra* $I(\mathcal{W}, \mathbb{K})$ of $\mathcal{W}$ over field $\mathbb{K}$ is the set of all mappings $f: \mathrm{Int}(\mathcal{W}) \to \mathbb{K}$ (we will write $f(x, y)$ for $f([x, y])$) with the usual vector space structure and the inner product $(f * g)(x, y) \equiv \sum_{z \in [x,y]} f(x, z) g(z, y)$ (f and g being elements of $I(\mathcal{W}, \mathbb{K})$). Note that this product is well defined because being $\mathcal{W}$ locally finite the sum contains a finite number of terms. It is easy to check that $I(\mathcal{W}, \mathbb{K})$ is an associative algebra with (two-sided) identity $\delta(x, y) \equiv 1$ if $[x, y] = [x, x] = \{x\}$ and 0 otherwise. Another useful function of $I(\mathcal{W}, \mathbb{K})$ is the *zeta function* $\zeta(x, y) = 1$ for all $[x, y] \in \mathrm{Int}(\mathcal{W})$.

An important result for incidence algebras is (the present statement is a simplified version of proposition 3.6.2 on p 114 of Stanley (1999)):

Proposition 1. *Let f be an element of $I(\mathcal{W}, \mathbb{K})$; then f has (two-sided) inverse (i.e., there exists $f^{-1} \in I(\mathcal{W}, \mathbb{K})$ such that $f * f^{-1} = f^{-1} * f = \delta$) if and only if $f(x, x) \neq 0$ for all $x \in \mathcal{W}$.*

It follows from this proposition that the zeta function ζ of a locally finite poset $\mathcal{W}$ is invertible; its inverse is the *Möbius function* of $\mathcal{W}$ and is denoted by $\mu_{\mathcal{W}}$. This function can be obtained recursively from the definition of the inverse:

Recursion 1. $(\boldsymbol{\zeta * \mu_{\mathcal{W}} = \delta})$

$$\begin{aligned} &\mu_{\mathcal{W}}(x, x) = 1, \qquad \text{for all } x \in \mathcal{W}, \\ &\mu_{\mathcal{W}}(x, y) = -\sum_{x < \dot{z} \leqslant y} \mu_{\mathcal{W}}(z, y), \qquad \text{for } \; x < y \text{ with } \; x, y \in \mathcal{W}. \end{aligned} \tag{3.3}$$

Recursion 2. $(\boldsymbol{\mu_{\mathcal{W}} * \zeta = \delta})$

$$\begin{aligned} &\mu_{\mathcal{W}}(x, x) = 1, \qquad \text{for all } \; x \in \mathcal{W}, \\ &\mu_{\mathcal{W}}(x, y) = -\sum_{x \leqslant \dot{z} < y} \mu_{\mathcal{W}}(x, z), \qquad \text{for } \; x < y \text{ with } x, y \in \mathcal{W}. \end{aligned} \tag{3.4}$$

In both recursions 1 and 2, we have used a dot to indicate the summation variable.

We are now ready to formulate the key theorem for the rigorous foundation of LFMT (from Stanley (1999), proposition 3.7.1 on p 116):

Theorem 1 (Möbius inversion formula). *Let $\mathcal{W}$ be a poset such that for every $x \in \mathcal{W}$ the subset $\{y \in \mathcal{W} | y \leqslant x\}$ is finite. Let $f, g: \mathcal{W} \longrightarrow V(\mathbb{K})$, ($V(\mathbb{K})$ being a vector space over field $\mathbb{K}$). Then,*

$$f(x) = \sum_{\dot{y} \leqslant x} g(y) \qquad \text{for all } \; x \in \mathcal{W} \tag{3.5}$$

if and only if

$$g(x) = \sum_{\dot{y} \leqslant x} f(y) \mu_{\mathcal{W}}(y, x) \qquad \text{for all } \; x \in \mathcal{W}. \tag{3.6}$$

In order to illustrate the meaning of the Möbius function let us first examine a few known examples.

The first example is provided by the poset $\mathbb{N}$. If f and g are two functions on $\mathbb{N}$ such that

$$g(n) = \sum_{k=1}^{n} f(k),$$

then it is immediate that

$$f(n) = g(n) - g(n-1), \qquad n > 1, \quad f(1) = g(1).$$

Thus,

$$f(n) = \sum_{k=1}^{n} g(k)\mu(k,n), \qquad \mu(k,n) = \begin{cases} 1 & \text{if } k = n, \\ -1 & \text{if } k = n-1, \\ 0 & \text{otherwise.} \end{cases}$$

This example reveals in its most obvious way the 'discrete derivative' character of the Möbius function.

For the second example, we again take the set $\mathbb{N}$ but this time ordered by divisibility (i.e., $n \leqslant m$ iff m is a multiple of n, denoted $n|m$). In that case, for any two functions f and g on $\mathbb{N}$, if we have

$$g(n) = \sum_{d|n} f(d),$$

then

$$f(n) = \sum_{d|n} g(d)\mu(d,n),$$

where $\mu(d,n) = \tilde{\mu}(n/d)$, $\tilde{\mu}(n)$ being the classical Möbius function of number theory defined as

$$\tilde{\mu}(n) = \begin{cases} (-1)^k & \text{if } n = p_1 \cdots p_k, k \text{ distinct primes}, \\ 0 & \text{otherwise.} \end{cases}$$

The third and last example is a rederivation of the well-known combinatorial inclusion–exclusion principle. Let us consider n distinct finite sets, $S_1, \ldots, S_n$, and let us form the poset P containing them along with all their intersections and the set $\Sigma \equiv S_1 \cup \cdots \cup S_n$. The order in P is defined by set inclusion. For any $T \in P$, let us define two functions: $f(T)$ as the number of elements of T which are in no other $T' \subset T$ and $g(T) = |T|$, the number of elements of T. Then,

$$|T| = \sum_{T' \subset T} f(T'), \qquad f(T) = \sum_{T' \subset T} |T'|\mu(T', T).$$

Now, by definition, $f(\Sigma) = 0$ and $\mu(\Sigma, \Sigma) = 1$; therefore,

$$|\Sigma| = -\sum_{T \neq \Sigma} |T|\mu(T, \Sigma).$$

It is not difficult to prove that if t is the intersection of k of the original sets, then $\mu(T, \Sigma) = (-1)^k$, and the above formula becomes the announced inclusion–exclusion principle.

In the following, we will show that Möbius inversion formula can be applied to obtain a cluster expansion of the exact free-energy functional of a general lattice gas with arbitrary interaction. After that, it will be straightforward to prove that LFMT amounts to taking a particular truncation of this expansion.

3.2. Cluster expansion of the free-energy functional

The cluster expansion we will propose here is based on the formulation of the cluster variation method by Morita (1994). There, Möbius inversion is used to approximate the entropy of a lattice model by a linear combination of the exact entropies of the same model restricted to a family of lattice clusters. This is exactly the same idea we find in approximation (3.1), but here it is applied to the free-energy density functional.

Let us consider a multicomponent lattice gas with underlying lattice $\mathcal{L}$. Let us assume that we have a poset $\mathcal{W}$ whose elements are (multicomponent) clusters of lattice $\mathcal{L}$ (as in the previous example), such that cluster $\boldsymbol{\mathcal{L}}$ is in $\mathcal{W}$. Note that by definition $\boldsymbol{\mathcal{C}} \leqslant \boldsymbol{\mathcal{L}}$ for all $\boldsymbol{\mathcal{C}} \in \mathcal{W}$. Now, let us consider the mapping $f(\boldsymbol{\mathcal{C}}) \equiv \mathcal{F}_{\boldsymbol{\mathcal{C}}}[\boldsymbol{\rho}]$, where $\boldsymbol{\mathcal{C}} \in \mathcal{W}$ and $\mathcal{F}_{\boldsymbol{\mathcal{C}}}[\boldsymbol{\rho}]$ is the *exact* free-energy functional of the given model restricted to cluster $\boldsymbol{\mathcal{C}}$. Particularizing $x = \boldsymbol{\mathcal{L}}$ in (3.6) and taking into account that $\mu_{\mathcal{W}}(\boldsymbol{\mathcal{L}}, \boldsymbol{\mathcal{L}}) = 1$, we have for the exact free-energy functional of the system

$$\mathcal{F}_{\boldsymbol{\mathcal{L}}}[\boldsymbol{\rho}] = \Psi_{\boldsymbol{\mathcal{L}}}[\boldsymbol{\rho}] + \sum_{\boldsymbol{\mathcal{C}} < \boldsymbol{\mathcal{L}}} [-\mu_{\mathcal{W}}(\boldsymbol{\mathcal{C}}, \boldsymbol{\mathcal{L}})] \mathcal{F}_{\boldsymbol{\mathcal{C}}}[\boldsymbol{\rho}], \tag{3.7}$$

where $\Psi_{\boldsymbol{\mathcal{L}}}[\boldsymbol{\rho}] = g(\boldsymbol{\mathcal{L}})$ is an unknown functional. Expression (3.7) provides a cluster expansion of the *exact* total free-energy density functional of an *arbitrary* system. In general, functional $\Psi_{\boldsymbol{\mathcal{L}}}[\boldsymbol{\rho}]$ cannot be computed exactly, but for suitable choices of the cluster set $\mathcal{W}$ some of its properties can be derived.

Let us make a particular choice: let us assume that $\mathcal{W}$ consists of $\boldsymbol{\mathcal{L}}$ as well as every non-empty intersection of the clusters of certain set $\mathcal{W}_{\max}$. We will then show that $\Psi_{\boldsymbol{\mathcal{L}}}[\boldsymbol{\rho}]$ vanishes for every density profile with support *any* cluster in $\mathcal{W}_{\max}$. (Note that, by construction, the clusters in $\mathcal{W}_{\max}$ are the maximal elements of $\mathcal{W} - \{\boldsymbol{\mathcal{L}}\}$ with respect to the order relation.)

