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Superposition Approximations from a Variation Principle

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A variation principle is introduced involving the n-particle molecular distribution function (where $1 \leq n \leq N$) for a fluid containing N molecules. An integral involving any approximate n-particle distribution function proves to define a *least* upper bound to the true system free energy. This integral can, therefore, be minimized with respect to the form of a trial distribution function to provide a best estimate to the exact distribution function. When no other constraints, save the requirement of normalization, are applied to the trial function, the extremum corresponds to the exact function. Using this variation principle, it is possible to demonstrate that the optimum triplet superposition approximation is the Krikwood approximation, and that the optimum quadruplet approximation is the form suggested by Fisher and Kopeliovich. Furthermore, all higher-order optimum superposition approximations are specified.

KEY WORDS: Correlation functions; hierarchy; upper bound; Helmholtz free energy.

1. INTRODUCTION

If we denote by P_{123} the triplet *specific* distribution function (Hill,⁽¹⁾ pp. 181–182), then the Kirkwood superposition approximation assumes the form

$$P_{123} = V^3 P_{12} P_{23} P_{13} \tag{1}$$

in which V is the volume of the system and P_{12}, P_{23}, and P_{13} are pair specific distribution functions on particles 1 and 2, 2 and 3, and 1 and 3, respectively.

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There are necessary relationships between P_{123} and any of the pair functions, of the type

$$P_{12} = \int_V P_{123}\, d\mathbf{r}_3 \tag{2}$$

where $d\mathbf{r}_3$ is the volume element of the coordinates of particle 3. Indeed, this is the meaning of the pair specific distribution functions appearing in Eq. (1).

On the other hand, we might replace Eq. (1) with

$$P_{123} = \phi_{12}\phi_{23}\phi_{13} \tag{3}$$

in which the ϕ's, which also depend upon pairs of the indicated coordinates, *need not* be pair specific distribution functions. Then, according to Eq. (2),

$$P_{12} = \phi_{12} \int_V \phi_{23}\phi_{13}\, d\mathbf{r}_3 \tag{4}$$

Thus, Eqs. (3) and (4) could be substituted into the hierarchial integrodifferential equation (Hill,(1) Sections 32 and 33), involving the triplet and pair distribution functions, to provide a determinate equation for the ϕ's.

Equation (3) is a more general superposition approximation, and one wonders whether ϕ functions can be found which would make it superior to Eq. (1). Even more important, if we advance to consideration of superposition for the higher-order distribution functions, many possible products and quotients of lower-order distribution functions are possible, and again we have the problem of the optimum choice.

In what follows, we shall formulate a variation principle in terms of which such choices can be made (relative, of course, to the variation principle). It turns out that for the triplet function, Eq. (3), is indeed the optimum choice. Furthermore, the optimum choice for any higher-order function is specified. Beyond the triplet, the ϕ's are *not* simply lower-order distribution functions.

2. THE VARIATION PRINCIPLE

Consider a fluid system containing N of one kind of molecule, in which n specified molecules are fixed at the points $\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n$. The configurational partition function for this system will be denoted by $Z(\{\mathbf{r}_n\})$, while the configurational partition function without the n particles constrained to fixed positions is Z. Clearly, the specific n-tuplet distribution function on the n particles will be

$$P(\{\mathbf{r}_n\}) = Z(\{\mathbf{r}_n\})/Z \tag{5}$$

The Helmholtz free energy with the n particles fixed is

$$A(\{\mathbf{r}_n\}) = -kT \ln Z(\{\mathbf{r}_n\}) \tag{6}$$

while in the case they are not fixed, it is

$$A = -kT \ln Z \tag{7}$$

Substitution of Eqs. (6) and (7) into Eq. (5) yields

$$A(\{\mathbf{r}_n\}) = -kT \ln P(\{\mathbf{r}_n\}) + A \tag{8}$$

Now, suppose we have some approximation, $P_*(\{\mathbf{r}_n\})$, to $P(\{\mathbf{r}_n\})$. Then we define the free energy corresponding to this approximate distribution[2]:

$$\begin{aligned} A_* = &\int \cdots \int_V P_*(\{\mathbf{r}_n\})\, A(\{\mathbf{r}_n\})\, d\{\mathbf{r}_n\} \\ &+ kT \int \cdots \int_V P_*(\{\mathbf{r}_n\}) \ln P_*(\{\mathbf{r}_n\})\, d\{\mathbf{r}_n\} \end{aligned} \tag{9}$$

