ON THE CONSTRUCTION OF PARTICLE DISTRIBUTIONS WITH SPECIFIED SINGLE AND PAIR DENSITIES

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Abstract. We discuss necessary conditions for the existence of probability distribution on particle configurations in \(d\)-dimensions i.e. a point process, compatible with a specified density \(\rho\) and radial distribution function \(g(r)\). In \(d = 1\) we give necessary and sufficient criteria on \(\rho g(r)\) for the existence of such a point process of renewal (Markov) type. We prove that these conditions are satisfied for the case \(g(r) = 0, r < D\) and \(g(r) = 1, r > D\), if and only if \(\rho D \leq e^{-1}\): the maximum density obtainable from diluting a Poisson process. We then describe briefly necessary and sufficient conditions, valid in every dimension, for \(\rho g(r)\) to specify a determinantal point process for which all \(n\)-particle densities, \(\rho_n(r_1, ..., r_n)\), are given explicitly as determinants. We give several examples.

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1. Introduction

The microscopic structure of macroscopic systems, such as fluids, is best described by the joint \( n \)-particle densities \( \rho_n(\mathbf{r}_1, \ldots, \mathbf{r}_n) \) where the \( \mathbf{r}_1, \ldots, \mathbf{r}_n \) are position vectors in \( d \)-dimensions [1]. The most important of these are the one particle density \( \rho_1(\mathbf{r}_1) \) and the pair density \( \rho_2(\mathbf{r}_1, \mathbf{r}_2) \). For spatially homogeneous systems, the only ones we shall consider here, From these one can also obtain an infinite set of conditions in the case where only \( \rho_1 \) and \( \rho_2 \) are given [11]. These conditions are very hard (or impossible) to check so the real question is whether one can get away with a smaller number of readily checkable conditions.

A simple subset of such positivity conditions, emphasized by Percus [2] and by Stillinger, Torquato, et al. [4]-[9], which follow directly from the definitions are,

\[
(1.1) \quad \rho > 0, \quad g(\mathbf{r}) \geq 0,
\]

\[
(1.2) \quad \hat{S}(\mathbf{k}) = \rho + \rho^2 \int_{\mathbb{R}^d} e^{i \mathbf{k} \cdot \mathbf{r}} [g(\mathbf{r}) - 1] d\mathbf{r} \geq 0
\]

Conditions (1.1) are obvious while (1.2) ensures that variances of one particle sum functions, \( \psi = \sum \phi(\mathbf{r}_i) \), are non-negative, since it follows
from the definition of $\rho_2$ that

\begin{equation}
\langle (\psi - \langle \psi \rangle)^2 \rangle = \left( \frac{1}{2\pi} \right)^d \int dk |\hat{\phi}(k)|^2 \hat{S}(k)
\end{equation}

where $\hat{\phi}(k) = \int e^{ikr} \phi(r) dr$, and the averages are with respect to the probability distribution of the point process. Simple considerations, see [3], show that one should add to these (1.1) and (1.2) at least one further requirement: the variance $V_\Lambda$ of the number of particles $N_\Lambda$ in a region $\Lambda$, which corresponds to $\phi(r)$ being the characteristic function of the region $\Lambda$, must be larger than $\theta(1 - \theta)$, where $\theta$ is the fractional part of the average number of particles in $\Lambda$. That is, if $\langle N_\Lambda \rangle = k + \theta$, for $k$ a non-negative integer, $k = 0, 1, 2, \ldots$, then

\begin{equation}
V_\Lambda = \langle (N_\Lambda - \langle N_\Lambda \rangle)^2 \rangle \geq \theta(1 - \theta).
\end{equation}

The bound (1.4) comes from the fact that $N_\Lambda$ can only take non-negative integer values, see [3] and Appendix where a more general condition of type (1.4) is proven.

A simple one dimensional example for which (1.1) and (1.2) are satisfied but (1.4) is violated, is

\begin{equation}
g(r) = \begin{cases} 
2(\rho r)^3, & r < \rho^{-1} \\
1, & r > \rho^{-1}
\end{cases}
\end{equation}
A direct computation shows that \( \hat{S}(k) \geq 0 \), with equality holding for \( k = 0 \), (see below), while the variance in an interval of length \( L \), \( L > \rho^{-1} \) is equal to \( 1/5 \), which violates (1.4), whenever \( \theta(1 - \theta) > 1/5 \). It is possible that the condition (1.4) becomes less important in higher dimensions where the minimal variance will go to infinity as the domain grows. For spherical domains it will grow at least like the surface area \( [12], [13] \). Note that by choosing the value of \( r \) beyond which \( g(r) = 1 \) as slightly smaller than \( \rho^{-1} \) (1.1) and (1.2) would be satisfied but (1.4) would not for some \( L \).

