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NEW DERIVATION OF THE CLUSTER CUMULANT FORMULA

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Dedicated to the memory of Professor Vladimir Šips

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The cluster cumulant formula of Kubo is derived by appealing only to elementary properties of subsets and binomial coefficients. It is shown to be a binomial transform of the grand potential. Extensivity is proven without introducing cumulants. A combinatorial inversion is used to reformulate the expansion in the activity to one in occupation probabilities, which explicitly control the convergence. The classical virial expansion is recovered to third order as an example.

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1. Introduction

The classical (Ursell–Yvon–Mayer) virial expansion is the traditional introduction to real-space particle correlations in basic statistical physics textbooks, both old [1] and new [2]. By contrast, cumulant methods, of which the virial expansion is a special case, are left to more specialized texts [3]. A possible disadvantage of this approach is that physical issues are initially confused with purely formal ones, stemming from the specific expression for the grand potential $\Omega = -k_\mathrm{B}T\,\Psi$ as a logarithm of a sum

$$\Psi = \ln \sum_N \mathrm{e}^{\beta\mu N} \mathrm{tr_N} \exp(-\beta \mathrm{H}). \tag{1}$$

The most prominent formal problem is extensivity: it is important to demonstrate that the "factorization property" of matrix elements leads to a Ψ proportional to the number of single-particle states.

The formal and physical side of the problem were neatly separated by Kubo [4]. The present article is built around a very compact formal derivation of one of his main formulas, appealing only to elementary algebra. Its principal feature is that the usual ordering, by which the cluster cumulant formula is a rearrangement of the cumulant expansion, is reversed, and the cluster formula appears as the basic one. At the level considered here, operative formulas can be developed without invoking cumulants at all, physics being determined by the choice of expansion parameter. Given the importance of cumulants in general, this is not necessarily a pedagogical advantage. Nevertheless, it is hoped that the present work gives a reasonable introduction to the issues involved in cumulant expansions, at a level still attainable in a general (graduate) course of statistical physics. To this end, the virial series is rederived as an example, not because this is the quickest way to do it (it obviously is not), but in order to provide a list, in some logical order, of the steps involved in reducing the fully general quantum formula to an operative classical limit. Once such a framework is established, each of these steps can be made the starting point of a more specialized development.

Being concerned with the interpretation, rather than the calculation, this article cannot do justice to the numerous, sometimes very refined, applications of the cumulant approach, in its broadest sense, which have developed over a period of more than half a century. Outside the original context of a classical gas [5], perhaps the most detailed work was done on spin models, under the heading of high-, and low-temperature expansions [6]. Another long-standing field of applications are polymers, including fundamental issues, such as the excluded-volume problem far from the gelation point [7]. Strong electron correlations [8−11] have also been described by cumulant expansions. The common denominator of all these applications is the need to treat correlations in real space, when neither scaling nor periodicity can be used to simplify the problem. An interesting variant of the cumulant method is when a finite system is treated in some "inverse" space, which diagonalizes part of the Hamiltonian; this is a classic microscopic approach in nuclear physics [12].

Concerning the presentation, I am aware that combinatorial manipulations are not part of the usual education of a physicist. If the binomial transform were as commonly known as the Fourier transform, the main result in the next section would be a "one-liner". I have tried to spell everything out, using familiar set-theoretic notation. A word of advice to the diligent reader: follow the formulas on a small example, with two, or at most three, single-particle states. If it suddenly appears easy, that is because it is.

2. The Kubo formula for fermions

2.1. The activity expansion

The diagonal matrix element in N-particle space is denoted

$$U_{i(N)} \equiv \langle i_1, \ldots, i_N | \, \mathrm{e}^{-\beta H} \, | i_1, \ldots, i_N \rangle \,, \tag{2}$$

where a set of occupied single-particle states (configuration) is denoted by $i(N) \equiv \{i_1, \ldots, i_N\}$. It is assumed that the total number of single-particle states is L, and they comprise a set, to be denoted $\mathcal{L}$. In this set-theoretic notation, the Massieu potential (1) reads

$$\Psi = \ln \sum_{\emptyset \subseteq i(N) \subseteq \mathcal{L}(\mathcal{L})} \mathrm{e}^{\beta\mu N} U_{i(N)}. \tag{3}$$

The sum is over all subsets of of the set $\mathcal{L}$ of single-particle states, which may of course take at most L fermions. It will be assumed throughout that the vacuum expectation $U_\emptyset = 1$. All derivations in this article will refer to fermions. Only the final expressions for bosons are given in the Appendix.

The motivation for the next step is that one would like a sum outside the logarithm, not inside it. So define an intermediate expression,

$$\Psi_M \equiv \sum_{j(M) \subseteq \mathcal{L}} \ln \sum_{\emptyset \subseteq i(N) \subseteq j(M)} \mathrm{e}^{\beta\mu N} U_{i(N)}. \tag{4}$$

Here the sum inside is limited to some subset of $\mathcal{L}$, with exactly M states, and the sum outside is over all possible choices of such a subset. Notice that $\Psi = \Psi_L$, since the sum outside the logarithm then reduces to a single term, $j(L) = \mathcal{L}$.