Let $\boldsymbol{\mathcal{D}} \in \mathcal{W}_{\max}$ and $\boldsymbol{\rho}_{\boldsymbol{\mathcal{D}}}$ a density profile with support $\boldsymbol{\mathcal{D}}$. If $\boldsymbol{\mathcal{C}} \in \mathcal{W}$ then we have

$$\mathcal{F}_{\boldsymbol{\mathcal{C}}}[\boldsymbol{\rho}_{\boldsymbol{\mathcal{D}}}] = \mathcal{F}_{\boldsymbol{\mathcal{C}} \cap \boldsymbol{\mathcal{D}}}[\boldsymbol{\rho}_{\boldsymbol{\mathcal{D}}}],$$

with $\mathcal{F}_{\varnothing}[\boldsymbol{\rho}_{\boldsymbol{\mathcal{D}}}] = 0$. By construction, if $\boldsymbol{\mathcal{C}} \cap \boldsymbol{\mathcal{D}} \neq \varnothing$ then $\boldsymbol{\mathcal{C}} \cap \boldsymbol{\mathcal{D}} \in \mathcal{W}$. Taking into account that $\mathcal{F}_{\boldsymbol{\mathcal{L}}}[\boldsymbol{\rho}_{\boldsymbol{\mathcal{D}}}] = \mathcal{F}_{\boldsymbol{\mathcal{D}}}[\boldsymbol{\rho}_{\boldsymbol{\mathcal{D}}}]$, from (3.7) we obtain

$$\Psi_{\boldsymbol{\mathcal{L}}}[\boldsymbol{\rho}_{\boldsymbol{\mathcal{D}}}] = \sum_{\boldsymbol{\mathcal{C}} \in \mathcal{W}} \mu_{\mathcal{W}}(\boldsymbol{\mathcal{C}}, \boldsymbol{\mathcal{L}}) \mathcal{F}_{\boldsymbol{\mathcal{C}} \cap \boldsymbol{\mathcal{D}}}[\boldsymbol{\rho}_{\boldsymbol{\mathcal{D}}}] = \sum_{\boldsymbol{\mathcal{E}} \leqslant \boldsymbol{\mathcal{D}}} \mathcal{F}_{\boldsymbol{\mathcal{E}}}[\boldsymbol{\rho}_{\boldsymbol{\mathcal{D}}}] \sum_{\boldsymbol{\mathcal{C}} \cap \boldsymbol{\mathcal{D}} = \boldsymbol{\mathcal{E}}} \mu_{\mathcal{W}}(\boldsymbol{\mathcal{C}}, \boldsymbol{\mathcal{L}}). \tag{3.8}$$

Now, we can use corollary A.1 in appendix A to show that

$$\sum_{\boldsymbol{\mathcal{C}} \cap \boldsymbol{\mathcal{D}} = \boldsymbol{\mathcal{E}}} \mu_{\mathcal{W}}(\boldsymbol{\mathcal{C}}, \boldsymbol{\mathcal{L}}) = 0 \qquad \text{for all} \quad \boldsymbol{\mathcal{D}} \in \mathcal{W}_{\max} \text{ and } \boldsymbol{\mathcal{E}} < \boldsymbol{\mathcal{L}}. \tag{3.9}$$

Therefore, the unknown functional $\Psi_{\boldsymbol{\mathcal{L}}}$ fulfils the condition

$$\Psi_{\boldsymbol{\mathcal{L}}}[\boldsymbol{\rho}_{\boldsymbol{\mathcal{C}}}] = 0 \qquad \text{for all} \quad \boldsymbol{\mathcal{C}} \in \mathcal{W} - \{\boldsymbol{\mathcal{L}}\}, \tag{3.10}$$

since for every such $\boldsymbol{\mathcal{C}}$ there exists at least one $\boldsymbol{\mathcal{D}} \in \mathcal{W}_{\max}$ such that $\boldsymbol{\mathcal{C}} \leqslant \boldsymbol{\mathcal{D}}$, and so $\boldsymbol{\rho}_{\boldsymbol{\mathcal{C}}}$ is a particular case of $\boldsymbol{\rho}_{\boldsymbol{\mathcal{D}}}$.

Now, if we approximate the exact free-energy functional in (3.7) by the truncation

$$\mathcal{F}_{\text{app}}[\boldsymbol{\rho}] = \sum_{\boldsymbol{\mathcal{C}} < \boldsymbol{\mathcal{L}}} [-\mu_{\mathcal{W}}(\boldsymbol{\mathcal{C}}, \boldsymbol{\mathcal{L}})] \mathcal{F}_{\boldsymbol{\mathcal{C}}}[\boldsymbol{\rho}], \tag{3.11}$$

the previous result (3.10) guarantees that this approximation is exact for any density profile with support any cluster of $\mathcal{W} - \{\boldsymbol{\mathcal{L}}\}$.

At this point, we have shown that given a specific lattice model there exists an approximation of the free-energy functional, given by (3.11), which is exact when the system

is restricted to a certain set of cavities (namely, those in $\mathcal{W}-\{\mathcal{L}\}$). But is it unique? Or in other words, how many free-energy functionals of the form

$$\mathcal{F}_{\rm app}[\rho]=\sum_{\mathcal{C}<\mathcal{L}} a(\mathcal{C})\mathcal{F}_{\mathcal{C}}[\rho] \tag{3.12}$$

are there which are exact in $\mathcal{W}-\{\mathcal{L}\}$? To end this section, we will prove that the only one with this property is (3.11). Before going into the technical details, note that, as in the proof of (3.10), it suffices that the above condition holds for the clusters in $\mathcal{W}_{\rm max}$ (the set of maximal elements of $\mathcal{W}-\{\mathcal{L}\}$).

Let us suppose that we have an approximation like (3.12) such that it satisfies

$$\mathcal{F}_{\rm app}[\rho_{\mathcal{D}}]=\mathcal{F}_{\mathcal{L}}[\rho_{\mathcal{D}}] \tag{3.13}$$

for every cluster $\mathcal{D}\in\mathcal{W}_{\rm max}$. Since $\mathcal{F}_{\mathcal{C}}[\rho_{\mathcal{D}}]=\mathcal{F}_{\mathcal{C}\cap\mathcal{D}}[\rho_{\mathcal{D}}]$, we have from (3.12) and (3.13)

$$\mathcal{F}_{\rm app}[\rho_{\mathcal{D}}]=\mathcal{F}_{\mathcal{D}}[\rho_{\mathcal{D}}]=a(\mathcal{D})\mathcal{F}_{\mathcal{D}}[\rho_{\mathcal{D}}]+\sum_{\mathcal{E}<\mathcal{D}}\mathcal{F}_{\mathcal{E}}[\rho_{\mathcal{D}}]\sum_{\mathcal{C}\cap\mathcal{D}=\mathcal{E}} a(\mathcal{C}). \tag{3.14}$$

This condition should be satisfied regardless the functional form of $\mathcal{F}_{\mathcal{C}}[\rho]$, therefore (3.14) can be cast in the recursion

$$a(\mathcal{D})=1 \qquad \text{for all} \quad \mathcal{D}\in\mathcal{W}_{\rm max}, \qquad \sum_{\mathcal{C}\cap\mathcal{D}=\mathcal{E}} a(\mathcal{C})=0 \qquad \text{for all} \quad \mathcal{E}<\mathcal{D}. \tag{3.15}$$

By defining $a(\mathcal{L})\equiv -1$, the first of these two equations can be rewritten as $\sum_{\mathcal{C}\cap\mathcal{D}=\mathcal{D}} a(\mathcal{C})=0$, so both equations can be gathered in the single one

$$\sum_{\mathcal{C}\cap\mathcal{D}=\mathcal{E}} a(\mathcal{C})=0 \qquad \text{for all} \quad \mathcal{D}\in\mathcal{W}_{\rm max} \text{ and } \mathcal{E}<\mathcal{L}. \tag{3.16}$$

From (3.15) it is clear that this recursion has a unique solution. On the other hand, (3.16) shows that it is formally identical to (3.9). Therefore, since it is linear, the solution must be $a(\mathcal{C})=\lambda\mu_{\mathcal{W}}(\mathcal{C},\mathcal{L})$ for some constant λ. Choosing $\mathcal{C}=\mathcal{L}$ shows that $\lambda=-1$; thus

$$a(\mathcal{C})=-\mu_{\mathcal{W}}(\mathcal{C},\mathcal{L}) \qquad \text{for all} \quad \mathcal{C}\in\mathcal{W}. \tag{3.17}$$

In other words, *the only functional of type* (3.12) *which is exact when restricted to the set of clusters* $\mathcal{W}-\{\mathcal{L}\}$ *is* (3.11).