It is however possible, especially for the type of \( g(r) \) considered in [4]–[9] that (1.1) and (1.2) are enough to ensure realizability. These \( g(r) \) have a hard core exclusion, prohibiting the centers of two particles from coming closer than a certain distance \( D \), i.e. \( g(r) = 0 \) for \( r < D \). In particular, it was conjectured in [4]–[8] that it is possible to find a point process with density \( \rho > 0 \) and a \( g(r) \) of the form

\[
g(r) = \begin{cases} 
0, & r < D \\
1, & r > D 
\end{cases}
\]

(1.6)

as long as \( \rho v(D) \leq 2^{-d} \), where \( v(D) \) is the volume of a \( d \)-dimensional sphere of radius \( D/2 \). For \( \rho v(D) > 2^{-d} \), \( \hat{S}(k) \) will be negative for \( k = 0 \). (1.6) also satisfies condition (1.4) for \( \rho v(D) \leq 2^{-d} \), although this was not explicitly imposed. There are also heuristic arguments, bolstered
by computer simulations \[7\] and by considerations of the \(d \to \infty\) limit 
\[14\], for the realizability of (1.6) when \(\rho v(D) \leq 2^{-d}\).

The case \(\rho v(D) = 2^{-d}\), for which \(\hat{S}(0) = 0\), is of particular interest,
since it yields a system for which the variance \(V_\Lambda\) grows only like the
surface area of the boundary of \(\Lambda\) rather than the volume. Such systems
are variously called superhomogeneous \[15\], \[17\], or hyperuniform \[8\].

In \(d = 1\), the variance in an interval of length \(L\), \(V_L\) can actually be
bounded uniformly in \(L\) as analyzed in \[15\], \[17\]. Thus for the example
(1.6) with \(\rho D = 1/2\),

(1.7) \[ V_L = \begin{cases} 
\rho L(1 - \rho L), & \rho L \leq \frac{1}{2} \\
\frac{1}{4}, & \rho L \geq \frac{1}{2} 
\end{cases} \]

This is, by (1.4), the minimum permissible variance when \(\rho L = k + \frac{1}{2}, k = 0, 1, 2, \ldots\).

Inspired by the work of Stillinger and Torquato we give here a proof of
realizability of the model \(g(r)\) in (1.6) for the case \(d = 1\) and \(\rho v(D) = \rho D \leq e^{-1}\). This is based on a particular construction of the point
process as a dilution of a Poisson process with \(\rho D = \lambda D \exp[-\lambda D]\). It
turns out that the new process is a Markov or renewal process \[18 - 19\]. This permits us to describe all higher order correlation functions
in terms of \(g(r)\). We do not know at present whether there exist non
renewal point processes for some or all \(\rho D \in (e^{-1}, \frac{1}{2}]\). We also do
not know whether the explicitly constructed process for $\rho D \leq e^{-1}$ is unique. In principle there can exist more than one process with the same $\rho$ and $g(r)$ but different higher order correlations; see sec. 5.

We note that one dimensional renewal processes, described in sec. 2, and determinantal processes for arbitrary dimension, described in sec. 4, are the only examples we know for which one can explicitly (and easily) construct higher order correlations from $\rho_1$ and $\rho_2$. In some cases these processes correspond to the distribution of particles in equilibrium systems. There is also a formula for the entropy of a renewal process in terms of $g(r)$ [19].

