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## Distribution of physical clusters

A Coniglio†§, U De Angelis‡, A Forlani‡ and G Lauro†

†Istituto di Fisica Teorica, Università di Napoli, Mostra d'Oltremare, Pad. 19, 80125 Napoli, Italy

‡Istituto di Fisica Sperimentale, Università di Napoli, Via A Tari 3, Napoli, Italy

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**Abstract.** A general formalism is developed to obtain series expansions of the average number of physical clusters of particles in the framework of Mayer's theory.

The special case of lattice systems is investigated in more detail and some preliminary results are given on the relation between percolation (namely the formation of an infinite cluster) and condensation in fluid systems in the lowest approximation (summation of chain diagrams).

### 1. Introduction

The concept of physical clusters has been widely applied in recent years. The distribution of physical clusters for a lattice system is directly connected to the percolation problem (for a review see Shante and Kirkpatrick 1971, Essam 1973) and has recently been investigated using the renormalization group technique (Harris *et al* 1975). This distribution is also of interest in the problem of the dilute ferromagnet (Elliot and Heap 1962, Rapaport 1972) and in a system of random resistors (Kirkpatrick 1973, and references cited therein).

A knowledge of the cluster distribution might also be of some help in the nucleation problem which has been studied by Binder (1975). The shape of the clusters in a lattice gas has been investigated by Domb (1976) and by Domb *et al* (1975) using Monte Carlo techniques. A connection between physical clusters and the phase transition in the Ising model has been found by Coniglio (1976) and Coniglio *et al* (1976, 1977). The location of the percolation point in the three-dimensional Ising model has also been investigated by Muller-Krumbhaar (1974) using Monte Carlo techniques.

Our aim in this paper is to find a general formulation of the distribution of physical clusters following Hill's initial work (Hill 1955). In § 2 we recall briefly the basic concepts of Hill's work and then proceed to find the general series expansions for the average number of physical clusters and a rule to build the successive coefficients in the activity expansion. In § 3 some of these coefficients are evaluated for a lattice system in terms of the lattice constants. In § 4 the general formalism is used to gain a first insight into the problem of condensation of fluid systems and the corresponding behaviour of physical clusters.

§ CNR, Gruppo Nazionale di Struttura della Materia.

**2. The general formalism**

In Hill’s work physical clusters are defined in phase space for systems which can be described in terms of pair potentials  $u(r_{ij})$ :  $n$  particles belong to the same cluster if they are pairwise bound in phase space (negative total energy) . This division of phase space leads to the introduction of two effective potentials:

$$u^+(r) = \begin{cases} +\infty & u(r) > 0 \\ u(r) - kT \ln F(r) & u(r) \leq 0 \end{cases}$$

$$u^*(r) = \begin{cases} u(r) & u(r) > 0 \\ u(r) - kT \ln(1 - F(r)) & u(r) \leq 0 \end{cases}$$

where  $F(r)$  is given by the incomplete  $\Gamma$  function:

$$F(r) = \Gamma(\frac{3}{2}, -\beta u(r)) / \Gamma(\frac{3}{2})$$

and

$$e^{-\beta u_{ij}(r)} = e^{-\beta u_{ij}^+(r)} + e^{-\beta u_{ij}^*(r)} \tag{1}$$

where  $\beta = 1/kT$ . These  $u^+$  and  $u^*$  are the effective potentials between bound and unbound pairs respectively and substitution of equation (1) into each of the  $\exp(-\beta u_{ij})$  appearing in the partition function  $Q_N$  of  $N$  particles gives an immediate recipe to obtain the partition functions  $Q_{N_1 N_2 \dots N_N}$  of specified sets of physical clusters ( $N_1$  clusters of 1 particle,  $N_2$  clusters of 2 particles . . .) such that

$$Q_N = \sum_{\{N_1 \dots N_N\}} Q_{N_1 \dots N_N} \tag{2}$$

where the sum is over all partitions of  $N$  with the condition

$$\sum_{s=1}^N s N_s = N. \tag{3}$$

This is most easily seen diagrammatically: if we associate a full circle with each particle, a full line with each  $\exp(-\beta u^+)$  bond and a broken line with each  $\exp(-\beta u^*)$  bond then for  $N = 3$  for instance we have:

$$Q_3 = Q_{300} + Q_{110} + Q_{001}$$

and the corresponding graphs are shown in figure 1 where the first diagram represents three clusters of one particle ( $Q_{300}$ ), the second one represents one cluster of two particles and one cluster of one particle ( $Q_{110}$ ) and the last two diagrams give the cluster of three particles ( $Q_{001}$ ). From the diagrams it is easy to write out the terms explicitly with a field integration for each point and an overall factor  $(N! \Lambda^N)^{-1}$  where  $\Lambda$  is the