where the last term is additional entropy due to the multiplicity of configurations available to the n particles and $d\{\mathbf{r}_n\}$ stands for the volume element $d\mathbf{r}_1\, d\mathbf{r}_2 \cdots d\mathbf{r}_n$.[3] If we substitute Eq. (8) into (9) and require the normalization

$$\int \cdots \int_V P_*(\{\mathbf{r}_n\})\, d\{\mathbf{r}_n\} = 1 \tag{10}$$

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[2] A word about the interpretation of A_* is in order. Suppose a powerful benefactor supplied us with partial information concerning the thermodynamic properties of the system by providing the *exact* values of $A(\{\mathbf{r}_n\})$ for all $\{\mathbf{r}_n\}$. We wish to make maximum use of this partial information to compute A, the full free energy of the system. To this end, we guess at the function $P(\{\mathbf{r}_n\})$ in the hope that some further principle will be available to assist with the process of guessing. We denote the guessed value of $P(\{\mathbf{r}_n\})$ by $P_*(\{\mathbf{r}_n\})$. Now, since, within the approximation, the fraction of time which the system spends in $d\{\mathbf{r}_n\}$ is given by $P_*(\{\mathbf{r}_n\})\, d\{\mathbf{r}_n\}$, the contribution to A from this configuration might, at first, be taken as $P_*(\{\mathbf{r}_n\}) A(\{\mathbf{r}_n\})\, d\{\mathbf{r}_n\}$, the integrand of the first integral in Eq. (9). The first integral itself is the sum of contributions from the whole of configuration space.

However, the full value of A must also include entropy contributions arising from the uncertainty as to which configuration, $\{\mathbf{r}_n\}$, the system is actually in. The second term accounts for this.

Thus, A_* represents the best estimate of A possible, based on the information in hand concerning $A(\{\mathbf{r}_n\})$ and a particular guess at $P(\{\mathbf{r}_n\})$, $P_*(\{\mathbf{r}_n\})$. In this sense, it represents the maximum immediate use of the information bequeathed by the benefactor. We improve this use by refining $P_*(\{\mathbf{r}_n\})$ through application of the variation principle.

[3] The origin of this last term is perhaps seen more clearly if we consider the exact distribution function $P(\{\mathbf{r}_n\})$ rather than $P_*(\{\mathbf{r}_n\})$, and compute

$$\Delta = A - \int \cdots \int_V P(\{\mathbf{r}_n\}) A(\{\mathbf{r}_n\})\, d\{\mathbf{r}_n\}$$

Substituting in this equation from Eqs. (5) and (6), we obtain

$$\Delta = kT \int \cdots \int_V P(\{\mathbf{r}_n\}) \ln P(\{\mathbf{r}_n\})\, d\{\mathbf{r}_n\}$$

we obtain

$$A_* = A + kT \int \cdots \int_V P_*(\{\mathbf{r}_n\}) \ln[P_*(\{\mathbf{r}_n\})/P(\{\mathbf{r}_n\})]\, d\{\mathbf{r}_n\} \tag{11}$$

Now, since in addition to Eq. (10) we have

$$\int \cdots \int_V P(\{\mathbf{r}_n\})\, d\{\mathbf{r}_n\} = 1 \tag{12}$$

it is easy to show (properties of convexity) that the integral at the right must always be positive, no matter the choice of $P_*(\{\mathbf{r}_n\})$. *Thus,* A^* *forms an upper bound to the true free energy* A. When $P_*(\{\mathbf{r}_n\}) = P(\{\mathbf{r}_n\})$, we note that $A_* = A$, as is expected.

Clearly, if $P_*(\{\mathbf{r}_n\})$ is varied subject to no other constraint than that of normalization, Eq. (1), the function which minimizes A_* will be the true distribution function $P(\{\mathbf{r}_n\})$. If other constraints are present, then the minimum of A_* with respect to the variation of $P_*(\{\mathbf{r}_n\})$ will give a best approximation *subject to those constraints*, in the sense of a least upper bound to the true free energy A. This is the variation principle we wish to use.