2. Renewal Processes

A translational invariant one dimensional particle system with density $\rho > 0$, is described by a renewal process (RP) whenever the conditional probability density of finding a particle (or point) at a position $q$ on the line, given the configuration of all particles to the left of $q$, say, ..., $q_{-1} < q_0 < q$, depends only on $x = q - q_1$ [18], [19]. Let us call that density $P_1(x)$. In other words, given that there is a particle at $q$, $P_1(x)$ is the probability density that the first particle to the right (left)
of $q$ is at $q + x$ ($q - x$). This corresponds, if we think of the points as events in time, to a Markov process. Clearly

\begin{equation}
(2.1) \quad \int_0^\infty P_1(x)dx = 1, \quad \int_0^\infty xP_1(x)dx = \rho^{-1}
\end{equation}

Calling $P_n(x)$ the probability density for finding the $n$th particle at a distance $x$ to the right of the specified position of a given particle we have

\begin{equation}
(2.2) \quad P_n(x) = \int_0^x P_{n-1}(x-y)P_1(y)dy, \quad n = 2, 3, ...
\end{equation}

By the definition of $\rho g(r)$ we have

\begin{equation}
(2.3) \quad \rho g(r) = \sum_{n=1}^{\infty} P_n(r)
\end{equation}

Taking the Laplace transform of (2.3), using (2.2), then gives

\[ \rho \tilde{g}(s) \equiv \rho \int_0^\infty e^{-sr}g(r)dr = \sum_{n=1}^{\infty} [P_1(s)]^n \]

\begin{equation}
(2.4) \quad = \frac{\tilde{P}_1(s)}{[1 - \tilde{P}_1(s)]}
\end{equation}

Conversely, a given $\rho$ and $g(r)$ will be realizable as a renewal point process if and only if

\begin{equation}
(2.5) \quad \tilde{Q}(s) = \frac{\rho \tilde{g}(s)}{[1 + \rho \tilde{g}(s)]}
\end{equation}
is the Laplace transform of a probability density, $P_1(r) \geq 0$, satisfying (2.1). We will show in the next section that for the one dimensional $g(r)$ given in (1.6) this is true when and only when $\rho D \leq e^{-1}$.

It is clear from the definition of a renewal process that the higher order correlation functions of such a system can be readily expressed in terms of $\rho$ and $g(r)$. More specifically given points $x_1 < x_2 < \ldots < x_n$ on the line we have for $n = 3, 4, \ldots$

\begin{equation}
(2.6) \quad \rho_n(x_1, \ldots, x_n)/\rho_{n-1}(x_1, \ldots, x_{n-1}) = \rho g(x_n - x_{n-1})
\end{equation}

since the left side is just the particle density at $x_n$ given that there are particles at $x_1, \ldots, x_{n-1}$. Thus

\begin{equation}
(2.7) \quad \rho_3(x_1, x_2, x_3) = \rho^3 g(x_2 - x_1)g(x_3 - x_2), \quad x_1 < x_2 < x_3,
\end{equation}

etc.

There is also a simple expression for $s$, the entropy per unit length of a renewal process [19], [17]. It is given by the following formula, see Aizenman-Goldstein-Lebowitz

\begin{equation}
(2.8) \quad s = -\rho \int_0^\infty P_1(r) \log[P_1(r)/W_0(r)]dr + \rho
\end{equation}

where $W_0(r) = \int_r^\infty P_1(y)dy$ is the probability that there is no particle between $q$ and $(q + r)$. 
We can realize an RP as an equilibrium system of particles in \( d = 1 \) in which only nearest neighbors interact: there are no interactions between non-nearest neighbor particles, whatever the distances between them. For such a pair interactions \( u(r) \), \( P_1(r) \) is given by

\[
P_1(r) = Ce^{-\beta[pr + u(r)]}, \quad r > 0,
\]

where \( \beta \) is the reciprocal temperature, \( p = p(\beta, \rho) \) is the pressure and \( C = \left[ \int_0^{\infty} e^{-\beta[pr + u(r)]} dr \right]^{-1} \) is a normalization constant. Conversely given \( P_1(r) \) we can always define a \( \beta u(r) \) and the corresponding \( \beta p \) by inverting \( 2.9 \).

A well known example of such an equilibrium system with only pair interactions is that of hard rods with diameter \( D \), \( u(r) = \infty \) for \( r < D \), \( u(r) = 0 \) for \( r > D \). For this system \( P_1(r) = 0 \), for \( r < D \), and

\[
P_1(r) = \beta pe^{-\beta p[r-D]}, \quad \text{for} \quad r \geq D
\]

with

\[
\beta p = \rho [1 - \rho D]^{-1}.
\]

Eq. \( 2.10 \) then gives the well known formula for the entropy density of this system

\[
s = -\rho \log[\rho/(1 - \rho D)] + \rho
\]
3. The Realizability of (1.6) as an RP

By general theorems a necessary and sufficient condition for a function of $s$, to be the Laplace transform of a non-negative density is that it be “completely monotone” for all $s \geq 0$ [20]. That is, it is required that its derivatives alternate in sign for all $s \geq 0$. Thus for $g$ to define a renewal process it is necessary and sufficient that $\bar{Q}(s)$ in (2.5) have the property that

\begin{equation}
(-1)^k Q^{(k)}(s) > 0, \text{ for all } k = 0, 1, 2, ..., \text{ and all } s \geq 0,
\end{equation}

where $f^{(k)}(s) \equiv d^k f(s)/ds^k$.