**Figure 1.** Clusters of three particles. The coefficients take care of permutations.

thermal wavelength. For instance

$$Q_{110} = 3 \left( \frac{1}{3! \Lambda^3} \right) \iiint e^{-\beta u_{12}^+} e^{-\beta u_{13}^+} e^{-\beta u_{23}^*} d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}_3.$$

These partition functions are used to write down the grand partition function of the system in a multicomponent formalism:

$$\Xi = \sum_{N \geq 0} \sum_{\{N_1, \dots, N_N\}} \lambda_1^{N_1} \dots \lambda_N^{N_N} Q_{N_1, \dots, N_N} \tag{4}$$

where  $\lambda_s$  is the absolute activity of the  $s$ -species (clusters of  $s$  particles) and at equilibrium  $\lambda_s = \lambda^s$  where  $\lambda$  is the absolute activity of the system. The multicomponent formalism allows one to calculate the average number of  $s$  clusters:

$$\langle N_s \rangle = \lambda_s \frac{\partial}{\partial \lambda_s} \ln \Xi |_{\lambda_s = \lambda^s} = \sum_{N \geq s} \lambda^N A_N. \tag{5}$$

Whereas in Hill's work only the first few terms in (5) are calculated explicitly we wish here to give an expression for the general term  $A_N$  in the activity expansion. In order to do this we have to calculate the coefficients in the logarithm of the multicomponent grand partition function which can be written in the general form:

$$\ln \Xi = \sum_{N \geq 1} \sum_{\{N_1, \dots, N_N\}} \lambda_1^{N_1} \dots \lambda_N^{N_N} b'_{N_1, \dots, N_N}. \tag{6}$$

Now  $b'_{N_1, \dots, N_N}$  can be obtained from the known  $Q_{N_1, \dots, N_N}$  using the same general theorem on graphs which allows one to calculate the connected integrals  $b_N$  from  $Q_N$  in Mayer's theory for one variable (see Domb 1974a, theorem 1). To make this theorem applicable to our case we define the functions:

$$f_{ij}^+ = e^{-\beta u_{ij}^+}; \quad f_{ij}^* = e^{-\beta u_{ij}^*} - 1 \tag{7}$$

and then replace each  $\exp(-\beta u^*)$  bond in the  $Q_{N_1, \dots, N_N}$  diagrams by  $(1 + f^*)$  and associate a wavy line with the  $f^*$  bond keeping the full lines to represent  $\exp(-\beta u^+) = f^+$  bonds; a new set of diagrams develops which, unlike the previous ones which are all fully connected, contains connected and unconnected graphs (connected graphs are those graphs where there is at least one path, formed of lines and/or wavy lines, between any two points). For these new sets of graphs all the conditions of theorem 1 are satisfied: thus the  $b'_{N_1, \dots, N_N}$  are the connected parts of the  $Q_{N_1, \dots, N_N}$ . An example for  $N = 3$  is given in figure 2 where the decomposition of the  $Q$  in terms of  $f$  bonds is shown; the  $b$  for this case are the connected graphs of figure 2 and are shown in figure 3.