Let us summarize the main results we have obtained in the following:

Theorem 2. *Given an arbitrary lattice model, a certain set of clusters* $\mathcal{W}_{\rm max}$ *and the poset* $\mathcal{W}$ *formed by all non-empty intersections of elements of* $\mathcal{W}_{\rm max}$ *as well as the cluster* $\mathcal{L}$*, then there exists a unique functional of type (3.12) which is exact when evaluated at density profiles* $\rho_{\mathcal{C}}$ *with support any cluster* $\mathcal{C}\in\mathcal{W}-\{\mathcal{L}\}$*. This functional is given by*

$$\mathcal{F}_{\rm app}[\rho]=\sum_{\mathcal{C}\in\mathcal{W}-\{\mathcal{L}\}}[-\mu_{\mathcal{W}}(\mathcal{C},\mathcal{L})]\mathcal{F}_{\mathcal{C}}[\rho], \tag{3.18}$$

where the integer coefficients $\mu_{\mathcal{W}}(\mathcal{C},\mathcal{L})$ *are defined by either recursion 1 (equation (3.3)) or recursion 2 (equation (3.4)).*

For some special cases, formula (3.18) is exact. One of them occurs if $\mathcal{F}_{\mathcal{C}}[\rho]$ is a local functional of $\rho(\boldsymbol{s})$, i.e.

$$\mathcal{F}_{\mathcal{C}}[\rho]=\sum_{i=1}^{p}\sum_{\boldsymbol{s}\in\mathcal{C}_i}\phi(\rho_i(\boldsymbol{s})). \tag{3.19}$$

With this particular choice for $\mathcal{F}_{\mathcal{C}}[\rho]$, the 'error' functional (3.7) becomes

$$\Psi_{\mathcal{L}}[\rho] = \sum_{\mathcal{C}\in\mathcal{W}} \mu_{\mathcal{W}}(\mathcal{C},\mathcal{L})\mathcal{F}_{\mathcal{C}}[\rho] = \sum_{i=1}^{p}\sum_{s\in\mathcal{L}} \phi(\rho_i(s))h_i(s),$$

where $h_i(s) = \sum_{\mathcal{C}\in\mathcal{W}} \mu_{\mathcal{W}}(\mathcal{C},\mathcal{L})\chi_{\mathcal{C}_i}(s)$ and $\chi_{\mathcal{C}_i}(s) = 1$, if $s \in \mathcal{C}_i$ and 0 otherwise, is the indicator function of the set $\mathcal{C}_i$. If we now define the cluster $\sigma_i(s) \equiv \big(\varnothing, \ldots, \varnothing, \overset{(i)}{\circ_s}, \varnothing, \ldots, \varnothing\big)$, the function $h_i(s)$ can be rewritten as

$$h_i(s) = \sum_{\mathcal{C}\cap\sigma_i(s)=\sigma_i(s)} \mu_{\mathcal{W}}(\mathcal{C},\mathcal{L}),$$

and because of corollary A.1 of appendix A, $h_i(s) = 0$ for all $s \in \mathcal{L}$ and $i = 1, \ldots, p$. Hence, $\Psi_{\mathcal{L}}[\rho] = 0$.

4. Lattice fundamental-measure theory revisited

The Möbius formalism developed in section 3 endows us with a very powerful alternative procedure to obtain the FM excess free-energy functional for any lattice system. The approximation (3.18) expresses the free-energy functional as a truncated cluster expansion of the form (3.1). Given that the ideal part of the functional is local in $\rho(s)$ and, as we have just shown, for local functionals this cluster expansion is exact (regardless the choice of $\mathcal{W}_{\max}$), the approximation is actually made on the excess free-energy functional. If we now take for $\mathcal{W}_{\max}$ the set of maximal 1-particle cavities of a given lattice model, then theorem 2 tells us that the FM excess free-energy functional will be given by

$$\mathcal{F}^{\mathrm{ex}}_{\mathrm{FM}}[\rho] = \sum_{\mathcal{C}\in\mathcal{W}-\{\mathcal{L}\}} [-\mu_{\mathcal{W}}(\mathcal{C},\mathcal{L})]\Phi_0(n_{\mathcal{C}}[\rho]), \tag{4.1}$$

where $n_{\mathcal{C}}[\rho] \equiv \sum_{i=1}^{p}\sum_{t\in\mathcal{C}_i}\rho_i(t)$. This expression is identical to (2.1), because in $\mathcal{W}-\{\mathcal{L}\}$ there will be clusters of different shapes (labelled by $k \in \mathcal{I}$ in (2.1)) and all their translates (labelled by $s \in \mathcal{L}$ in (2.1)). But now, thanks to theorem 2, we know that this is the only functional one can obtain with this particular choice for $\mathcal{W}_{\max}$ which is exact when evaluated at any 0D density profile with support any cluster in $\mathcal{W}-\{\mathcal{L}\}$.

A remark is in order here. If $\mathcal{C}$ is a cluster of $\mathcal{W}-\{\mathcal{L}\}$, and $\mathcal{C}'$ is a translation of $\mathcal{C}$, then $\mu_{\mathcal{W}}(\mathcal{C},\mathcal{L}) = \mu_{\mathcal{W}}(\mathcal{C}',\mathcal{L})$ because the Möbius function $\mu_{\mathcal{W}}(x,y)$ only depends on the interval $[x,y]$ (cf its definition in recursion 1) and the translation operation is an obvious order-preserving isomorphism between $[\mathcal{C},\mathcal{L}]$ and $[\mathcal{C}',\mathcal{L}]$. This justifies why the coefficients a_k in (2.1) are independent of $s \in \mathcal{L}$.

With this new formulation of LFMT it is a simple task to obtain the unknowns in expression (2.1). As an illustration, let us re-derive functional (2.12). The first step is to fix the appropriate cluster poset $\mathcal{W}$, which contains all non-empty intersections of maximal 1-particle cavities in the set $\mathcal{W}_{\max}$ described in (2.5), as well as the cluster $\mathcal{L}$. Thus,

$\mathcal{W} = \{$ $\mid s \in \mathcal{L}\} \cup \{\mathcal{L}\}.$ (4.2)

The second and last step is to compute the Möbius function $\mu_{\mathcal{W}}(\mathcal{C},\mathcal{L})$ for every $\mathcal{C} \in \mathcal{W}-\{\mathcal{L}\}$ (note that $\mu_{\mathcal{W}}(\mathcal{L},\mathcal{L}) = 1$). This can be easily done by resorting to recursion 1. The natural iteration prescribed by this recursion is to start with maximal clusters (those in $\mathcal{W}_{\max}$) and then to follow a decreasing path. Note that in order to obtain $\mu_{\mathcal{W}}(\mathcal{C},\mathcal{L})$ we only need the values of the Möbius function for the clusters in the interval $(\mathcal{C},\mathcal{L}]$. To carry on this task, it is useful to draw the Hasse diagram of the corresponding interval (see figure 1), since it explicitly shows the order structure of the latter.

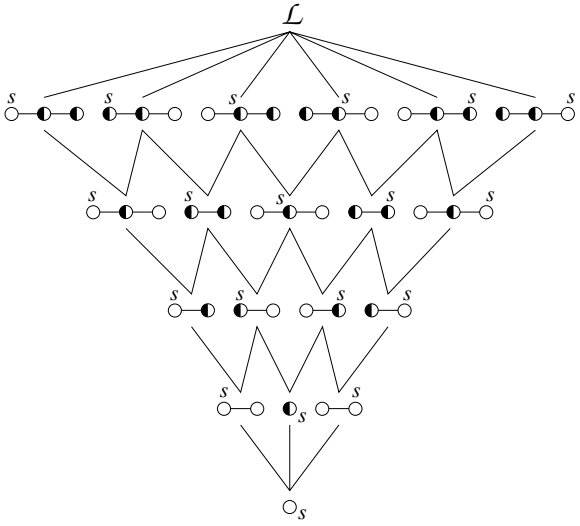


Figure 1. The *Hasse diagram* of a finite poset $\mathcal{W}$ is defined as the graph whose vertices are the elements of the poset and there is an edge between elements x and y if $x < y$ and there is no $z \in \mathcal{W}$ such that $x < z < y$. If $x < y$, then y is drawn at higher level than x. This figure shows the Hasse diagram of the interval $[\circ_s, \mathcal{L}]$. Nodes connected through a descending path are ordered by transitivity. For this reason, a Hasse diagram is a very practical way of visualizing the order in a finite set.

In our example, we should start with $\overset{s}{\bullet}\!\!-\!\!\bullet\!\!-\!\!\circ$ and $\overset{s}{\circ}\!\!-\!\!\bullet\!\!-\!\!\bullet$ (remember that the Möbius function does not depend on s, so what follows holds for any $s \in \mathcal{L}$). As both are maximal clusters, then $(\overset{s}{\bullet}\!\!-\!\!\bullet\!\!-\!\!\circ, \mathcal{L}] = (\overset{s}{\circ}\!\!-\!\!\bullet\!\!-\!\!\bullet, \mathcal{L}] = \{\mathcal{L}\}$ (by definition, this is always true for maximal clusters). By applying recursion 1, we obtain

$$\mu_{\mathcal{W}}(\overset{s}{\bullet}\!\!-\!\!\bullet\!\!-\!\!\circ, \mathcal{L}) = \mu_{\mathcal{W}}(\overset{s}{\circ}\!\!-\!\!\bullet\!\!-\!\!\bullet, \mathcal{L}) = -\mu_{\mathcal{W}}(\mathcal{L}, \mathcal{L}) = -1.$$

In decreasing order, the next set of clusters involves $\overset{s}{\circ}\!\!-\!\!\bullet\!\!-\!\!\circ$ and $\overset{s}{\bullet}\!\!-\!\!\bullet$, and we have

$$(\overset{s}{\circ}\!\!-\!\!\bullet\!\!-\!\!\circ, \mathcal{L}] = \{\overset{s}{\bullet}\!\!-\!\!\bullet\!\!-\!\!\circ, \overset{s}{\circ}\!\!-\!\!\bullet\!\!-\!\!\bullet, \mathcal{L}\}, \qquad (\overset{s}{\bullet}\!\!-\!\!\bullet, \mathcal{L}] = \{\overset{s}{\bullet}\!\!-\!\!\bullet\!\!-\!\!\circ, \circ\!\!-\!\!\overset{s}{\bullet}\!\!-\!\!\bullet, \mathcal{L}\}.$$