For the $g(r)$ in (1.6),

\begin{equation}
\bar{g}(s) = \rho \int_{D}^{\infty} e^{-sr} dr = \rho s^{-1} e^{-sD}
\end{equation}

and the corresponding $\bar{Q}(s)$ in (2.5) is

\begin{equation}
\bar{Q}(s) = \rho e^{-sD}/[s + \rho e^{-sD}]
\end{equation}

It can be shown that (3.1) will be satisfied by (3.3) if and only if $\rho D \leq e^{-1}$ [21]. Here we provide a simple construction of this point process by starting with a Poisson process on the line, $x \in (-\infty, \infty)$, with density $\lambda$ and removing points which are too close ending up with a density $\rho = \lambda e^{-\lambda D}$ and the step $g(r)$ of (1.6).
The procedure is as follows. Denote the points of the Poisson process, by ..., −$x_2$, −$x_1$, $x_0$, $x_1$, $x_2$, $x_3$, ..., with $x_i \leq x_{i+1}$. Then if $(x_{i+1} - x_i) < D$, $x_i$ is removed; if $(x_{i+1} - x_i) \geq D$, $x_i$ stays. Now the probability that $(x_{i+1} - x_i)$ is greater than $D$ is $e^{-\lambda D}$ so the density of remaining points is

\begin{equation}
\rho D = \lambda De^{-\lambda D} \leq e^{-1}.
\end{equation}

The last inequality follows from the fact that $ye^{-y}$ has its maximum value $e^{-1}$ at $y = 1$. Note that for $\rho D < e^{-1}$ there are two different values of $\lambda$ which lead to the same RP with density $\rho$ (see below).

The new translation invariant process with density $\rho$ clearly has $g(r) = 0$ for $r < D$. To see that $g(r) = 1$ for $r > D$ we note that, given a surviving point at position $q$, the density of other surviving points at $q + r$ is, for $r \geq D$, just the density of points for the Poisson processes which have survived, i.e. $\lambda e^{-\lambda D} = \rho$.

It is clear from the above construction that the new process satisfies the conditions at the beginning of sec. 2 and so is an RP with $P_1(r) = Q(r)$, the inverse Laplace transform of $\bar{Q}(s)$ in (3.3). To compute $Q(r)$ we use units in which $\rho = 1$. Define

\begin{equation}
Q(r) = Q(y + nD) = w_n(y), \quad \text{for } nD \leq r < (n + 1)D
\end{equation}
and \(0 \leq y \leq D\), \(n = 0, 1, 2, \ldots\). It is then easy to deduce from (3.3) that

\[
(3.6) \quad w_{n+1}(y) = w_{n+1}(0) - \int_0^y w_n(x)dx, \quad n \geq 0
\]

with

\[
(3.7) \quad w_0(y) = 0, \quad w_1(y) = 1, \quad w_2(y) = 1 - y, \ldots
\]

Furthermore

\[
(3.8) \quad w_{n+1}(0) = w_n(D) \quad \text{for } n \geq 1
\]

i.e., \(Q(r)\) is continuous for \(r > D\).

Define now,

\[
(3.9) \quad \psi(\lambda; y) = \sum_{n=1}^{\infty} \lambda^n w_n(y)
\]

It follows then from (3.6) that

\[
(3.10) \quad \psi(\lambda, y) = \psi(\lambda, 0) - \lambda \int_0^y \psi(\lambda, x)dx
\]

and thus

\[
(3.11) \quad \psi(\lambda; y) = \psi(\lambda; 0)e^{-\lambda y}.
\]

Putting \(y = D\) then yields

\[
(3.12) \quad \psi(\lambda; D) = \psi(\lambda; 0)e^{-\lambda D} = \frac{1}{\lambda}[\psi(\lambda; 0) - \lambda]
\]
where the last equality follows from (3.8) and (3.7). This gives

\[ \psi(\lambda; 0) = \frac{\lambda}{1 - \lambda e^{-\lambda D}}. \]