The explicit integrals for the  $b$  now have to be written in terms of  $f$  functions, e.g.:

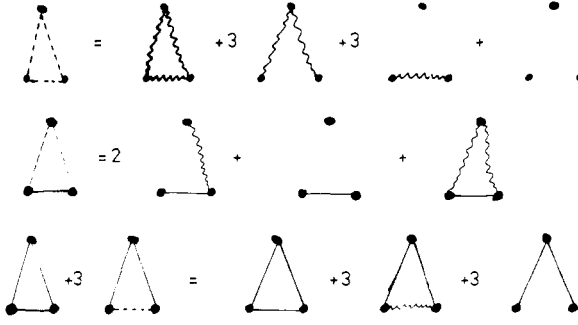
$$b'_{110} = \frac{1}{3! \Lambda^3} \left( 6 \iiint f_{12}^+ f_{23}^* d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}_3 + 3 \iiint f_{12}^+ f_{23}^* f_{13}^* d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}_3 \right).$$

Then from equations (5) and (6) we get:

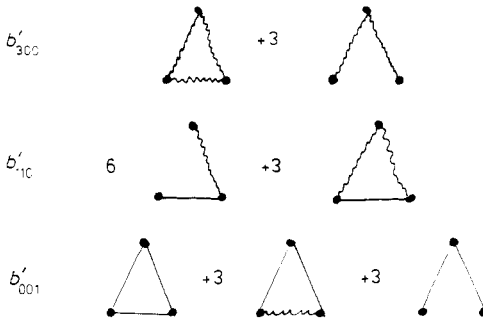
$$\langle N_s \rangle = \sum_{N \geq s} \lambda^N \sum_{\{N\}} N_s b'_{\{N\}}$$

where the sum is over all partitions  $\{N\} = \{N_1 \dots N_N\}$  satisfying condition (3). Introducing a new set of coefficients

$$b_{\{N\}} = \frac{\Lambda^N}{V} b'_{\{N\}}$$



**Figure 2.** Decomposition of clusters of three particles in terms of  $f^+$  and  $f^*$  bonds defined by equation (7) in the text.



**Figure 3.** Connected graphs of figure (2) corresponding to the coefficients  $b_{N_1 N_2 N_3}$ .

we can write the expansion in terms of the activity  $z = \lambda/\Lambda$  as:

$$\langle N_s \rangle = V \sum_{N \geq s} z^N \sum_{\{N\}} N_s b_{\{N\}} \tag{8}$$

Equation (8) gives the general activity expansion for the equilibrium distribution of physical clusters in the system with coefficients defined in terms of partial partition functions  $Q_{N_1 \dots N_N}$ .

We wish to point out however that there is a more direct way of obtaining the coefficients  $b_{N_1 \dots N_N}$  starting with Mayer's connected integrals

$$b_N = \frac{1}{N! V} \sum_{\{CN\}} \int \dots \int d\mathbf{r}_1 \dots d\mathbf{r}_N (\prod f_{ij}) \tag{9}$$

where the sum is over all connected graphs of  $N$  particles and the  $f_{ij}$  are Mayer's functions:

$$f_{ij} = \exp(-\beta u_{ij}) - 1 = f_{ij}^+ + f_{ij}^* \tag{10}$$

It is easy to show that if each  $f$  bond in the graphs for  $b_N$  is changed according to (10) then the resulting graphs can be collected to give back the  $b_{\{N\}}$  and

$$b_N = \sum_{\{N\}} b_{\{N\}}, \quad \sum_{s=1}^N s N_s = N. \tag{11}$$

Equation (11) gives an immediate recipe to obtain the coefficients in the activity

expansion (8). We write out explicitly the first few terms for the partial densities  $\rho_s = \langle N_s \rangle / V$ :

$$\begin{aligned} \rho_1 &= b_{1z} + 2b_{20}z^2 + (3b_{300} + b_{110})z^3 + \dots \\ \rho_2 &= b_{01}z^2 + b_{110}z^3 + (2b_{0200} + b_{2100})z^4 + \dots \\ \rho_3 &= b_{001}z^3 + b_{1010}z^4 + (b_{20100} + b_{01100})z^5 + \dots \end{aligned} \tag{12}$$

and it is easy to check that these coincide with Hill's results.