Thus $\mu_{\mathcal{W}}(\overset{s}{\circ}\!\!-\!\!\bullet\!\!-\!\!\circ, \mathcal{L}) = \mu_{\mathcal{W}}(\bullet\!\!-\!\!\bullet, \mathcal{L}) = 1$. Then we find $\overset{s}{\bullet}\!\!-\!\!\circ$ and $\overset{s}{\circ}\!\!-\!\!\bullet$. By symmetry, $\mu_{\mathcal{W}}(\overset{s}{\bullet}\!\!-\!\!\circ, \mathcal{L}) = \mu_{\mathcal{W}}(\overset{s}{\circ}\!\!-\!\!\bullet, \mathcal{L})$, and since

$$(\overset{s}{\bullet}\!\!-\!\!\circ, \mathcal{L}] = \{\overset{s}{\bullet}\!\!-\!\!\bullet, \circ\!\!-\!\!\overset{s}{\bullet}, \overset{s}{\bullet}\!\!-\!\!\bullet\!\!-\!\!\circ, \bullet\!\!-\!\!\overset{s}{\bullet}\!\!-\!\!\circ, \circ\!\!-\!\!\overset{s}{\bullet}\!\!-\!\!\bullet, \mathcal{L}\},$$

we will have $\mu_{\mathcal{W}}(\overset{s}{\bullet}\!\!-\!\!\circ, \mathcal{L}) = 0$. Next, we have $\overset{s}{\circ}\!\!-\!\!\circ$ and $\bullet_s$. The corresponding intervals are, respectively,

$$(\overset{s}{\circ}\!\!-\!\!\circ, \mathcal{L}] = \{\overset{s}{\bullet}\!\!-\!\!\circ, \overset{s}{\circ}\!\!-\!\!\bullet, \overset{s}{\bullet}\!\!-\!\!\bullet, \overset{s}{\circ}\!\!-\!\!\bullet\!\!-\!\!\circ, \circ\!\!-\!\!\overset{s}{\bullet}\!\!-\!\!\circ, \overset{s}{\bullet}\!\!-\!\!\bullet\!\!-\!\!\circ, \bullet\!\!-\!\!\overset{s}{\bullet}\!\!-\!\!\circ, \overset{s}{\circ}\!\!-\!\!\bullet\!\!-\!\!\bullet, \circ\!\!-\!\!\overset{s}{\bullet}\!\!-\!\!\bullet, \mathcal{L}\}$$

and

$$(\bullet_s, \mathcal{L}] = \{\overset{s}{\bullet}\!\!-\!\!\circ, \circ\!\!-\!\!\overset{s}{\bullet}, \overset{s}{\bullet}\!\!-\!\!\bullet, \bullet\!\!-\!\!\overset{s}{\bullet}, \circ\!\!-\!\!\overset{s}{\bullet}\!\!-\!\!\circ, \overset{s}{\bullet}\!\!-\!\!\bullet\!\!-\!\!\circ, \bullet\!\!-\!\!\overset{s}{\bullet}\!\!-\!\!\circ, \circ\!\!-\!\!\overset{s}{\bullet}\!\!-\!\!\bullet, \circ\!\!-\!\!\bullet\!\!-\!\!\overset{s}{\bullet}, \mathcal{L}\}.$$

Therefore $\mu_{\mathcal{W}}(\overset{s}{\circ}\!\!-\!\!\circ, \mathcal{L}) = 0$ and $\mu_{\mathcal{W}}(\bullet, \mathcal{L}) = 0$. Finally, for $\circ_s$ we have

$$(\circ_s, \mathcal{L}] = \{\bullet_s, \overset{s}{\circ}\!\!-\!\!\circ, \circ\!\!-\!\!\overset{s}{\circ}, \overset{s}{\bullet}\!\!-\!\!\circ, \bullet\!\!-\!\!\overset{s}{\circ}, \overset{s}{\circ}\!\!-\!\!\bullet, \circ\!\!-\!\!\overset{s}{\bullet}, \overset{s}{\bullet}\!\!-\!\!\bullet, \bullet\!\!-\!\!\overset{s}{\bullet},$$
$$\overset{s}{\circ}\!\!-\!\!\bullet\!\!-\!\!\circ, \circ\!\!-\!\!\overset{s}{\bullet}\!\!-\!\!\circ, \circ\!\!-\!\!\bullet\!\!-\!\!\overset{s}{\circ}, \overset{s}{\bullet}\!\!-\!\!\bullet\!\!-\!\!\circ, \bullet\!\!-\!\!\overset{s}{\bullet}\!\!-\!\!\circ, \bullet\!\!-\!\!\bullet\!\!-\!\!\overset{s}{\circ}, \overset{s}{\circ}\!\!-\!\!\bullet\!\!-\!\!\bullet, \circ\!\!-\!\!\overset{s}{\bullet}\!\!-\!\!\bullet, \circ\!\!-\!\!\bullet\!\!-\!\!\overset{s}{\bullet}, \mathcal{L}\}$$

which leads to $\mu_{\mathcal{W}}(\circ, \mathcal{L}) = 0$. Substituting these values of the Möbius function in the general expression (4.1), we recover functional (2.12).

Although in our example we have shown that recursion 1 is enough to compute the Möbius function, there are many alternative (more efficient) techniques which exploit the order structure of the cluster poset associated with the given lattice model and make use of specialized results of the theory of posets (see section 3.8 of Stanley (1999) and section IV.3 of Aigner (1979)). In appendix A, we provide a few examples of these tools.

5. Extending lattice fundamental-measure theory

In the previous section, we have achieved a reformulation of LFMT based on theorem 2. Actually, we have shown that LFMT is a particular case of the approximation proposed in that theorem, where we choose the cluster set $\mathcal{W}_{\max}$ as the set of maximal 1-particle cavities. We will see in the next section that, for hard-core models, this choice is an excellent balance between accuracy and simplicity of the approximate functional. But we want to stress that the cluster expansion of the free-energy functional (3.7) is more general: it applies not only to hard-core models, but is valid for *any* lattice model.

In order to illustrate this, let us consider the Ising lattice gas in an arbitrary lattice $\mathcal{L}$. The interaction potential for this system is such that each lattice node can be occupied at most by one particle and two particles interact with an energy J if they are placed at nodes which are nearest neighbours. If we applied theorem 2 to this system with $\mathcal{W}_{\max}$ the set of maximal 1-particle cavities (which for this model is just the set of all single lattice nodes), the approximate free-energy functional we would obtain would be that of the site-excluding ideal lattice gas, i.e.

$$\mathcal{F}_{\rm app}[\rho] = \sum_{\boldsymbol{s}\in\mathcal{L}} [\rho(\boldsymbol{s})\ln\rho(\boldsymbol{s}) + (1-\rho(\boldsymbol{s}))\ln(1-\rho(\boldsymbol{s}))].$$

This choice for $\mathcal{W}_{\max}$ is clearly inappropriate for this system, since it ignores the interaction between nearest neighbours. In order to account for it we should go beyond the standard LFMT and take 2-particle cavities. Thus, $\mathcal{W}_{\max}$ = {all pairs of nearest neighbours} and $\mathcal{W} = \{\mathcal{L}\} \cup \{\text{all pairs of nearest neighbours}\} \cup \{\text{all single nodes}\}$. The Möbius function takes the values $\mu_{\mathcal{W}}(\{\boldsymbol{s},\boldsymbol{t}\},\mathcal{L}) = -1$ and $\mu_{\mathcal{W}}(\{\boldsymbol{s}\},\mathcal{L}) = q(\boldsymbol{s})-1$ for every pair of nearest neighbours $\{\boldsymbol{s},\boldsymbol{t}\}$ and every single node $\{\boldsymbol{s}\}$ of $\mathcal{L}$, $q(\boldsymbol{s})$ being the coordination number at node $\boldsymbol{s}$. From theorem 2, the approximate free-energy functional will be

$$\mathcal{F}_{\rm app}[\rho] = \sum_{\text{all n.n. } \{\boldsymbol{s},\boldsymbol{t}\}} \mathcal{F}_{\{\boldsymbol{s},\boldsymbol{t}\}}[\rho] - \sum_{\boldsymbol{s}\in\mathcal{L}} [q(\boldsymbol{s})-1]\mathcal{F}_{\{\boldsymbol{s}\}}[\rho], \tag{5.1}$$

where

$$\begin{aligned}\mathcal{F}_{\{\boldsymbol{s},\boldsymbol{t}\}}[\rho] = {}& \rho(\boldsymbol{s})\ln[\rho(\boldsymbol{s}) - \rho^{(2)}(\boldsymbol{s},\boldsymbol{t})] + \rho(\boldsymbol{t})\ln[\rho(\boldsymbol{t}) - \rho^{(2)}(\boldsymbol{s},\boldsymbol{t})] \\ & + [1-\rho(\boldsymbol{s})-\rho(\boldsymbol{t})]\ln[1-\rho(\boldsymbol{s})-\rho(\boldsymbol{t})+\rho^{(2)}(\boldsymbol{s},\boldsymbol{t})],\end{aligned} \tag{5.2}$$

$\rho^{(2)}(\boldsymbol{s},\boldsymbol{t})$ being the joint probability of finding two particles at nodes $\boldsymbol{s}$ and $\boldsymbol{t}$. For this model, it is not difficult to show that it can be eliminated in terms of $\rho(\boldsymbol{s})$ and $\rho(\boldsymbol{t})$ as

$$\rho^{(2)}(\boldsymbol{s},\boldsymbol{t}) = \frac{1+\zeta[\rho(\boldsymbol{s})+\rho(\boldsymbol{t})] - \sqrt{\{1+\zeta[\rho(\boldsymbol{s})+\rho(\boldsymbol{t})]\}^2 - 4\zeta(1+\zeta)\rho(\boldsymbol{s})\rho(\boldsymbol{t})}}{2\zeta},$$

with $\zeta = \exp(-J) - 1$ (J is the interaction energy between nearest neighbours in kT units). Note that $\mathcal{F}_{\{\boldsymbol{s}\}}[\rho]$ can be obtained from (5.2) by setting $\rho(\boldsymbol{t}) = 0$ and is just $\Phi_0(\rho(\boldsymbol{s}))$. This approximation is exact when the lattice $\mathcal{L}$ is a Bethe lattice[4] (even with node-dependent coordination numbers) and it is equivalent to the Bethe approximation for any other lattice (Bowman and Levin 1982).

[4] In particular, it is exact for the one-dimensional lattice $\mathbb{Z}$.

6. Low-density limit of the approximate functional

A fundamental ingredient of many approximate density functional theories is the available exact information about the low-density limit (Evans 1992; see also Cuesta *et al* (2002) for an analysis of this limit in the construction of the *weighted density approximation* and the FMT). In contrast, in our formulation of LFMT we have input the exact functionals for the system restricted to a certain set of clusters and used a combinatorial tool (Möbius inversion) to construct an 'optimal' functional out of them. The latter is so restrictive that the only freedom we have in the resulting approximation (3.18) is limited to the choice of $\mathcal{W}_{\max}$. In this section, we will prove that with an appropriate choice of this set we can assure the correct behaviour of the approximate functional in the low-density limit up to, at least, third order.