The positivity of \( Q(r) \) is equivalent to the requirement that all the coefficients \( C_j \) in the expansion of

\[ \psi(\lambda; 0) = \sum_{j=0}^{\infty} C_j \lambda^j \]

are positive. This again leads to the requirement that \( D \leq e^{-1} \), with the explicit formula (due to E. Speer)

\[ w_n(y) = \sum_{k=1}^{n} [(n - k)D + y]^{k-1}(-1)^{k-1}/(k - 1)!, \quad D \leq e^{-1}. \]

4. **Determinantal Point Process**

We review here briefly how one can obtain point processes from a \( g(r) \) satisfying certain inequalities in any dimension. The construction of such processes is a subject of great current interest in mathematics and we refer the reader to [22] for more information. We again restrict ourselves to homogeneous systems and choose units in which \( \rho = 1 \). Let \( B(r) \) be a complex function such that

\[ B(r) = B^*(-r), \quad B(0) = 1 \]
and

\[
0 \leq \hat{B}(k) \equiv \int_{\mathbb{R}^d} e^{-ik \cdot r} B(r) d\mathbf{r} \leq 1
\]

It can then be proven that conditions (4.1) and (4.2) are necessary and sufficient for the existence of a point process with \(n\)-particle densities given by the determinants (4.3)

\[
\rho_n(\mathbf{r}_1, \ldots, \mathbf{r}_n) = \begin{vmatrix}
1 & B(\mathbf{r}_{12}) & \cdots & B(\mathbf{r}_{1n}) \\
B(\mathbf{r}_{21}) & 1 & \cdots & B(\mathbf{r}_{2n}) \\
\vdots & \vdots & \ddots & \vdots \\
B(\mathbf{r}_{n1}) & B(\mathbf{r}_{n2}) & \cdots & 1
\end{vmatrix}
\]

where \(\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j\). In particular we have

\[
g(\mathbf{r}) = 1 - |B(\mathbf{r})|^2
\]

with \(g(0) = 0\), \(g(r) \leq 1\).

Such a process is called a determinantal point process (DP). It follows that given a \(g(\mathbf{r})\), such that the Fourier transform of \(B(\mathbf{r}) \equiv [1-g(\mathbf{r})]^{1/2}\) satisfies (4.2) and \(g(0) = 0\), \(0 \leq g(r) \leq 1\), we can construct a point process with explicit correlations (4.3). This gives a large (uncountable) class of \(g(\mathbf{r})\) which have the realizability property. For all details, see [22] and references there.

We make two remarks:
1) To obtain a superhomogeneous system \[13\]–\[17\] with \( \hat{S}(0) = 0 \), for the determinantal point process specified by some \( B(r) \), it is necessary and sufficient that \( \hat{B}(k) \) be a characteristic function of a set \( \Omega \) in \( k \)-space, i.e. \( \hat{B}(k) = 1 \) for \( k \in \Omega \), \( \hat{B}(k) = 0 \) otherwise. This is the case for the well known one dimensional system of particles on a circle with pair interaction \( \phi(r_{ij}) = -e^2 \log |r_{ij}| \), at reciprocal temperature \( \beta = 2e^{-2} \). For this system, with \( \rho = 1 \), the infinite volume limit of the radial distribution function is given by \( g(r) = 1 - (\sin \pi r/\pi r)^2 \) and the variance \( V_L \) of the number of particles in an interval of length \( L \) grows like \( \log L \). This system is sometimes referred to as the Dyson gas: the \( \rho_n \) describe the correlations of the eigenvalues of random Gaussian Hermitian Matrices \[23\], \[24\].