Now introduce the pair of inverse relations

$$\Psi_M = \sum_{m=0}^{M} \binom{L-m}{M-m} \Psi(m), \tag{5}$$

$$\Psi(m) = \sum_{M=0}^{m} (-1)^{m-M} \binom{L-M}{m-M} \Psi_M. \tag{6}$$

If one of these is inserted into the other, the result is an identity, so they are valid independently of what the Ψ_M may be. Taking Eq. (5) at $M = L$, one finds

$$\boxed{\Psi = \sum_{m \geq 1} \Psi(m),} \tag{7}$$

which is the Kubo cluster cumulant expansion, when $\Psi(m)$ is given in terms of the Ψ_M in Eq. (4). (Note that $\Psi(0) = \Psi_0 = \ln U_\emptyset = 0$.)

To see the structure of Eq. (7), look at the first two terms, reverting to standard notation for a moment

$$\Psi(1) = \sum_{i=1}^{L} \ln\left(1 + \mathrm{e}^{\beta\mu} \langle i| \, \mathrm{e}^{-\beta H} \, |i\rangle\right), \tag{8}$$

$$\Psi(2) = \sum_{1 \leq i < j \leq L} \ln\Big(1 + \mathrm{e}^{\beta\mu} \langle i| \, \mathrm{e}^{-\beta H} \, |i\rangle + \mathrm{e}^{\beta\mu} \langle j| \, \mathrm{e}^{-\beta H} \, |j\rangle$$

$$
\begin{aligned}
&\qquad +\mathrm{e}^{2\beta\mu}\left\langle i,j\right|\mathrm{e}^{-\beta H}\left|i,j\right\rangle\big) - (L-1)\sum_{i=1}^{L}\ln\left(1+\mathrm{e}^{\beta\mu}\left\langle i\right|\mathrm{e}^{-\beta H}\left|i\right\rangle\right) \\
= &\sum_{1\le i<j\le L}\big[\ln\big(1+\mathrm{e}^{\beta\mu}\left\langle i\right|\mathrm{e}^{-\beta H}\left|i\right\rangle+\mathrm{e}^{\beta\mu}\left\langle j\right|\mathrm{e}^{-\beta H}\left|j\right\rangle \\
&\qquad +\mathrm{e}^{2\beta\mu}\left\langle i,j\right|\mathrm{e}^{-\beta H}\left|i,j\right\rangle\big) - \ln\left(1+\mathrm{e}^{\beta\mu}\left\langle i\right|\mathrm{e}^{-\beta H}\left|i\right\rangle\right) \\
&\qquad\quad - \ln\left(1+\mathrm{e}^{\beta\mu}\left\langle j\right|\mathrm{e}^{-\beta H}\left|j\right\rangle\right)\big] \\
= &\sum_{1\le i<j\le L}\ln\left(1+\mathrm{e}^{2\beta\mu}\frac{\left\langle i,j\right|\mathrm{e}^{-\beta H}\left|i,j\right\rangle-\left\langle i\right|\mathrm{e}^{-\beta H}\left|i\right\rangle\left\langle j\right|\mathrm{e}^{-\beta H}\left|j\right\rangle}{\left(1+\mathrm{e}^{\beta\mu}\left\langle i\right|\mathrm{e}^{-\beta H}\left|i\right\rangle\right)\left(1+\mathrm{e}^{\beta\mu}\left\langle j\right|\mathrm{e}^{-\beta H}\left|j\right\rangle\right)}\right). \qquad (9)
\end{aligned}
$$

This should be compared with Eq. 6.22 of Ref. [4]. The lowest power of the activity appearing in $\Psi(m)$ is $\mathrm{e}^{\beta\mu m}$, so I shall refer to this as the "activity expansion". If the basis $|i\rangle$ diagonalizes the Hamiltonian, $\Psi(1)$ gives the exact solution of the trivial problem, and the other $\Psi(m)$ are zero.

Of the three forms in which $\Psi(2)$ is given, the first is the defining form Eq. (6). The second is Kubo's original form, where all terms appear under a single sum. The original form reads (Eq. 4.13 of Ref. [4])

$$\Psi(m) = \sum_{i(m)\subseteq\mathcal{L}} \psi[i(m)], \qquad (10)$$

where

$$\psi[i(m)] \equiv \sum_{\emptyset\subseteq j(M)\subseteq i(m)} (-1)^{m-M} \ln\left[\sum_{\emptyset\subseteq k(l)\subseteq j(M)} \mathrm{e}^{l\beta\mu} U_{k(l)}\right]. \qquad (11)$$

Finally, the third is a compact form, where all logarithms have been collapsed to a single one. The trouble with it, although it looks prettiest for small m, is a large (hyperexponential) explosion with m in the number of terms in the fraction multiplying $\mathrm{e}^{\beta\mu m}$, as the fraction arises from multiplication of the polynomials in $\mathrm{e}^{\beta\mu}$ which appear under the logarithms in the original form.