### 3. Lattice gas

The formalism of § 2 has a straightforward application to a lattice system where the coordinates of the particles can only assume discrete values and integrations are replaced by summations over lattice sites:

The interaction is

$$u_{ij} = \begin{cases} +\infty & \text{if } i = j \\ -\epsilon & \text{if } i \text{ and } j \text{ are nearest neighbours} \\ 0 & \text{otherwise.} \end{cases}$$

Let us define the function

$$\theta_{ij} = \begin{cases} +1 & \text{if } i \text{ and } j \text{ are nearest neighbours} \\ 0 & \text{otherwise.} \end{cases}$$

Then the effective potentials introduced in § 2 are:

$$\begin{aligned} \exp(-\beta u_{ij}^+) &= \theta_{ij} \exp(-\beta u_{ij}) = \theta_{ij} \exp(\beta \epsilon) \\ \exp(-\beta u_{ij}^*) &= (1 - \theta_{ij}) \exp(-\beta u_{ij}) = 1 - \theta_{ij} - \delta_{ij} \end{aligned} \tag{13}$$

and there is a major simplification in this case as the coefficients in the activity expansion can be expressed in terms of weak lattice constants (for the terminology see Domb 1974b) which are tabulated. For instance for  $N = 2$  only the weak lattice constant  $q/2$  of the two-point graph is needed and we have:

$$b_{20} = -\frac{1}{2} - \frac{1}{2}q; \quad b_{01} = \frac{1}{2}q e^{\beta \epsilon}.$$

Thus

$$b_2 = b_{01} + b_{20} = -\frac{1}{2} + \frac{1}{2}qf \quad (f = e^{\beta \epsilon} - 1)$$

which is in agreement with Domb (1974b). For  $N = 3$  both  $q/2$  and  $p_3$  are needed, where  $p_3$  is the weak lattice constant of the three-point graph (triangle) and we have:

$$\begin{aligned} b_{001} &= p_3(e^{3\beta \epsilon} - 3e^{2\beta \epsilon}) - e^{2\beta \epsilon}(\frac{1}{2}q^2 - \frac{1}{2}q) \\ b_{110} &= (3p_3 - q^2)e^{\beta \epsilon} \\ b_{300} &= \frac{1}{3} + \frac{1}{2}q(q - 1) - p_3 \end{aligned}$$

which gives

$$b_3 = \frac{1}{3} - qf + \frac{1}{2}q(q - 1)f^2 + p_3f^3$$

in agreement with Domb (1974b).

The recipe to obtain the coefficients explicitly is to use equation (13) for the bonds in the graphs for the  $b$ : then the summation over lattice sites for the resulting configurations gives the appropriate weak lattice constants.

#### 4. Percolation and condensation

##### 4.1. Preliminary considerations

In this section we consider the possible connection between condensation and percolation, which has been investigated, as far as we know, only for lattice systems. For a system of interacting spins Coniglio (1976) has shown on the Bethe lattice that the percolation point, i.e. the point where an infinite cluster begins to appear in the system, is located on the coexistence curve before the critical point. For a ferromagnetic Ising model it has been proved by Coniglio *et al* (1976, 1977) that spontaneous magnetization can only exist upon formation of an infinite cluster of spins. It is also proved that in the two-dimensional model the critical point is also a percolation point, whereas in the three-dimensional model there is some indication that the percolation point should appear before the critical point is reached (Muller-Krumbhaar 1974, Sykes and Gaunt 1976).

We have started an investigation of the problem of condensation and percolation in the framework of the general formalism developed in the present work which is valid for lattice as well as fluid systems. The idea is to use the formalism to evaluate, in successive approximations, i.e. summation of diagrams, the percolation quantities related to condensation as well as explicit contributions of the physical clusters to the thermodynamics of the system. The results of the lowest-order approximation are presented here mainly to show the possible use of the general formalism.

The idea that condensation of a fluid system is somehow related to the formation of large 'clusters' in the system is the central point in the theories of Frenkel, Band and Mayer (see Hill 1956).

Here we wish to show, in the lowest approximation, how the condensation point (in the sense of Mayer's theory) is also a percolation point (formation of a large 'physical' cluster in the system).

The relevant quantities in percolation theory are:

$$\bar{n} = \sum_{s=1}^{\infty} \rho_s; \quad P = 1 - \frac{\sum_{s=1}^{\infty} s \rho_s}{\rho}; \quad S = \frac{\sum_{s=1}^{\infty} s^2 \rho_s}{\sum_{s=1}^{\infty} s \rho_s} \quad (14)$$

where  $\bar{n}$  is the mean number density of clusters,  $P$  is the percolation probability (i.e. the probability that a given particle belongs to an infinite cluster) and  $S$  is the mean cluster size (for finite clusters).