2. To get translation invariant determinantal correlation functions as in \[4.3\] it is not necessary that \( B \) depend only on \( r_{12} \). It is only necessary that \( B(r_1, r_2) = F(r_{12})e^{i[\phi(r_1) - \phi(r_2)]} \) with \( B(r_1, r_2) \) satisfying, as an operator, the analogue of \[4.1\] \[22\], \[25\]. This is the case for a two dimensional one component plasma with \( \phi(r_{ij}) = -e^2 \log |r_{ij}| \), \( \beta = 2e^{-2} \[13\], \[23\]. For this system the variance in the number of particles in a disc of radius \( R \) grows like \( R \[13\]. \)
5. Example and Discussion

We illustrate here the construction of a DP in $d$ dimensions from a given $\rho_1$ and $\rho_2$ which is, in $d = 1$, also an RP. As in the example (1.6) this can be done for only a subset of the parameter for which (1.1), (1.2) and (1.4) are satisfied. On the other hand everything here can be computed explicitly in an elementary way. Using units in which $\rho = 1$, let

$$g(r) = 1 - e^{-\lambda r}, \quad \lambda \geq 0.$$  \hfill (5.1)

It is easily checked that this $g$ satisfies (1.1), (1.2) and (1.4), whenever $\lambda \geq \lambda_d$, $\lambda_1 = 2$, $\lambda_3 = (8\pi)^{1/3}$, ... It follows from (4.4) that this $g$ determines a DP with $B = e^{-\lambda r/2}$ whenever $\lambda \geq 2\lambda_d$.

On the other hand, using (2.5), we get for $d = 1$, the Laplace transform,

$$\bar{Q}(s) = \lambda/[s^2 + \lambda s + \lambda]$$  \hfill (5.2)

from which one readily finds, by factorizing the denominator in (5.2) and using criteria (3.1), that (5.1) determines a RP if and only if $\lambda \geq 2\lambda_1 = 4$. For such values of $\lambda$, it is then easily found that
\[ P_1(r) = \lambda[\lambda^2 - 4\lambda]^{-1/2}e^{-\lambda r}\left\{\exp\left[-\frac{\lambda + \sqrt{\lambda^2 - 4\lambda}}{2}r\right]ight. - \left.\exp\left[-\frac{\lambda - \sqrt{\lambda^2 - 4\lambda}}{2}r\right]\right\}. \quad \lambda > 4, \quad (5.3a) \]

\[ P_1(r) = 4re^{-2r}. \quad \lambda = 4 \quad (5.3b) \]

It is now easy to check that (2.6) and (4.3) give the same correlations \( \rho_n(x_1, ..., x_n) \). In particular for all \( \lambda \geq 4 \),

\[ (5.4) \quad \rho_3(x_1, x_2, x_3) = (1 - e^{-\lambda(x_2-x_1)})(1 - e^{-\lambda(x_3-x_2)}), \quad x_1 \leq x_2 \leq x_3 \]

Using (5.3) in (2.8) one can also obtain the entropy of this system for \( \lambda \geq 4 \).

The fact that the DP and RP constructions in the above example yield the same point process might suggest that an RP or DP determines a unique point process. This may indeed be the case. We note, however, that uniqueness is not true in general, as can be seen from considerations of systems with non-reflection invariant correlations. Thus, while we always have, for translation invariant systems, that \( g(r) = g(-r) \), there is no such symmetry for the higher order \( \rho_n \). In particular there is an explicit construction of a translation invariant point processes in \( d = 1 \) which, when run “backwards” will have the same \( \rho \) and \( g(r) \) as the original process but a \( \rho'_3 \) obtained from the original one.
by reflection, i.e. $\rho_3(x_1, x_2, x_3) = \rho_3(-x_1, -x_2, -x_3) \neq \rho_3(x_1, x_2, x_3)$; see [15] for details.

The sufficiency of conditions (1.1), (1.2) and (1.4) (with the generalizations given in the Appendix) remains open although it seems unlikely that any finite number of conditions would suffice for the general case [2], [3], [11]. The construction of the step $g(r)$ in (1.6) by the dilution of a Poisson process does not seem to work in $d > 1$. On the other hand we have not found any counterexample so far.

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Appendix: Proof of (1.4)

We give here an elementary proof of (1.4) and its generalizations, see also [3]. Let $P(k) = \text{Prob. of having } k \text{ particles in } \Lambda$ such that

$$
\langle k \rangle = \sum_{k=0}^{\infty} k P(k) = N + \theta, \quad N = 0, 1, 2, \ldots, \quad 0 \leq \theta \leq 1.
$$
Then the variance

\[ V_\Lambda = \sum_k [k - N - \theta]^2 P(k) \]

\[ = \sum (k - N)^2 P(k) - 2\theta[\langle k \rangle - N] + \theta^2 \]

\[ = \sum (k - N)^2 P(k) - \theta^2 \]

\[ \geq \sum (k - N)P(k) - \theta^2 = \theta(1 - \theta). \]

The inequality follows from the fact that \( n^2 \geq n \) for \( n \) an integer. Equality occurs when \( P(k) = \alpha \delta_{k,N} + (1 - \alpha) \delta_{k,N+1} \), with \( \alpha \) determined by \( \theta \).