The second functional derivative of the excess part of the exact free-energy functional yields the exact pair direct correlation functional

$$c_{ij}^{(2)}(\boldsymbol{s}_1,\boldsymbol{s}_2) = -\frac{\delta^2\mathcal{F}_{\mathcal{L}}^{\mathrm{ex}}[\boldsymbol{\rho}]}{\delta\rho_i(\boldsymbol{s}_1)\delta\rho_j(\boldsymbol{s}_2)} \sim f_{ij}(\boldsymbol{s}_1,\boldsymbol{s}_2)\left[1+\sum_k\sum_{\boldsymbol{s}_3\in\mathcal{L}} f_{ik}(\boldsymbol{s}_1,\boldsymbol{s}_3)\rho_k(\boldsymbol{s}_3)f_{kj}(\boldsymbol{s}_3,\boldsymbol{s}_2)\right] \qquad (\boldsymbol{\rho}\to\mathbf{0}), \tag{6.1}$$

where $f_{ij}(\boldsymbol{s}_1,\boldsymbol{s}_2)\equiv \mathrm{e}^{-\phi_{ij}(\boldsymbol{s}_1,\boldsymbol{s}_2)}-1$ is the Mayer function, $\phi_{ij}(\boldsymbol{s}_1,\boldsymbol{s}_2)$ being the interaction potential (in kT units) between a particle of species i at node $\boldsymbol{s}_1$ and another of species j at node $\boldsymbol{s}_2$ (the interaction potential is assumed pairwise). If we compute the pair direct correlation functional from the approximate functional (3.18), we obtain

$$c_{\mathrm{app},ij}^{(2)}(\boldsymbol{s}_1,\boldsymbol{s}_2) = \sum_{\mathcal{C}\in\mathcal{W}-\{\mathcal{L}\}}[-\mu_{\mathcal{W}}(\mathcal{C},\mathcal{L})]c_{ij}^{(2)}(\boldsymbol{s}_1,\boldsymbol{s}_2|\mathcal{C}),$$

where we have introduced

$$c_{ij}^{(2)}(\boldsymbol{s}_1,\boldsymbol{s}_2|\mathcal{C}) \equiv -\frac{\delta^2\mathcal{F}_{\mathcal{C}}^{\mathrm{ex}}[\boldsymbol{\rho}]}{\delta\rho_i(\boldsymbol{s}_1)\delta\rho_j(\boldsymbol{s}_2)},$$

the *exact* pair direct correlation functional for the system restricted to cluster $\mathcal{C}$. Since $c_{ij}^{(2)}(\boldsymbol{s}_1,\boldsymbol{s}_2|\mathcal{C})$ is exact, we have from (6.1) that

$$c_{ij}^{(2)}(\boldsymbol{s}_1,\boldsymbol{s}_2|\mathcal{C}) \sim f_{ij}(\boldsymbol{s}_1,\boldsymbol{s}_2)\chi_{\mathcal{C}_i}(\boldsymbol{s}_1)\chi_{\mathcal{C}_j}(\boldsymbol{s}_2) \qquad (\boldsymbol{\rho}\to\mathbf{0}),$$

with $\chi_{\mathcal{C}}(\boldsymbol{s})$ the indicator function of $\mathcal{C}$. Thus, the approximate pair direct correlation functional satisfies

$$c_{\mathrm{app},ij}^{(2)}(\boldsymbol{s}_1,\boldsymbol{s}_2) \sim f_{ij}(\boldsymbol{s}_1,\boldsymbol{s}_2)\sum_{\mathcal{C}\in\mathcal{W}-\{\mathcal{L}\}}[-\mu_{\mathcal{W}}(\mathcal{C},\mathcal{L})]\chi_{\mathcal{C}_i}(\boldsymbol{s}_1)\chi_{\mathcal{C}_j}(\boldsymbol{s}_2) \qquad (\boldsymbol{\rho}\to\mathbf{0}).$$

Taking into account that $\mu_{\mathcal{W}}(\mathcal{L},\mathcal{L})=1$ and denoting $\boldsymbol{\sigma}_{ij}(\boldsymbol{s}_1,\boldsymbol{s}_2)\equiv\boldsymbol{\sigma}_i(\boldsymbol{s}_1)\cup\boldsymbol{\sigma}_j(\boldsymbol{s}_2)$, we can rewrite the above expression in the more suitable form

$$c_{\mathrm{app},ij}^{(2)}(\boldsymbol{s}_1,\boldsymbol{s}_2) \sim f_{ij}(\boldsymbol{s}_1,\boldsymbol{s}_2)\left(1-\sum_{\boldsymbol{\sigma}_{ij}(\boldsymbol{s}_1,\boldsymbol{s}_2)\leqslant\mathcal{C}}\mu_{\mathcal{W}}(\mathcal{C},\mathcal{L})\right) \qquad (\boldsymbol{\rho}\to\mathbf{0}).$$

Now, inasmuch as $\boldsymbol{\sigma}_{ij}(\boldsymbol{s}_1,\boldsymbol{s}_2)\leqslant\mathcal{C}$ is equivalent to $\mathcal{C}\cap\boldsymbol{\sigma}_{ij}(\boldsymbol{s}_1,\boldsymbol{s}_2)=\boldsymbol{\sigma}_{ij}(\boldsymbol{s}_1,\boldsymbol{s}_2)$, it is a direct consequence of corollary A.1 in appendix A that, when $\boldsymbol{\rho}\to\mathbf{0}$,

$$c_{\mathrm{app},ij}^{(2)}(\boldsymbol{s}_1,\boldsymbol{s}_2) \sim \begin{cases} f_{ij}(\boldsymbol{s}_1,\boldsymbol{s}_2) & \text{if } \boldsymbol{\sigma}_{ij}(\boldsymbol{s}_1,\boldsymbol{s}_2) \text{ is contained in any cluster of } \mathcal{W}_{\max},\\ 0 & \text{otherwise.}\end{cases}$$

The Mayer function has the same range of the interaction potential; when the latter is short ranged, $f_{ij}(\boldsymbol{s}_1, \boldsymbol{s}_2)$ vanishes if a particle of species i at $\boldsymbol{s}_1$ does not interact with a particle of species j at $\boldsymbol{s}_2$. Therefore, if we want the approximate pair direct correlation functional to recover the exact low-density limit, it must happen that *for any pair of nodes $\boldsymbol{s}_1$ and $\boldsymbol{s}_2$ for which $f_{ij}(\boldsymbol{s}_1, \boldsymbol{s}_2) \neq 0$ the cluster $\boldsymbol{\sigma}_{ij}(\boldsymbol{s}_1, \boldsymbol{s}_2)$ is contained in at least one cluster of $\mathcal{W}_{\max}$*.

Let us consider the particular case of hard-core interaction. For these systems, two particles interact if and only if they overlap. Accordingly, we can define a 0D cavity as *any cluster such that if we place two particles in any pair of its nodes* (of any allowed species), *they necessarily interact* (in which case the corresponding Mayer function is non-zero). From this definition, it should be clear that if we let $\mathcal{W}_{\max}$ be the set of maximal 0D cavities, the density expansion of the approximate pair direct correlation functional will recover the exact zeroth order.

Also, let us reconsider the Ising lattice gas. We showed in section 5 that the choice of $\mathcal{W}_{\max}$ as the set of maximal 1-particle cavities gives rise to a very poor approximation of the free-energy functional. Then, we took $\mathcal{W}_{\max}$ as the set of all pairs of nearest neighbours in order to account for the interaction. This set contains all maximal 0D cavities according to the new definition, and so we are certain that the free-energy functional (5.1) gives a pair direct correlation functional with the exact zeroth-order term in the density expansion. The new definition of 0D cavity that we have just introduced is thus suitable for any kind of interaction (whether hard or soft, repulsive or attractive) in the sense that choosing $\mathcal{W}_{\max}$ as the set of all maximal 0D cavities guarantees the correct low-density limit of $c^{(2)}_{\mathrm{app},ij}(\boldsymbol{s}_1, \boldsymbol{s}_2)$.

Now, let us look at higher order terms in the density expansion. If we take the third functional derivative of the excess part of the free-energy functional, we obtain the so-called triplet direct correlation functional

$$c^{(3)}_{ijk}(\boldsymbol{s}_1, \boldsymbol{s}_2, \boldsymbol{s}_3) = -\frac{\delta^3 \mathcal{F}^{\mathrm{ex}}_{\mathcal{L}}[\boldsymbol{\rho}]}{\delta\rho_i(\boldsymbol{s}_1)\delta\rho_j(\boldsymbol{s}_2)\delta\rho_k(\boldsymbol{s}_3)} \sim f_{ij}(\boldsymbol{s}_1, \boldsymbol{s}_2) f_{jk}(\boldsymbol{s}_2, \boldsymbol{s}_3) f_{ki}(\boldsymbol{s}_3, \boldsymbol{s}_1) \qquad (\boldsymbol{\rho} \to \mathbf{0}).$$

Computing $c^{(3)}_{\mathrm{app},ijk}(\boldsymbol{s}_1, \boldsymbol{s}_2, \boldsymbol{s}_3)$ from (3.18) yields (reproducing the arguments given to obtain $c^{(2)}_{\mathrm{app},ij}$), when $\boldsymbol{\rho} \to \mathbf{0}$,

$$c^{(3)}_{ijk}(\boldsymbol{s}_1, \boldsymbol{s}_2, \boldsymbol{s}_3) \sim f_{ij}(\boldsymbol{s}_1, \boldsymbol{s}_2) f_{jk}(\boldsymbol{s}_2, \boldsymbol{s}_3) f_{ki}(\boldsymbol{s}_3, \boldsymbol{s}_1) \left(1 - \sum_{\boldsymbol{\sigma}_{ijk}(\boldsymbol{s}_1, \boldsymbol{s}_2, \boldsymbol{s}_3) \lesssim \mathcal{C}} \mu_{\mathcal{W}}(\mathcal{C}, \mathcal{L}) \right),$$

where $\boldsymbol{\sigma}_{ijk}(\boldsymbol{s}_1, \boldsymbol{s}_2, \boldsymbol{s}_3) \equiv \boldsymbol{\sigma}_i(\boldsymbol{s}_1) \cup \boldsymbol{\sigma}_j(\boldsymbol{s}_2) \cup \boldsymbol{\sigma}_k(\boldsymbol{s}_3)$. Therefore, we are guaranteed to recover the exact low-density limit of the triplet direct correlation functional if *every cluster $\boldsymbol{\sigma}_{ijk}(\boldsymbol{s}_1, \boldsymbol{s}_2, \boldsymbol{s}_3)$ for which the product $f_{ij}(\boldsymbol{s}_1, \boldsymbol{s}_2) f_{jk}(\boldsymbol{s}_2, \boldsymbol{s}_3) f_{ki}(\boldsymbol{s}_3, \boldsymbol{s}_1)$ does not vanish is contained in at least one cluster of $\mathcal{W}_{\max}$*. Note that because of (6.1), if this holds then the density expansion of the pair direct correlation functional is exact up to first order in $\boldsymbol{\rho}$.