It is easy to prove that the defining and original forms are equivalent. One starts from Eq. (10) and simply interchanges the order of summation

$$
\begin{aligned}
\Psi(m) &= \sum_{i(m)\subseteq\mathcal{L}}\ \sum_{\emptyset\subseteq j(M)\subseteq i(m)} (-1)^{m-M}\ln[\ldots j(M)\ldots] \\
&= \sum_{\emptyset\subseteq j(M)\subseteq\mathcal{L}} (-1)^{m-M}\ln[\ldots j(M)\ldots] \sum_{j(M)\subseteq i(m)\subseteq\mathcal{L}} 1 \\
&= \sum_{\emptyset\subseteq j(M)\subseteq\mathcal{L}} (-1)^{m-M}\ln[\ldots j(M)\ldots]\binom{L-M}{m-M}
\end{aligned}
$$

$$= \sum_{M \geq 0} (-1)^{m-M} \binom{L-M}{m-M} \Psi_M,$$

where, in the last step, all M-particle configurations have been grouped together. From this point of view, the present article rests on the observation that the last line above is formally invertible.

2.2. Extensivity

Extensivity itself cannot, of course, be proven without some reference to the interactions involved. What will be proven here is more properly called size-consistency: if the matrix element $U_{i(m)}$ in a configuration $i(m)$ can be expressed as a product of lower matrix elements, then the contribution of that configuration to the grand potential is zero.

We shall need an elementary property of sets. Namely, if a set $i(m)$ is written as the union of two non-overlapping, non-empty subsets, $i(m) = i_1(m_1) \cup i_2(m_2)$, then all the subsets of $i(m)$ may be obtained by writing down all subsets of $i_1(m_1)$ and $i_2(m_2)$, and combining them in all possible ways. In particular, if there is a sum over subsets of $i(m)$, it can be written as two sums

$$\sum_{\emptyset \subseteq j(n) \subseteq i(m)} [\ldots j(n) \ldots] = \sum_{\emptyset \subseteq j_1(n_1) \subseteq i_1(m_1)} \sum_{\emptyset \subseteq j_2(n_2) \subseteq i_2(m_2)} [\ldots j_1(n_1) \cup j_2(n_2) \ldots] . \quad (12)$$

This will be referred to below as the "subset property".

The proposition is as follows: let $i(m)$ be the union of two non-overlapping, non-empty subsets, $i(m) = i_1(m_1) \cup i_2(m_2)$. Let

$$U_{k_1(l_1) \cup k_2(l_2)} = U_{k_1(l_1)} U_{k_2(l_2)} \quad (13)$$

whenever $k_1(l_1) \subseteq i_1(m_1)$ and $k_2(l_2) \subseteq i_2(m_2)$. Then $\psi[i(m)] = 0$, where $\psi[i(m)]$ is the contribution of the set $i(m)$ to $\Psi(m)$ in Eq. (10).

For the proof, first observe that under these assumptions, the sum under the logarithm in Eq. (11) factorizes,

$$\sum_{\emptyset \subseteq k(l) \subseteq j(n)} U_{k(l)} = \sum_{\emptyset \subseteq k_1(l_1) \subseteq j_1(n_1)} U_{k_1(l_1)} \sum_{\emptyset \subseteq k_2(l_2) \subseteq j_2(n_2)} U_{k_2(l_2)} . \quad (14)$$

This is trivial: the sum is rewritten by the subset property, and the matrix element factorizes by assumption. It follows that the logarithm of (14) is the sum of two logarithms, one a function of $j_1(n_1)$ alone, the other of $j_2(n_2)$. The contribution of the first logarithm to Eq. (11) reads

$$\sum_{\emptyset \subseteq j(n) \subseteq i(m)} (-1)^{m-n} \ln [\ldots j_1(n_1) \ldots] \quad (15)$$

$$= \sum_{\emptyset \subseteq j_1(n_1) \subseteq i_1(m_1)} (-1)^{m_1 - n_1} \ln [\ldots j_1(n_1) \ldots] \sum_{\emptyset \subseteq j_2(n_2) \subseteq i_2(m_2)} (-1)^{m_2 - n_2},$$

because the sum here can also be written by the subset property. Since the number of subsets of $i(m_2)$ with fixed number n_2 is given by a binomial coefficient,

$$\sum_{\emptyset\subseteq j_2(n_2)\subseteq i_2(m_2)} (-1)^{m_2-n_2} = \sum_{n_2=0}^{m_2} \binom{m_2}{n_2} (-1)^{m_2-n_2} = (1-1)^{m_2}, \tag{16}$$

this is zero for $m_2 \geq 1$; similarly the contribution of the second logarithm is zero for $m_1 \geq 1$, so the proposition is proved.