We note that the same argument works also for the variance of a linear combination of the number of particles \( N_{\Lambda_i} \) in regions \( \Lambda_i \). Let \( Y = \sum_{i=1}^k m_i N_{\Lambda_i} \), where \( m_i \) are integer coefficients, then \( \langle (Y - \langle Y \rangle)^2 \rangle \geq \theta(1 - \theta) \). In particular consider the difference between the number of particles in a region \( \Lambda_1 \) and a region \( \Lambda_2 \). Letting \( \langle (N_{\Lambda_1} - N_{\Lambda_2}) \rangle = K + \theta, K = 0, \pm 1, \pm 2, \ldots \) we again have

\[ V_{1,2} = \langle [N_{\Lambda_1} - N_{\Lambda_2} - K - \theta]^2 \rangle \geq \theta(1 - \theta) \]

References

[1] J. -P. Hansen and I. R. McDonald, Theory of Simple Liquids (Academic Press, New York, 1986). See articles by J. K. Percus, G. Stell and others in Classical Fluids, H. L. Frisch and J. L. Lebowitz, editors, (Benjamin, N.Y., 1964).
Liquid State of Matter: Fluids, Simple and Complex, E. W. Montroll and J. L. Lebowitz, Editors. (North-Holland, N.Y. 1982).

[2] J. K. Percus, article in reference 1 and references there.

[3] M. Yamada, Progress of Theoretical Physics, 25, 579 (1961).

[4] F. H. Stillinger, S. Torquato, J. M. Eroles, and R. M. Truskett, J. Phys. Chem. B 105, 6592 (2000).

[5] S. Torquato and F. H. Stillinger, Journal of Physical Chemistry B, 106, 8354 (2002); Erratum: ibid, 11406 (2002)

[6] H. Sakai, S. Torquato, and F. H. Stillinger, Journal of Chemical Physics, 117, 297 (2002).

[7] J. Crawford, S. Torquato, and F. H. Stillinger, Journal of Chemical Physics, 119, 7065-7074 (2003)

[8] S. Torquato and F. H. Stillinger, Physical Review E 68, 041113 (2003).

[9] F. H. Stillinger and S. Torquato, “Pair Correlation Function Realizability: Lattice Model Implications”, to appear.

[10] A. Lenard, Comm. Math. Phys. 30, 35 (1973); Arch. Rat. Mech. Anal. 59, 219 and 240 (1975).

[11] T. Kuna and J. L. Lebowitz, in preparation.

[12] J. Beck, Acta. Math. 1, 159 (1987).

[13] J. L. Lebowitz, Phys. Rev. A 27, 1491 (1983); D. Levesque, J. Weis and J. L. Lebowitz, Journal of Statistical Physics, 100, 209-222, 2000

[14] S. Torquato and F. H. Stillinger, in preparation.

[15] S. Goldstein, J. L. Lebowitz, E. R. Speer, in preparation.
[16] A. Gabrielli, B. Jancovici, M. Joyce, J. L. Lebowitz, L. Pietronero, and F. S. Labini, *Phys. Rev. D*, 67, 043506 (2003).

[17] M. Aizenman, S. Goldstein and J. L. Lebowitz, *J. Stat. Phys.* 103, 601 (2001).

[18] D. J. Daley and D. Vere-Jones, *An Introduction to the Theory of Point Processes*, (Springer-Verlag, N.Y., 1988).

[19] R. R. Cox and V. Isham, *Point Processes*, (Chapman and Hall, London, 1980).

[20] D. V. Widder, *Laplace Transforms*, Princeton University Press, 1941.

[21] O. Costin, Three applications of Complex Analysis and Resurgence, submitted.

[22] A. Soshnikov, *Russian Math. Surveys* 55:5, 923 (2000); *J. Stat. Phys.* 100, 491 (2000).

[23] M. L. Mehta, *Random Matrices*. (Academic Press, N.Y. 1991).

[24] O. Costin and J. L. Lebowitz, *Physical Review Letters*, 75, 69-72, 1995.

[25] A. Soshnikov, private communication.