Again, the choice of $\mathcal{W}_{\max}$ as the set of maximal 0D cavities (according to the new definition) is enough to assure the correct behaviour of $c^{(3)}_{\mathrm{app},ijk}(\boldsymbol{s}_1, \boldsymbol{s}_2, \boldsymbol{s}_3)$ in the low-density limit. To see this, just note that the product $f_{ij}(\boldsymbol{s}_1, \boldsymbol{s}_2) f_{jk}(\boldsymbol{s}_2, \boldsymbol{s}_3) f_{ki}(\boldsymbol{s}_3, \boldsymbol{s}_1)$ is different from zero only if two particles of the corresponding species placed at any pair of nodes $\{\boldsymbol{s}_1, \boldsymbol{s}_2, \boldsymbol{s}_3\}$ interact; in other words, only if the nodes belong to the same maximal 0D cavity. Therefore, *the approximate free-energy functional (3.18) with $\mathcal{W}_{\max}$ the set of maximal 0D cavities, recovers the exact density expansion of the pair and triplet direct correlation functional up to first and zeroth order, respectively*.

All this analysis can in principle be extended to higher order direct correlation functions. In general, we will have that $c^{(n)}_{\mathrm{app},i_1 \ldots i_n}(\boldsymbol{s}_1, \ldots, \boldsymbol{s}_n)$ will recover the exact low-density limit

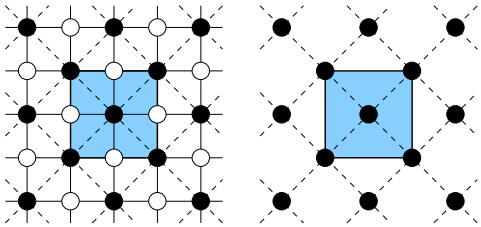


Figure 2. The system on the left corresponds to the lattice gas with first- and second-neighbour exclusion in the square lattice. If the position of the particles is confined to the black nodes of the lattice, the system behaves exactly like the lattice gas with nearest-neighbour exclusion in the square lattice (represented in the right figure).

(This figure is in colour only in the electronic version)

provided the cluster $\boldsymbol{\sigma}_{i_1\dots i_n}(\boldsymbol{s}_1,\dots,\boldsymbol{s}_n) \equiv \bigcup_{l=1}^{n}\boldsymbol{\sigma}_{i_l}(\boldsymbol{s}_l)$ is contained in at least one cluster of $\mathcal{W}_{\max}$ for any combination of $\{(i_1,\boldsymbol{s}_1),\dots,(i_n,\boldsymbol{s}_n)\}$ for which the exact low-density limit is different from zero. In practice, this is a very demanding task, because guaranteeing the correct low-density limit of $c^{(n)}_{\mathrm{app},i_1\dots i_n}(\boldsymbol{s}_1,\dots,\boldsymbol{s}_n)$ amounts to consider all diagrams in the virial expansion of the n th order direct correlation function, and the number of them grows exponentially. Clearly, the current definition of 0D cavity is insufficient to provide the correct behaviour beyond $n = 3$ (except in very particular cases, like some one-dimensional systems for which the approximation becomes exact), so if we want more terms in the low-density expansion we have to take bigger maximal 0D cavities. Bigger cavities means more particles in one cavity and an increasingly higher difficulty to obtain the exact free-energy functional for a single cavity. So, as is usual with expansions, although a systematic improvement is possible, going beyond the lowest terms may be too involved in practice.

7. Dimensional reduction

One of the most remarkable properties of FMT is dimensional crossover. This property means that by confining the particles of a d-dimensional system to lie in a $(d-1)$-dimensional subset we obtain the $(d-1)$-dimensional FM functional out of the d-dimensional one. In the case of LFMT, a typical example would be to start with the lattice gas with nearest neighbour exclusion in the simple cubic lattice in three dimensions and constrain the position of the particles to the nodes of one of the coordinate planes in order to obtain an effective system equivalent to the lattice gas with nearest-neighbour exclusion in the square lattice. In this example, the real dimension of the system is reduced from 3 to 2, and in so doing, the FM functional of the three-dimensional system is transformed into that of the two-dimensional one (Lafuente and Cuesta 2003). But this is only an instance of a more general class of mappings between different models. As another example, the FM of the lattice gas with nearest-neighbour exclusion in the square lattice can also be obtained from the one of the lattice gas with first- and second-neighbour exclusion in the same lattice, as figure 2 illustrates (Lafuente and Cuesta 2003).

The aim of this section is to define a general mapping between models (of which these two examples are particular cases) and to prove that the approximation (3.18) is 'closed' with respect to this kind of mapping, i.e. that the FM functional of the original model becomes the one of the transformed model under the action of that mapping on the density profile.

Let us assume that the original system has an underlying lattice $\mathcal{L}$ and its approximate free-energy functional is given from theorem 2 by

$$\mathcal{F}_{\rm app}[\rho] = \sum_{\mathcal{C}\in\mathcal{W}-\{\mathcal{L}\}} [-\mu_{\mathcal{W}}(\mathcal{C},\mathcal{L})]\mathcal{F}_{\mathcal{C}}[\rho] \tag{7.1}$$

for a given choice of $\mathcal{W}_{\rm max}$. If we restrict the position of the particles of species i to the embedded lattice $\mathcal{L}'_i \subset \mathcal{L}$ (lattice here is a general term which refers to any subset of $\mathcal{L}$, finite or infinite), the approximate functional of the new effective system can be obtained by specializing functional (7.1) to a density profile with support $\mathcal{L}' = (\mathcal{L}'_1, \ldots, \mathcal{L}'_p)$, i.e.

$$\mathcal{F}'_{\rm app}[\rho] = \mathcal{F}_{\rm app}[\rho_{\mathcal{L}'}] = \sum_{\mathcal{C}\in\mathcal{W}-\{\mathcal{L}\}} [-\mu_{\mathcal{W}}(\mathcal{C},\mathcal{L})]\mathcal{F}_{\mathcal{C}\cap\mathcal{L}'}[\rho_{\mathcal{L}'}],$$

where $\rho_{\mathcal{L}'}(s) = \rho(s)$ if $s \in \mathcal{L}'$ and is 0 otherwise. Since $\mathcal{F}_\phi[\rho] = 0$, if $\mathcal{W}'$ denotes the set of all non-empty intersections of the clusters in $\mathcal{W}$ with the cluster $\mathcal{L}'$, the above expression becomes

$$\mathcal{F}'_{\rm app}[\rho] = \sum_{\mathcal{C}'\in\mathcal{W}'} \mathcal{F}_{\mathcal{C}'}[\rho] \sum_{\mathcal{C}\cap\mathcal{L}'=\mathcal{C}'} [-\mu_{\mathcal{W}}(\mathcal{C},\mathcal{L}) + \delta(\mathcal{C},\mathcal{L})], \tag{7.2}$$

where we have introduced the identity of $I(\mathcal{W},\mathbb{K})$, $\delta(\mathcal{C},\mathcal{L})$, to compensate for the inclusion of $\mu_{\mathcal{W}}(\mathcal{L},\mathcal{L}) = 1$ in the sum. We can now apply corollary A.1 in appendix A to evaluate the constrained sum on the rhs of (7.2). If $\mathcal{L}'$ is contained in some cluster of $\mathcal{W} - \{\mathcal{L}\}$, then we recover the result of section 3, i.e. $\mathcal{F}'_{\rm app}[\rho] = \mathcal{F}_{\mathcal{L}'}[\rho]$ (in other words, the approximate functional of the effective system coincides with the exact one). In contrast, if $\mathcal{L}'$ is not contained in any cluster of $\mathcal{W} - \{\mathcal{L}\}$, then

$$\mathcal{F}'_{\rm app}[\rho] = \sum_{\mathcal{C}'\in\mathcal{W}'-\{\mathcal{L}'\}} [-\mu_{\mathcal{W}'}(\mathcal{C}',\mathcal{L}')]\mathcal{F}_{\mathcal{C}'}[\rho]. \tag{7.3}$$

Note that if $\mathcal{W}_{\rm max}$ denotes the set of maximal clusters of $\mathcal{W} - \{\mathcal{L}\}$, then the set of maximal clusters of $\mathcal{W}' - \{\mathcal{L}'\}$ is the set $\mathcal{W}'_{\rm max}$ of all non-empty intersections of the clusters of $\mathcal{W}_{\rm max}$ with $\mathcal{L}'$. Consequently, the set $\mathcal{W}'$ coincides with the set of all non-empty intersections of the clusters in $\mathcal{W}'_{\rm max}$, as well as the cluster $\mathcal{L}'$. In other words, the approximate functional (7.3) is the one that would have been obtained directly from theorem 2 if it had been applied to the effective system (the original system constrained to $\mathcal{L}'$) choosing $\mathcal{W}'_{\rm max}$ as the set of maximal clusters.