3. *The probability expansion*

In this section, the combinatorial approach is pushed a step further, to rewrite Kubo's formula (7) in a particularly transparent way. (The derivations are written somewhat more tersely than in the other sections.) Probabilities will replace the activity as the expansion parameters, analogously to passing from activity to concentration in the classical case. It should be emphasized that this only affects the form of $\psi[i(m)]$ in Eq. (10), their value remaining the same, term for term, for all $i(m)$ with $m \geq 2$. The simplest example of the transformation is in the two ways one may write $\Psi(1)$,

$$\Psi(1) = \sum_i \ln\left(1 + \mathrm{e}^{\beta\mu} U_i\right) = -\sum_i \ln\left(1 - p_i\right), \tag{17}$$

where the "occupation probability"

$$p_i = \frac{\mathrm{e}^{\beta\mu} U_i}{1 + \mathrm{e}^{\beta\mu} U_i} \tag{18}$$

would be just the Fermi function in the quantum non-interacting case.

3.1. *The general transformation*

Here a whole class of ways to expand Ψ will be shown to be equal to the activity expansion (7), term for term. In other words, they are merely different ways to rearrange the contributions under the logarithms in Eq. (11). A special choice then gives the expansion in the probabilities (18), alluded to above.

Take an arbitrary set of L variables ε_i, $i = 1, \ldots, L$. Define the quantities W by the pair of inverse relations

$$U_{i(m)} = \mathrm{e}^{-\beta \sum_{i\in i(m)} \varepsilon_i} \sum_{\emptyset\subseteq j(n)\subseteq i(m)} W_{j(n)}, \tag{19}$$

$$W_{j(n)} = \sum_{\emptyset\subseteq i(m)\subseteq j(n)} (-1)^{m-n} \mathrm{e}^{+\beta \sum_{i\in i(m)} \varepsilon_i} U_{i(m)}. \tag{20}$$

One could just write x_i instead of $\mathrm{e}^{-\beta\varepsilon_i}$, but the notation is meant to be suggestive. The grand partition function may now be written

$$\sum_{\emptyset\subseteq i(N)\subseteq\mathcal{L}(\mathcal{L})} \mathrm{e}^{\beta\mu N} U_{i(N)} = \prod_{i=1}^{L}\left(1+\mathrm{e}^{\beta(\mu-\varepsilon_i)}\right) \sum_{\emptyset\subseteq j(n)\subseteq\mathcal{L}(\mathcal{L})} f_{j_1}\dots f_{j_n} W_{j(n)}. \tag{21}$$

Here the f_i's are just the Fermi functions corresponding to the ε_i. Obviously, they are the new variables of the partition function, replacing the activity. Now define, by analogy with Eq. (4),

$$\widetilde{\Psi}_M \equiv \sum_{i(M)\subseteq\mathcal{L}} \ln \sum_{\emptyset\subseteq j(n)\subseteq i(M)} f_{j_1}\dots f_{j_n} W_{j(n)}, \tag{22}$$

and the main statement of this section is as follows

$$\widetilde{\Psi}(m) = \Psi(m), \quad m \geq 2, \tag{23}$$

where $\widetilde{\Psi}(m)$ is the binomial transform (6) of $\widetilde{\Psi}_M$. In other words, the activity expansion is unaffected by the transformation, except in the first term; it is easy to show that

$$\sum_i \ln\left(1+\mathrm{e}^{\beta(\mu-\varepsilon_i)}\right) + \widetilde{\Psi}(1) = \Psi(1), \tag{24}$$

with the ε_i's cancelling exactly. Of course, since the partition function (left-hand side of Eq. (21)) does not depend on them, they all must cancel in the end; but the statement here is that they do so term by term in the expansion, when $m \geq 2$.

To prove this, express W in $\widetilde{\Psi}_M$ back in terms of the U's,

$$\sum_{\emptyset\subseteq j(n)\subseteq i(M)} f_{j_1}\dots f_{j_n} W_{j(n)} \quad = \prod_{i\in i(M)}\left(1+\mathrm{e}^{\beta(\mu-\varepsilon_i)}\right)^{-1} \sum_{\emptyset\subseteq k(N)\subseteq i(M)} \mathrm{e}^{\beta\mu N} U_{k(N)}. \tag{25}$$

Now observe that the sum is the same one as appears in the definition (4) of Ψ_M, by which $\Psi(m)$ is defined. The difference $\Psi(m) - \widetilde{\Psi}(m)$ is thus due to the product in front, and reads explicitly, by Eq. (6),

$$\sum_M (-1)^{m-M}\binom{L-M}{m-M} \sum_{i(M)\subseteq\mathcal{L}} \sum_{i\in i(M)} \ln\left(1+\mathrm{e}^{\beta(\mu-\varepsilon_i)}\right), \tag{26}$$

which is, in fact, zero for $m \geq 2$. Namely, in the sum over configurations (subsets of $\mathcal{L}$), each *given* single-particle state will appear $\binom{L-1}{M-1}$ times, so upon exchanging the last two sums, one gets

$$\sum_{i=1}^{L} \ln\left(1+\mathrm{e}^{\beta(\mu-\varepsilon_i)}\right) \sum_M (-1)^{m-M}\binom{L-M}{m-M}\binom{L-1}{M-1}, \tag{27}$$

and after rearranging the binomial coefficients, the last sum is equal to

$$\binom{L-1}{m-1}\sum_M (-1)^{m-M}\binom{m-1}{M-1} \sim (1-1)^{m-1}, \tag{28}$$

so the statement is proven.