These quantities near the percolation point behave, respectively, like the free energy, the spontaneous magnetization and the susceptibility of a ferromagnet near the critical point (Essam 1973): in fact at the percolation point  $\bar{n}$  shows a weak singularity,  $P$  starts increasing from its zero value and  $S$  diverges.

Since we approach the percolation critical density from lower densities (gaseous system) we have

$$\rho = \sum_{s=1}^{\infty} s \rho_s \quad (15)$$

and  $P$  is always identically zero. We are thus left with  $\bar{n}$  and  $S$ .

#### 4.2. Gas of ideal clusters in the chain approximation

From equations (8) and (11) the activity expansions for the pressure and  $\bar{n}$  can be written as:

$$\beta P = \sum_N z^N \sum_{\{N\}} b_{\{N\}}; \quad \bar{n} = \sum_N z^N \sum_{\{N\}} \left( \sum_s N_s \right) b_{\{N\}}. \quad (16)$$

First we sum over all chain diagrams (no nodal points and no closed loops) with only  $f^+$  bonds: clearly in this case each diagram of  $S$  points is a cluster of  $S$  particles, i.e.,  $\sum_s N_s = 1$  and  $\sum_{\{N\}} b_{\{N\}}^{(0)} = b_{00\dots 1}^{(0)}$ . So in this approximation

$$\beta P^{(0)} = \bar{n}^{(0)} \quad (17)$$

where the superscript means that no  $f^*$  bond has been considered (pressure of an ideal gas of clusters). Now in the chain approximation:

$$b_{00\dots 1}^{(0)} = \frac{1}{2V} \int \dots \int d\mathbf{r}_1 \dots d\mathbf{r}_N f^+(1, 2) f^+(2, 3) \dots f^+(N-1, N) = \frac{1}{2} (b^+)^{N-1} \quad (18)$$

where 2 is the symmetry factor of the chain diagrams and:

$$b^+ = \int f^+(r) d\mathbf{r}. \quad (19)$$

Then

$$\beta P^{(0)} = \bar{n}^{(0)} = z + \frac{1}{2} \sum_{N \geq 2} z^N (b^+)^{N-1} = \frac{z}{2} \frac{2 - zb^+}{1 - zb^+}. \quad (20)$$

The density in the same approximation is:

$$\rho^{(0)} = \sum_s s \rho_s^{(0)} = z \frac{\partial}{\partial z} \beta P^{(0)} = \frac{z}{2} \frac{2 - zb^+}{1 - zb^+} + \frac{z^2}{2} \frac{b^+}{(1 - zb^+)^2} \quad (21)$$

and the mean size of the physical clusters is given by  $\rho^{(0)} S^{(0)} = z \partial \rho^{(0)} / \partial z$ .

When interpreted in the framework of Mayer's theory these results show that the condensation point is given by  $z_{\text{crit}} = 1/b^+ > 0$  which is also the point where  $S^{(0)}$  diverges thus showing coincidence of condensation and formation of an infinite physical cluster.

#### 4.3. Gas of interacting clusters in the chain approximation

Insertion of a single  $f^*$  bond in the chains will now produce the lowest-order contribution of the interaction between clusters.

It is easy to see that the effect of an  $f^*$  bond in any chain is to break the single cluster into two separate 'interacting' clusters so that now  $\sum_s N_s = 2$  and then (the superscript 1 refers to the presence of a single  $f^*$ ):

$$\bar{n}^{(1)} = \bar{n}^{(0)} + \sum_{N \geq 2} z^N (N-1) b^* (b^+)^{N-2}; \quad b^* = \int f^*(r) d\mathbf{r} \quad (22)$$

where  $(N-1)$  accounts for the ways of inserting  $f^*$  in an  $N$ -particle chain. Summation of equation (22) gives:

$$\bar{n}^{(1)} = \bar{n}^{(0)} + \frac{z^2 b^*}{(1 - zb^+)^2}. \quad (23)$$



The pressure in the same approximation is:

$$\beta P^{(1)} = \beta P^{(0)} + \frac{1}{2} \frac{z^2 b^*}{(1 - zb^*)^2}. \quad (24)$$