The previous result shows that LFMT is closed under dimensional reduction. Note that if we have the approximate free-energy functional prescribed by theorem 2 for a given system and a certain $\mathcal{W}_{\rm max}$, then for all systems that can be obtained from it through dimensional reduction, the resulting approximate functional for the lower dimensional system coincides with the one deduced from theorem 2 with $\mathcal{W}'_{\rm max}$ obtained from $\mathcal{W}_{\rm max}$ as above. In LFMT, the prescription for $\mathcal{W}_{\rm max}$ for a given system is to choose all maximal 0D cavities. In order for LFMT to be closed under dimensional reduction, $\mathcal{W}'_{\rm max}$ should coincide with the set of maximal 0D cavities of the lower dimensional system. A little reflection will convince the reader that this is indeed the case (and it is so as well for other kind of mappings, like the one described in figure 2). In general, if a system s is transformed by the action of a mapping $\mathcal{T}$ into another system $\mathcal{T}\{S\}$, and if its set of maximal clusters $\mathcal{W}_{\rm max}(S)$ is transformed as described above into the set $\mathcal{W}'_{\rm max} = \mathcal{T}\{\mathcal{W}_{\rm max}(S)\}$, then the approximate functional (3.18) behaves consistently under $\mathcal{T}$ provided the prescription to choose $\mathcal{W}_{\rm max}(S)$ is such that $\mathcal{W}_{\rm max}(\mathcal{T}\{S\}) = \mathcal{T}\{\mathcal{W}_{\rm max}(S)\}$.

8. Conclusions

In this work, we have provided a rigorous foundation of LFMT based on the formalism of Möbius inversion in posets of lattice clusters. The free-energy density functional is thus expressed in the form of a cluster expansion: given a set of basic clusters $\mathcal{W}_{\max}$, there is a unique functional of the form (3.18) which is exact for every density profile with support any subcluster of $\mathcal{W}_{\max}$. The cluster expansion (3.18) requires the exact expressions of the free-energy functional on the clusters of $\mathcal{W}_{\max}$. The low-density limit of the functional (3.18) dictates a definition of the clusters of $\mathcal{W}_{\max}$—the 0D cavities, or those clusters such that if there are two or more particles in them they necessarily interact—which guarantees the exact zero-density limit of the pair and triplet direct correlation functions. This redefinition subsumes the previous version of LFMT, valid for hard-core models (Lafuente and Cuesta 2004), and extends it to include any lattice model with short-range interaction (the Ising lattice gas is an explicit example). The Möbius function formalism also allows us to analyse the behaviour of functional (3.18) under mappings between models, and a consequence of this analysis is the proof that LFMT behaves consistently under dimensional reduction (or confinements of the density into lower dimensional sets of the lattice).

Going beyond LFMT—to account for higher order correlations, for instance—is, in principle, possible, but requires choices of $\mathcal{W}_{\max}$ containing larger clusters. But then the exact free-energy functional of these larger clusters is required, what may be too involved. In some cases, though, e.g., in some one-dimensional models, it is known that LFMT as such is exact, so no improvement is required in those cases. The reason behind this fact is not clear to us yet, and it is certainly a matter that deserves further thought.

Another interesting point concerns the continuum limit. For some models (e.g., hard cubes in a d-dimensional cubic lattice), this limit is feasible and in fact the limit functional coincides with the FM functional of the continuum model (Lafuente 2004). But there are important cases, like hard spheres, which are not easy to obtain as a limit of discrete lattice models. If this were possible, the result would be a functional which would recover the exact zero-density limit of the pair and triplet direct correlation functions, something that the best current FM functional for hard spheres does not accomplish (Tarazona and Rosenfeld 1997, Tarazona 2000, Cuesta *et al* 2002). From the insight provided by the present analysis of LFMT we dare to say that Tarazona's functional for hard spheres (Tarazona 2000) is the limit of the functionals (3.18) of a sequence of lattice models with an incomplete choice of $\mathcal{W}_{\max}$ (incomplete in the sense that some maximal 0D cavities are not in $\mathcal{W}_{\max}$). This is another problem certainly worth exploring.

Acknowledgment

This work is supported by project BFM2003-0180 of the Spanish Ministerio de Ciencia y Tecnología.

Appendix. Some technical results

In order to make this work self-contained, in this appendix we will collect and prove some technical results of the theory of posets (and in particular of cluster posets) that we have used along this paper. We will only prove those which are original contributions, while for the rest we simply address the reader to the specialized literature, where complete proofs can be found.

In section 3, we have proved that the approximate functional (3.18) is exact when it is restricted to any cluster of the set $\mathcal{W} - \{\mathcal{L}\}$. Also, in sections 6 and 7 we have analysed the low-density limit and the dimensional crossover of the functional (3.18), respectively. In all cases, the key point was to evaluate a constrained sum of the Möbius function $\mu_{\mathcal{W}}(\mathcal{C}, \mathcal{L})$ for those $\mathcal{C} \in \mathcal{W}$ satisfying $\mathcal{C} \cap \mathcal{D} = \mathcal{E}$, $\mathcal{D}$ and $\mathcal{E}$ being clusters of the lattice $\mathcal{L}$ not necessarily in $\mathcal{W}$. This relation can be expressed in the more general form $\sigma^+\mathcal{C} = \mathcal{E}$, where σ^+ is a mapping from the cluster poset $\mathcal{W}$ to another cluster poset which, in general, is different from $\mathcal{W}$. For all instances in this work, σ^+ is just the intersection with a given fixed cluster of the underlying lattice, and hence is an order-preserving map.

The advantage of the above interpretation of the constraint over the clusters in $\mathcal{W}$ is that it allows us to use an important result of the theory of posets which relates the Möbius functions of two posets, $\mathcal{W}$ and $\mathcal{V}$, if both are connected through a pair (σ, σ^+) of order-preserving maps, where σ is a *Galois function*, defined as

Definition A.1. *Let $\mathcal{V}$ and $\mathcal{W}$ be posets. A mapping $\sigma: \mathcal{V} \to \mathcal{W}$ is called a Galois function if there exists a function $\sigma^+: \mathcal{W} \to \mathcal{V}$ such that*

(i) σ, σ^+ are order-preserving maps;
(ii) $\sigma^+\sigma x \geqslant x$ for all $x \in \mathcal{V}$ and $\sigma\sigma^+ z \leqslant z$ for all $z \in \mathcal{W}$.

Given two posets $\mathcal{V}$ and $\mathcal{W}$, their Möbius functions are related via a Galois function in the precise way expressed in the following theorem (which is a version of theorem 4.39 in p 173 of Aigner (1979)):

Theorem A.1. *Let $\sigma: \mathcal{V} \to \mathcal{W}$ be a Galois function. Then, for all $x \in \mathcal{V}$, $y \in \mathcal{W}$,*

$$\sum_{z \in \mathcal{W}, \sigma^+ z = x} \mu_{\mathcal{W}}(z, y) = \begin{cases} \mu_{\sigma^+\mathcal{W}}(x, \sigma^+ y) & \text{if } x \in \sigma^+\mathcal{W} \text{ and } y \in \sigma\mathcal{V}, \\ 0 & \text{otherwise.} \end{cases}$$

This theorem is exactly what we need to derive a result which can be applied directly to evaluate the constrained sums that appear along this paper:

Corollary A.1. *Let $\mathcal{W}$ be a cluster poset with underlying lattice $\mathcal{L}$ which contains all non-empty intersections of certain cluster poset $\mathcal{W}_{\max}$ as well as the cluster $\mathcal{L}$. Let $\mathcal{L}'$ be a cluster of lattice $\mathcal{L}$ not necessarily in $\mathcal{W}$, and let us consider the poset $\mathcal{W}'$ of all non-empty intersections of the clusters in $\mathcal{W}$ with $\mathcal{L}'$. Then, for all $\mathcal{C}' \in \mathcal{W}'$,*

$$\sum_{\mathcal{C} \in \mathcal{W}, \mathcal{C} \cap \mathcal{L}' = \mathcal{C}'} \mu_{\mathcal{W}}(\mathcal{C}, \mathcal{L}) = \begin{cases} \mu_{\mathcal{W}'}(\mathcal{C}', \mathcal{L}') & \text{if } \mathcal{L}' \not\subset \mathcal{D} \text{ for all } \mathcal{D} \in \mathcal{W} - \{\mathcal{L}\}, \\ 0 & \text{otherwise.} \end{cases}$$

Proof. To apply theorem 3, we first need to rewrite the constrained sum in the statement of the corollary in a more convenient form. Since $\varnothing \notin \mathcal{W}'$, the only clusters in $\mathcal{W}$ contributing to the sum are those whose intersection with $\mathcal{L}'$ is non-empty. Let $\widetilde{\mathcal{W}}$ denote the poset of such clusters; then we have that $\mu_{\mathcal{W}}(\mathcal{C}, \mathcal{L}) = \mu_{\widetilde{\mathcal{W}}}(\mathcal{C}, \mathcal{L})$ for all $\mathcal{C} \in \widetilde{\mathcal{W}}$, since $[\mathcal{C}, \mathcal{L}]$ in $\mathcal{W}$ is identical to $[\mathcal{C}, \mathcal{L}]$ in $\widetilde{\mathcal{W}}$. Therefore, we have the identity

$$\sum_{\mathcal{C} \in \mathcal{W}, \mathcal{C} \cap \mathcal{L}' = \mathcal{C}'} \mu_{\mathcal{W}}(\mathcal{C}, \mathcal{L}) = \sum_{\mathcal{C} \in \widetilde{\mathcal{W}}, \sigma^+ \mathcal{C} = \mathcal{C}'} \mu_{\widetilde{\mathcal{W}}}(\mathcal{C}, \mathcal{L}), \tag{A.1}$$

where $\sigma^+: \widetilde{\mathcal{W}} \to \mathcal{W}'$ is defined as $\sigma^+\mathcal{C} \equiv \mathcal{C} \cap \mathcal{L}'$.