3. THE OPTIMUM SUPERPOSITION APPROXIMATION

We introduce a superposition approximation in the sense of Eq. (3). Thus, we approximate $P(\{\mathbf{r}_n\})$ by

$$P_*(\{\mathbf{r}_n\}) = \phi_n \phi_{n-1} \cdots \phi_1 \tag{13}$$

where, for example, ϕ_n is a function of all coordinates of the group of n particles save those of particle n. Thus, in the special case of the triplet function in Eq. (3), we could write, using this notation,

$$\phi_{12} = \phi_3, \qquad \phi_{23} = \phi_1, \qquad \phi_{13} = \phi_2 \tag{14}$$

Substitution of Eq. (13) into (11) yields

$$\begin{aligned} A_* = A &+ kT \int \cdots \int_V \phi_n \phi_{n-1} \cdots \phi_1 \ln \phi_n \, d\{\mathbf{r}_n\} \\ &+ kT \int \cdots \int_V \phi_n \phi_{n-1} \cdots \phi_1 \ln \phi_{n-1} \, d\{\mathbf{r}_n\} \\ &+ \cdots + kT \int \cdots \int_V \phi_n \phi_{n-1} \cdots \phi_1 \ln \phi_1 \, d\{\mathbf{r}_n\} \\ &- kT \int \cdots \int_V \phi_n \phi_{n-1} \cdots \phi_1 \ln P(\{\mathbf{r}_n\})\, d\{\mathbf{r}_n\} \end{aligned} \tag{15}$$

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which is just the last term in question for the case that $P^*(\{\mathbf{r}_n\})$ is exact. From this, it is clear that the integral in the first expression for Δ, even though it averages $A(\{\mathbf{r}_n\})$ properly over configuration space, misses supplying the value of A because it omits certain entropic contributions.

The constraints in this problem are the special decomposed form of $P_*(\{\mathbf{r}_n\})$ and normalization of Eq. (10).

Taking the variation of A_* with respect to the form of the ϕ's, and subject to Eq. (10), we find, upon setting $\delta A_* = 0$, that the coefficient of $\delta\phi_n$ is given by

$$(kT + \lambda) \int_V [P_*(\{\mathbf{r}_n\})/\phi_n]\, d\mathbf{r}_n$$
$$+ kT \int_V [P_*(\{\mathbf{r}_n\})/\phi_n] \ln[P_*(\{\mathbf{r}_n\})/P(\{\mathbf{r}_n\})]\, d\mathbf{r}_n = 0 \tag{16}$$

in which λ is an undetermined multiplier. By symmetry, since all the ϕ's are the same function of their arguments, the same equation holds for ϕ_{n-1}, etc. We need only investigate Eq. (16).

Recognizing that ϕ_n does not depend upon $\mathbf{r}_n$, and setting $kT + \lambda$ equal to the "constant" $kT\alpha$, we may rewrite Eq. (16) as

$$(\alpha + \ln \phi_n) \int_V [P_*(\{\mathbf{r}_n\})/\phi_n]\, d\mathbf{r}_n$$
$$= \int_V [P_*(\{\mathbf{r}_n\})/\phi_n] \ln[P(\{\mathbf{r}_n\})\, \phi_n/P_*(\{\mathbf{r}_n\})]\, d\mathbf{r}_n \tag{17}$$

Writing $\alpha = -\ln K$, this can be further rearranged to

$$\ln \frac{\phi_n}{K} = \frac{\int_V [P_*(\{\mathbf{r}_n\})/\phi_n] \ln[P(\{\mathbf{r}_n\})\, \phi_n/P_*(\{\mathbf{r}_n\})]\, d\mathbf{r}_n}{\int_V [P_*(\{\mathbf{r}_n\})/\phi_n]\, d\mathbf{r}_n} \tag{18}$$