3.2. *The probability expansion*

By choosing

$$\mathrm{e}^{-\beta\epsilon_i} = \langle i|\,\mathrm{e}^{-\beta H}\,|i\rangle = U_i, \tag{29}$$

one finds $f_i = p_i$ (Eq. (18)), and $W_i = 0$. This is the useful case, so let us denote the W's for this special choice by the letter S, for "subtracted",

$$S_{i(m)} = \sum_{\emptyset\subseteq j(n)\subseteq i(m)} (-1)^{n-m}\widetilde{U}_{j(n)}, \tag{30}$$

where

$$\widetilde{U}_{j(n)} = \frac{U_{j(n)}}{U_{j_1}\cdots U_{j_n}}, \qquad \widetilde{U}_{\emptyset} = 1. \tag{31}$$

In terms of these, the $\psi[i(m)]$ in Eq. (11) read, for $m \geq 2$,

$$\psi[i(m)] = \sum_{\emptyset\subseteq j(n)\subseteq i(m)} (-1)^{m-n}\ln\left[\sum_{\emptyset\subseteq k(l)\subseteq j(n)} p_{k_1}\dots p_{k_l}S_{k(l)}\right], \tag{32}$$

noting that $S_\emptyset = 1$. This is the "probability expansion". For example,

$$S_{ij} = \frac{U_{ij}}{U_iU_j} - 1, \qquad \Psi(2) = \sum_{1\leq i<j\leq L} \ln(1 + p_ip_jS_{ij}), \tag{33}$$

and even $\Psi(3)$ is short, in the compact form

$$\Psi(3) = \sum_{1\leq i<j<k\leq L} \ln\Bigg[1 + p_ip_jp_k \times\frac{S_{ijk} - p_iS_{ij}S_{ik} - p_jS_{ij}S_{jk} - p_kS_{ik}S_{jk} - p_ip_jp_kS_{ij}S_{ik}S_{jk}}{(1+p_ip_jS_{ij})(1+p_ip_kS_{ik})(1+p_jp_kS_{jk})}\Bigg]. \tag{34}$$

These are higher-order corrections to the non-interacting ("undergraduate") formula (17). It is obvious how the probabilities control the convergence.

It should be noted that the "compact" form has not become really compact, but rather that the hyperexponential explosion takes off a little later (because $S_i = 0$, so the polynomials being multiplied are shorter). For instance, the numerator in the compact form of $\Psi(4)$ has 15 629 terms in the activity expansion, and "only" 505 terms in the probability expansion, still far less practical than the 15 distinct terms, 65 additions and 16 logarithms in the original form.

4. *Example*

In this section, the classical virial expansion will be obtained term by term, from the formulas developed so far. While such a derivation is nothing new in itself, the purpose is to comment on it from the present "combinatorial" point of view, and give a familiar interpretation of the subtracted matrix elements, formally introduced in the previous section.

4.1. *The classical limit*

Begin with a Hamiltonian of the form $H = K + V$, where K is the usual kinetic energy, and V a sum of two-body interactions depending on mutual distance. The first choice to be made in the formal expansion is, which basis to use for the single-particle states. If the momentum basis, which diagonalizes K, is used as a starting point, the term $\Psi(1)$ will be the exact solution of the non-interacting problem, valid down to zero temperature, and the corrections will correspond to an expansion in quasiparticle occupation probabilities, after a canonical transformation to particles and holes. (If this transformation is not made, the occupation probabilities of levels below the Fermi energy tend to unity, leading to convergence problems.) Such an example, inspired by nuclear physics [12], is beyond the scope of the present work.

If, on the other hand, the position basis is chosen, one is led straight to the Mayer expansion. This is particularly easy to see on a lattice. The one-particle matrix element is then just the normalized one-particle partition function, independently of the position,

$$U_i = \left\langle \boldsymbol{r}_i \left| \mathrm{e}^{-\beta(K+V)} \right| \boldsymbol{r}_i \right\rangle = \frac{1}{L} \sum_{\boldsymbol{k}} \exp\left[-\beta\varepsilon(\boldsymbol{k})\right] = \frac{Z_1}{L}, \tag{35}$$

where L is now the number of lattice sites, and $\varepsilon(\boldsymbol{k})$ is the non-interacting dispersion derived from K. (In the limit of vanishing activity, $\Psi(1) = L \ln(1 + \mathrm{e}^{\beta\mu} Z_1/L) \to \mathrm{e}^{\beta\mu} Z_1$, which is the classical non-interacting result.) The occupation probabilities (18) are then also independent of position, and to the first order in the activity, they become equal to the (dimensionless) fugacity,

$$p = \frac{\mathrm{e}^{\beta\mu} Z_1/L}{1 + \mathrm{e}^{\beta\mu} Z_1/L} \to \mathrm{e}^{\beta\mu} \frac{Z_1}{L} \to \mathrm{e}^{\beta\mu} \left(\frac{a}{\lambda_T}\right)^3, \tag{36}$$

where the second limit is of high temperature, with a the lattice constant and λ_T the thermal wavelength.