Now, in order to apply theorem A.1 to (A.1) we need a pair (σ, σ^+) of order-preserving maps $\sigma: \mathcal{W}' \to \widetilde{\mathcal{W}}$ and $\sigma^+: \widetilde{\mathcal{W}} \to \mathcal{W}'$, σ being a Galois function. Let us take for σ^+ the one

introduced in (A.1) and let us define σ as $\sigma\mathcal{C}' \equiv \inf\{\mathcal{C} \in \widetilde{\mathcal{W}} | \mathcal{C}' \subset \mathcal{C}\}$ for all $\mathcal{C}' \in \mathcal{W}'$. It is a direct consequence of the definition that σ and σ^+ both are order-preserving maps. Moreover, for all $\mathcal{C} \in \widetilde{\mathcal{W}}$ we have

$$\sigma\sigma^+\mathcal{C} = \inf\{\mathcal{D} \in \widetilde{\mathcal{W}} | \sigma^+\mathcal{C} \subset \mathcal{D}\} \leqslant \mathcal{C}$$

because $\sigma^+\mathcal{C} \subset \mathcal{C}$, and for all $\mathcal{C}' \in \mathcal{W}'$,

$$\sigma^+\sigma\mathcal{C}' = \sigma^+ \inf\{\mathcal{D} \in \widetilde{\mathcal{W}} | \mathcal{C}' \subset \mathcal{D}\} = \inf\{\mathcal{D}' \in \mathcal{W}' | \mathcal{C}' \subset \mathcal{D}'\} = \mathcal{C}'.$$

Therefore, σ and σ^+ satisfy condition (ii) of definition A.1 and thus σ is a Galois function.

At this point, we can apply theorem A.1 to the rhs of (A.1). Since for this case $\sigma^+\widetilde{\mathcal{W}} = \mathcal{W}'$, for all $\mathcal{C}' \in \mathcal{W}'$ we can write

$$\sum_{\mathcal{C}\in\mathcal{W}, \mathcal{C}\cap\mathcal{L}=\mathcal{C}'} \mu_{\mathcal{W}}(\mathcal{C}, \mathcal{L}) = \begin{cases} \mu_{\mathcal{W}'}(\mathcal{C}', \mathcal{L}') & \text{if } \mathcal{L} \in \sigma\mathcal{W}', \\ 0 & \text{otherwise.} \end{cases}$$

The last step of the proof just amounts to showing that $\mathcal{L} \in \sigma\mathcal{W}'$ if and only if $\mathcal{L}'$ is not contained in any cluster of $\mathcal{W} - \{\mathcal{L}\}$. Note that the latter is equivalent to $\sigma\mathcal{L}' = \mathcal{L}$, therefore if it holds then $\mathcal{L} \in \sigma\mathcal{W}'$. Now, let us assume that $\mathcal{L} \in \sigma\mathcal{W}'$, then there exists a cluster $\mathcal{C}'$ in $\mathcal{W}'$ such that $\sigma\mathcal{C}' = \mathcal{L}$. Since σ is order-preserving and $\mathcal{C}' \leqslant \mathcal{L}'$, we have $\mathcal{L} = \sigma\mathcal{C}' \leqslant \sigma\mathcal{L}'$, where only the equality $\sigma\mathcal{L}' = \mathcal{L}$ can hold, and the proof is complete. □

To end this appendix, we will bring about some results which simplify the calculation of the Möbius function of certain posets, and we will provide two applications related to the cluster posets involved in this work. First of all, we will give some definitions relative to a special type of posets: *lattices* (a mathematical notion not to be confused with physical lattices), since they appear in a natural way when we have to compute the Möbius function of a locally finite poset.

A poset $\mathcal{P}$ is a *lattice* if for any $x, y \in \mathcal{P}$, $\sup\{x, y\}$ and $\inf\{x, y\}$ are in $\mathcal{P}$. One instance of a lattice is any interval $[\mathcal{C}, \mathcal{C}']$ of a cluster poset $\mathcal{W}$ such as those involved in theorem 2 (which are closed under non-empty intersections). A 0-element of a poset $\mathcal{W}$, denoted $\hat{0}$, is an element satisfying $\hat{0} \leqslant x$ for all $x \in \mathcal{W}$. Dually, a 1-element of $\mathcal{W}$, denoted $\hat{1}$, is an element satisfying $x \leqslant \hat{1}$ for all $x \in \mathcal{W}$. Obviously, any finite lattice has a $\hat{0}$ and a $\hat{1}$. A *point* of a finite lattice $\mathcal{P}$ is an element satisfying $\hat{0} < x$ for which there is no element $z \in \mathcal{P}$ such that $z < x$. A *copoint* of a finite lattice $\mathcal{P}$ is an element satisfying $x < \hat{1}$ for which there is no element $z \in \mathcal{P}$ such that $x < z$. A subset $\mathcal{M}$ of a finite lattice $\mathcal{P}$ is called a *lower cross-cut* if $\hat{0} \notin \mathcal{M}$ and for all $\hat{0} \neq x \in \mathcal{P}$ with $x \notin \mathcal{M}$ there is an element $y \in \mathcal{M}$ with $y \leqslant x$. Dually, a subset $\mathcal{M}$ is an *upper cross-cut* if $\hat{1} \notin \mathcal{M}$ and for all $\hat{1} \neq x \in \mathcal{P}$ with $x \notin \mathcal{M}$ there is an element $y \in \mathcal{M}$ with $x \leqslant y$. An example of lower (upper) cross-cut is the set of all points (copoints) of a given finite lattice.

In our particular case, we have to compute the Möbius function $\mu_{\mathcal{W}}(\mathcal{C}, \mathcal{L})$, where $\mathcal{W}$ is a cluster poset which contains all non-empty intersections of the elements of certain cluster set $\mathcal{W}_{\max}$ as well as the cluster $\mathcal{L}$, which is a 1-element. Let us consider the finite lattice $\mathcal{P}_{\mathcal{C}} \equiv [\mathcal{C}, \mathcal{L}]$ with $\hat{0} = \mathcal{C}$ and $\hat{1} = \mathcal{L}$. From recursion 1, it is straightforward that for all $\mathcal{C} \in \mathcal{W}$ we have $\mu_{\mathcal{W}}(\mathcal{C}, \mathcal{L}) = \mu_{\mathcal{P}_{\mathcal{C}}}(\hat{0}, \hat{1})$. Therefore, in all cases we have to compute the Möbius function $\mu_{\mathcal{P}}(\hat{0}, \hat{1})$ of certain finite lattice $\mathcal{P}$. This fact makes the following theorem (from Aigner (1979), theorem 4.42 on p 175) very useful:

Theorem A.2 (cross-cut theorem). *Let $\mathcal{P}$ be a finite lattice, $\mathcal{M}$ a lower (upper) cross-cut and n_k the number of sets $\mathcal{A} \subset \mathcal{M}$ with k elements such that* $\sup \mathcal{A} = \hat{1}$ ($\inf \mathcal{A} = \hat{0}$). *Then,*

$$\mu_{\mathcal{P}}(\hat{0}, \hat{1}) = \sum_{k\geqslant 0} (-1)^k n_k.$$

When we take in this theorem the lower (upper) cross-cut as the set of all points (copoints) of the lattice $\mathcal{P}$, a direct consequence is the following corollary (from Stanley (1999), corollary 3.9.5 on p 126):

Corollary A.2. *Let $\mathcal{P}$ be a finite lattice with point set $\mathcal{Q}$ and copoint set $\mathcal{R}$. Then,*

$$\mu_{\mathcal{P}}(\hat{0}, \hat{1}) = 0$$

if $\hat{0} \neq \inf \mathcal{R}$ or $\hat{1} \neq \sup \mathcal{Q}$.

Note that this result renders the calculation of some values of the Möbius function of a given poset straightforward.

A practical application of corollary A.2 is to easily compute some values of the Möbius function, say, in the working example of section 4. Note that it is a direct consequence of this corollary that $\mu_{\mathcal{W}}(\overset{s}{\circ}\!\!-\!\!\circ, \mathcal{L}) = \mu_{\mathcal{W}}(\overset{s}{\circ}\!\!-\!\!\bullet, \mathcal{L}) = \mu_{\mathcal{W}}(\overset{s}{\bullet}\!\!-\!\!\circ, \mathcal{L}) = \mu_{\mathcal{W}}(\bullet_s, \mathcal{L}) = \mu_{\mathcal{W}}(\circ_s, \mathcal{L}) = 0$ for any $s \in \mathcal{L}$, since for all these clusters the supremum of the sets of points of the corresponding interval is different from $\mathcal{L}$ (a glance at figure 1 is enough to realize it).

An important consequence of corollary A.2 concerns the form of the cluster expansion of the free-energy functional. Note that in our formulation of LFMT we have worked with $\mathcal{W}$ defined as the cluster $\mathcal{L}$ and the non-empty intersections of the clusters in $\mathcal{W}_{\max}$. We have shown that this choice is enough to ensure our main purpose, building an approximate free-energy functional which is exact in the clusters of $\mathcal{W}_{\max}$. Having this idea in mind, we could have started with the cluster poset $\mathcal{V}$ made of cluster $\mathcal{L}$ and all non-empty clusters contained in some cluster of $\mathcal{W}_{\max}$ (note that $\mathcal{W} \subset \mathcal{V}$). Now, if we compute $\mu_{\mathcal{V}}(\mathcal{C}, \mathcal{L})$ for all $\mathcal{C} \in \mathcal{V}$, then the application of corollary A.2 implies that if $\mathcal{C}$ is in $\mathcal{V}$ but not in $\mathcal{W}$ then $\mu_{\mathcal{V}}(\mathcal{C}, \mathcal{L}) = 0$, while $\mu_{\mathcal{V}}(\mathcal{C}, \mathcal{L}) = \mu_{\mathcal{W}}(\mathcal{C}, \mathcal{L})$ otherwise. In other words, if a cluster $\mathcal{C}$ is not the intersection of maximal clusters, then it does not contribute to the cluster expansion of the free-energy functional. So, we can constrain this cluster expansion to $\mathcal{W}$ without loss of generality, as we have indeed done.

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