Thus

$$\beta P^{(1)} = \bar{n}^{(1)} - \frac{1}{2} \frac{z^2 b^*}{(1 - zb^*)^2} \quad (25)$$

and the second term now gives the contribution of the interaction to the pressure to first order in  $b^*$ . The density in this approximation is then given by  $z \partial \beta P^{(1)} / \partial z$ . The mean cluster size is now:

$$S^{(1)} = S^{(0)} + \sum_{N \geq 2} z^N \sum_{\{N\}} \left( \sum_s s^2 N_s \right) b_{\{N\}}^{(1)}. \quad (26)$$

It is easy to show that

$$\sum_s s^2 N_s = \sum_{s=1}^{N-1} [s^2 + (N-s)^2] = -\frac{1}{3} N^3 + N^2 + \frac{1}{3} N$$

and then

$$S^{(1)} = S^{(0)} + \frac{1}{3} z \frac{\partial}{\partial z} Q(z) + \left( z \frac{\partial}{\partial z} \right)^2 Q(z) - \frac{1}{3} \left( z \frac{\partial}{\partial z} \right)^3 Q(z) \quad (27)$$

where

$$Q(z) = \frac{1}{2} \frac{z^2 b^*}{1 - zb^*}. \quad (28)$$

The conclusion is that the effect of interaction to this order does not change the critical point of percolation and condensation. Insertion of all possible  $f^*$  bonds in the chains however does change the condensation point. In this case in fact (the chain with all possible bonds):

$$\sum_{\{N\}} b_{\{N\}} = b_N = \frac{1}{2} b^{N-1} \quad (29)$$

where

$$b = \int f(r) d\mathbf{r} = -2B_2(T)$$

where  $B_2(T)$  is the second virial coefficient. The pressure and density are then:

$$\beta P = \frac{z}{2} \frac{2 - zb}{1 - zb}; \quad \rho = \frac{z}{2} \frac{2 - zb}{1 - zb} + \frac{z^2}{2} \frac{b}{(1 - zb)^2}. \quad (30)$$

The condensation point is now given by  $z_{\text{crit}} = 1/b$  for  $b > 0$ : it is interesting to note that since  $b(T)$  can also be negative this implies the existence of a critical temperature  $b(T_c) = 0$  above which there is no condensation.

What can be said in this approximation for the percolation point? It is not too difficult to calculate  $\bar{n}$ : in fact in this case

$$\sum_s N_s = \text{number of } f^* \text{ bonds} + 1$$

and this leads us to write the general term as:

$$\frac{1}{2} z^n \frac{\partial}{\partial \lambda} \lambda (b^+ + \lambda b^*)^{n-1} \Big|_{\lambda=1}.$$

Thus:

$$\bar{n} = \frac{z}{2} \frac{2-zb}{1-zb} + \frac{z}{2} \frac{zb^*}{(1-zb)^2}. \quad (31)$$

Comparison with equation (30) gives:

$$\beta P = \bar{n} - \frac{z}{2} \frac{zb^*}{(1-zb)^2}. \quad (32)$$

Direct calculation of  $S$  is not straightforward in this approximation: however, since in percolation theory the singular point for  $\bar{n}$  is also the point where  $S$  is divergent (though there is no general proof of this) one might be led to the conclusion (from equation (31)) that the percolation point still coincides with the condensation point in the chain approximation. We are now trying to give a definite answer to this question by a calculation of  $S$  via the pair connectedness function (Essam 1973).

## 5. Conclusions

General series expansions have been obtained for the distribution of physical clusters in interacting systems.

These can be used in connection with the general techniques for diagrammatic summations in a variety of problems. For the particular case of the relation between condensation and percolation we have shown that the percolation and condensation point (in the sense of Mayer's theory) coincide to lowest order in the chain approximation. It is now of great interest to carry on to sum all tree diagrams: this approximation is known to be equivalent to the second virial coefficient in the density expansion of the pressure and gives condensation in the sense of Van der Waal's theory; the question is which is the behaviour of the percolation quantities in the same approximation.

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