The classical limit for $\Psi(2)$ is taken in the usual two steps: first, the commutator $[K,V]$ is neglected, being at least of the order of $\hbar$, so that disentanglement is trivial:

$$\mathrm{e}^{-\beta(K+V)} \to \mathrm{e}^{-\beta K}\mathrm{e}^{-\beta V}, \tag{37}$$

after which the two-particle matrix element reads

$$\begin{aligned} U_{ij} &= \left\langle \boldsymbol{r}_i, \boldsymbol{r}_j \left| \mathrm{e}^{-\beta K}\mathrm{e}^{-\beta V} \right| \boldsymbol{r}_i, \boldsymbol{r}_j \right\rangle \\ &= \frac{1}{L^2}\mathrm{e}^{-\beta v_{ij}} \left[Z_1^2 - \left(\sum_{\boldsymbol{k}} \cos\left[\boldsymbol{k}\cdot(\boldsymbol{r}_i - \boldsymbol{r}_j)\right] \exp\left[-\beta\varepsilon(\boldsymbol{k})\right] \right)^2 \right]. \end{aligned} \tag{38}$$

Here $v_{ij} = V(|\boldsymbol{r}_i - \boldsymbol{r}_j|)$, and the interference term from $|\langle \boldsymbol{r}_i, \boldsymbol{r}_j | \boldsymbol{k}_i, \boldsymbol{k}_j\rangle|^2$ is shown explicitly. In physical units $\boldsymbol{k} = \boldsymbol{p}/\hbar$, so one observes in the second step that as $\hbar \to 0$, it gets "washed out" by the sum, giving

$$\Psi(2) = \sum_{1\le i<j\le L} \ln\left[1 + p^2\left(\mathrm{e}^{-\beta v_{ij}} - 1\right)\right], \tag{39}$$

where p is the position-independent occupation probability (36), multiplying Mayer's expansion parameter, $g_{ij} = \mathrm{e}^{-\beta v_{ij}} - 1$. In other words, the subtracted matrix elements S_{ij} in the probability expansion become equal to Mayer's parameter: $S_{ij} \to g_{ij}$ in the classical limit.

This is as far as one can go without invoking the probability expansion (32) explicitly, since only $\Psi(2)$ is easy to rewrite in probabilities "by hand". It is not difficult to show that, when $\hbar \to 0$ and all interference terms are neglected, the normalized matrix elements (31) in the position basis take the familiar form

$$\widetilde{U}_{j(n)} = \exp\left(-\beta \sum_{1\le k<l\le n} v_{j_k j_l} \right) = \prod_{1\le k<l\le n} (1 + g_{j_k j_l}), \tag{40}$$

where the sum (product) is over all pairs of indices in $j(n) = \{j_1, \ldots, j_n\}$. It follows that Ψ is a function of g_{ij} alone, as is well known. The S's vanish with the interaction, as they should, because all the $\widetilde{U}$'s in Eq. (40) are then equal to unity, so they cancel in the definition (30).

This is a good place to pause, and put the result (40) in a physical perspective. One could have worried: the same one-particle terms (29) appear in the denominator of the normalized matrix elements (31) as in the numerator of the probabilities (18). Might they not cancel, leaving the probabilities in expressions like (35) only formally, but not really, in control of the convergence? Not so: equation (40) shows that one should rather expect one-particle ("kinetic") terms to cancel between the numerator and denominator of $\widetilde{U}$ itself, this cancellation being complete in the classical limit. This is just the statement, that in the classical limit one can integrate out momenta from the partition function exactly. Quantum effects are manifested as incomplete cancellation: interactions do affect the momentum distribution. However, to believe that a $\widetilde{U}$ would not tend to a limit when a one-particle matrix element in the denominator became small, is to believe that the many-body state in the numerator has an "infinite stopping power" for that particle, if it can avoid making the numerator small as well, despite containing that same single-particle state of high momentum. Such drastic effects of the interaction on the momentum distribution are not unimaginable. In fact, one of the persistent worries in high-temperature superconductivity is that they could preclude any "semiclassical" description of the conducting electrons [13]. Nevertheless, it may be (vaguely) concluded, that only "exotic" collective states would spoil the *numerical* convergence of Kubo's expansion for the grand potential, as long as the occupation probabilities (18) are reasonably small.

4.2. The virial series

Going back from probability to fugacity,

$$p = \frac{f}{1+f}, \tag{41}$$

and expanding to second order in f, in the classical limit one obtains

$$\Psi(1) + \Psi(2) = Lf + \left(-\frac{L}{2} + \sum_{1 \le i < j \le L} g_{ij} \right) f^2 + \mathcal{O}(f^3), \tag{42}$$

and since it is consistent to write $g_{ii} = -1$, the term multiplying f^2 may be written as the *unrestricted* double sum

$$\frac{1}{2} \sum_{i=1}^{L} \sum_{j=1}^{L} g_{ij} \to \frac{L}{2} \sum_{\Delta} g(\Delta) \equiv LB_1, \tag{43}$$

where it has been used that $g_{ij} \equiv g(|\boldsymbol{r}_i - \boldsymbol{r}_j|)$ depends only on the differences, and the arrow means the large-volume limit. It is obvious that B_1 is just the usual "second virial coefficient", except that it is dimensionless; this can be repaired by transferring the factor a^3 from the fugacity (36), and putting $La^3 = V$.

The third-order result is recovered along the same lines. All that remains of $\Psi(3)$ (35) to third order in the fugacity is $f^3 \sum S_{ijk}$, where, in the classical limit (inserting Eq. (40) into Eq. (30)),

$$S_{ijk} = g_{ij} g_{ik} g_{jk} + g_{ij} g_{ik} + g_{ij} g_{jk} + g_{ik} g_{jk}, \tag{44}$$

from which it is clear that the subtracted matrix elements $S_{i(n)}$ are the generating functions of labelled graphs with n vertices. Adding the terms in f^3 from $\Psi(1)$ and $\Psi(2)$, one gets

$$\left(\frac{L}{3} - 2\sum_{i<j} g_{ij} + \sum_{i<j<k} S_{ijk}\right) f^3. \tag{45}$$

To pass from restricted to unrestricted summation, use

$$3!\sum_{i<j<k} = \sum_{i\neq j\neq k} = \sum_{i,j,k} - \sum_{i,j=k} - \sum_{i=k,j} - \sum_{i=j,k} + 2\sum_{i=j=k}, \tag{46}$$

which may be derived by inverting successive expressions of the type

$$\sum_{i,j} = \sum_{i\neq j} + \sum_{i=j}. \tag{47}$$

Inserting Eqs. (44) and (46) into Eq. (45), it becomes

$$\frac{1}{6}\sum_{i,j,k} S_{ijk} f^3, \tag{48}$$

so that, just as in second order, all that the lower terms in Eq. (45) do is to remove restrictions on the sum in the highest one. In the large-volume limit, the first term in Eq. (44) gives

$$\frac{1}{6}\sum_{i,j,k} g_{ij}g_{ik}g_{jk} \to \frac{L}{6}\sum_{\Delta_1,\Delta_2} g(\Delta_1)g(\Delta_2)g(|\Delta_1 - \Delta_2|) \equiv LB_2, \tag{49}$$

and the remaining three give a total contribution

$$\frac{1}{2}\sum_{i,j,k} g_{ij}g_{ik} \to \frac{L}{2}\sum_{\Delta_1}\sum_{\Delta_2} g(\Delta_1)g(\Delta_2) = 2LB_1^2, \tag{50}$$

because the g's depend only on the differences, so one finds, finally,

$$\Psi = Lf + LB_1 f^2 + L(2B_1^2 + B_2)f^3 + \mathcal{O}(f^4), \tag{51}$$

which is the virial series to third order, in one notation [2].

The statement that the interacting problem is "reduced to quadrature" in the classical limit is interpreted here, that the coefficient of f^n in Ψ becomes an *unrestricted* sum, namely

$$\frac{1}{n!}\sum_{i_1,\ldots,i_n} \{\text{coefficient of } f^n \text{ in } \psi[i(n)]\}. \tag{52}$$

From this point of view, in the classical limit there appears a "conspiracy of terms" which removes quantum restrictions from the sums in Kubo's formula.

5. *Discussion*

This article gives a combinatorial interpretation of Kubo's cluster cumulant expansion, as a binomial transform of the grand potential. It primarily explores the pedagogical implications of having such a brief, but formal, derivation. The idea is to develop a self-contained, general point of departure to treat problems which require a formulation in real space, assuming only undergraduate prior knowledge. In particular, it is found there is no need to introduce cumulants explicitly at this level, in order to produce operative size-consistent expressions. This was demonstrated in detail for the classical limit.

In this approach, the distinction is kept between rearranging the series for Ψ, which (effectively) resums different infinite classes of terms, and generating the terms themselves, which requires the evaluation of matrix elements. It becomes clear in principle, how different choices of expansion parameter (probability, fugacity, coupling constant, ...) necessarily yield different rules for which terms appear, and how either quantum entanglement, or topological restrictions from the Hamiltonian, both of which change the form of (40), can spoil the "conspiracy" by which the classical expressions simplify.

The basic operational problem in quantum mechanics is to replace sums by integrals, or, in more sophisticated language, to pass from functions defined on sets (of quantum states) to functions of real numbers (parameters of the Hamiltonian). This is trivial with unrestricted sums, which is the combinatorial content of the simplification in the classical limit. The converse is formally the most difficult problem of strong correlations: when dynamical effects restrict a (multiple) sum to an "arbitrary" subset of discrete states, *i.e.* such that no ordering can be defined on it, there is no controlled way to express the sum as an integral in the large- volume limit. Otherwise, the standard way to obtain integrals is to introduce ordering by the time variable, leading to Dyson diagrams, or along the fermion line, leading to Feynman diagrams. The relationship between the cluster cumulant expansion and the diagrammatic approach has been discussed by Dunn [14], for the case of a particular self-energy. He showed that cutting the expansion off at m-th order amounted to calculating all diagrams with at most m-fold momentum integrals exactly, and all others approximately (the logarithms in $\Psi(m)$ necessarily generate diagrams to infinite order).

On the other hand, rewriting Kubo's formula as a probability expansion, equation (32), shows that convergence can be expected even when one does not have the complete solution of the problem. All that is required is that the probabilities in the chosen basis are bounded away from unity (and zero). The prototype for this is precisely the real-space basis, because position states are never stationary, due to the uncertainty principle.

Two other properties of the cluster expansion are readily obtained. First, it was stressed by Kubo [4] that all described operations remain exactly correct even if the "matrix elements" are not c-numbers, and the exponential functions are replaced by various rules. This follows directly from the fact that the binomial inversion is a formal identity. For example, when the probability expansion can be written in Fock-space operators, the Pauli principle is exactly preserved, even if one stops at the first order [9, 10]. Second, if matrix elements are used, the Pauli principle cannot be satisfied for the whole assembly of particles, as soon as the expansion is cut off. Its form then indicates that antisymmetrization is taken into account by an "inclusion-exclusion" procedure; for instance, the same two-particle terms, involving S_{ij}, appear with opposite sign in $\Psi(2)$ and $\Psi(3)$ (Eq. (35)). So the cluster cumulant series is expected to alternate, whenever many-body correlations are important. This can be striking in practice [15].

To conclude, the second and fourth sections of this article give a compact and hopefully useful introduction to an established general treatment of correlations in real space. In the third section, it is shown that the parameters controlling convergence can be interpreted as probabilities, and their associated subtracted matrix elements appear as the basic building blocks of more elaborate calculations.

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Appendix

The probability expansion (32) is formally the same for fermions and bosons, only the definitions of the various quantities change. For bosons, the occupation probabilities are

$$p_i = \frac{\sum\limits_{k=1}^{\infty} \mathrm{e}^{k\beta\mu} \left\langle i^k \right| \mathrm{e}^{-\beta H} \left| i^k \right\rangle}{\sum\limits_{k=0}^{\infty} \mathrm{e}^{k\beta\mu} \left\langle i^k \right| \mathrm{e}^{-\beta H} \left| i^k \right\rangle}, \tag{53}$$

where i^k means, i-th state, occupied by k bosons. In the non-interacting case, this reduces to the familiar $\mathrm{e}^{\beta(\mu-\varepsilon_i)}$, justifying the use of the term "occupation probability".

The normalized matrix elements (31) become, for bosons,

$$\widetilde{U}_{j(n)} = \frac{\sum\limits_{k_1,\ldots,k_n=1}^{\infty} \mathrm{e}^{(k_1+\ldots+k_n)\beta\mu} \left\langle j_1^{k_1} \ldots j_n^{k_n} \right| \mathrm{e}^{-\beta H} \left| j_1^{k_1} \ldots j_n^{k_n} \right\rangle}{\left(\sum\limits_{k_1=1}^{\infty} \mathrm{e}^{k_1\beta\mu} \left\langle j_1^{k_1} \right| \mathrm{e}^{-\beta H} \left| j_1^{k_1} \right\rangle \right) \cdots \left(\sum\limits_{k_n=1}^{\infty} \mathrm{e}^{k_n\beta\mu} \left\langle j_n^{k_n} \right| \mathrm{e}^{-\beta H} \left| j_n^{k_n} \right\rangle \right)}. \tag{54}$$

To the lowest order in the activity, these expressions are of course equal to the fermion ones, which accounts for the classical limit. Note, finally, that the denominator of $\widetilde{U}_{j(n)}$ contains the "occupation numbers"

$$n_i = \sum_{k=1}^{\infty} \mathrm{e}^{k\beta\mu} \left\langle i^k \right| \mathrm{e}^{-\beta H} \left| i^k \right\rangle , \tag{55}$$

in terms of which $p_i = n_i/(n_i + 1)$, as with non-interacting bosons.

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NOVI IZVOD RAZVOJA U GROZDOVE KUMULANATA

Kubov razvoj u kumulantne grozdove se izvede pozivanjem samo na elementarna svojstva skupova i binomnih koeficijenata. Pokaže se da je on upravo binomni transformat velekanonskog potencijala. Ekstenzivnost se dokaže bez pozivanja na kumulante. Razvoj po aktivnosti se preuredi kombinatoričkom inverzijom u razvoj po vjerojatnostima zaposjednuća, koje izravno upravljaju konvergencijom. Klasični virijalni razvoj se izvede do trećeg reda kao primjer.