Consider $P(\{\mathbf{r}_n\})$ in the case that the nth particle is far removed (beyond the range of correlation) from the remaining $n - 1$ particles. Under this condition, denote $P(\{\mathbf{r}_n\})$ by $\bar{P}(\{\mathbf{r}_n\})$. Denote $P_*(\{\mathbf{r}_n\})$, under the similar condition, by $\bar{P}_*(\{\mathbf{r}_n\})$. Clearly, both $\bar{P}(\{\mathbf{r}_n\})$ and $\bar{P}_*(\{\mathbf{r}_n\})$ depend only on the coordinates of the $n - 1$ particles other than the nth. In fact, we know that

$$\bar{P}(\{\mathbf{r}_n\}) = (1/V) P(\{\mathbf{r}_{n-1}\}) \tag{19}$$

Equation (18) can be further transformed. Thus, by adding and subtracting $\bar{P}(\{\mathbf{r}_n\})$ in the numerator behind the logarithm on the right and $\bar{P}_*(\{\mathbf{r}_n\})$ in the denominator, we obtain

$$\ln \frac{\phi_n}{K} = \biggl(\int_V \frac{P_*(\{\mathbf{r}_n\})}{\phi_n} \ln \frac{\phi_n \bar{P}(\{\mathbf{r}_n\})}{\bar{P}_*(\{\mathbf{r}_n\})}\, d\mathbf{r}_n$$
$$+ \int_V \frac{P_*(\{\mathbf{r}_n\})}{\phi_n} \ln \frac{1 + \{[P(\{\mathbf{r}_n\}) - \bar{P}(\{\mathbf{r}_n\})]/\bar{P}(\{\mathbf{r}_n\})\}}{1 + \{[P_*(\{\mathbf{r}_n\}) - \bar{P}_*(\{\mathbf{r}_n\})]/\bar{P}_*(\{\mathbf{r}_n\})\}}\, d\mathbf{r}_n \biggr)$$
$$\times \biggl(\int_V \frac{P_*(\{\mathbf{r}_n\})}{\phi_n}\, d\mathbf{r}_n \biggr)^{-1} \tag{20}$$

The quantity

$$\ln \frac{1 + \{[P(\{\mathbf{r}_n\}) - \bar{P}(\{\mathbf{r}_n\})]/\bar{P}(\{\mathbf{r}_n\})\}}{1 + \{[P_*(\{\mathbf{r}_n\}) - \bar{P}_*(\{\mathbf{r}_n\})]/\bar{P}_*(\{\mathbf{r}_n\})\}} \tag{21}$$

in Eq. (20) has about the range of the intermolecular potential since $P(\{\mathbf{r}_n\})$ and $P_*(\{\mathbf{r}_n\})$ go to $\bar{P}(\{\mathbf{r}_n\})$ and $\bar{P}_*(\{\mathbf{r}_n\})$, respectively, when $\mathbf{r}_n$ is outside the range of correlation of the remaining $n - 1$ particles. Thus, the second integral in the numerator on the right of Eq. (20) is of the order of

$$(v/V) \int_V [P_*(\{\mathbf{r}_n\})/\phi_n]\, d\mathbf{r}_n \tag{22}$$

where v is the "volume of correlation" and $v/V = O(1/N)$, where N is the total number of molecules in the fluid. In the first integral in the numerator on the right of Eq. (20), the quantity

$$\phi_n \bar{P}(\{\mathbf{r}_n\})/\bar{P}_*(\{\mathbf{r}_n\}) \tag{23}$$

is independent of $\mathbf{r}_n$ and may be moved in front of the integral. Thus, this term becomes

$$\ln[\phi_n \bar{P}(\{\mathbf{r}_n\})/\bar{P}_*(\{\mathbf{r}_n\})] \int_V [P_*(\{\mathbf{r}_n\})/\phi_n]\, d\mathbf{r}_n \tag{24}$$

Substituting Eqs. (22) and (24) into (20) yields

$$\ln \frac{\phi_n}{K} = \ln \frac{\phi_n \bar{P}(\{\mathbf{r}_n\})}{\bar{P}_*(\{\mathbf{r}_n\})} + \frac{v}{V} = \ln \frac{\phi_n \bar{P}(\{\mathbf{r}_n\})}{\bar{P}_*(\{\mathbf{r}_n\})} + O\left(\frac{1}{N}\right) \tag{25}$$

and since we consider the thermodynamic limit in which $N \to \infty$, this becomes

$$1/K = \bar{P}(\{\mathbf{r}_n\})/\bar{P}_*(\{\mathbf{r}_n\}) \tag{26}$$

Using Eq. (19), we have

$$\bar{P}_*(\{\mathbf{r}_n\}) = (K/V) P(\{\mathbf{r}_{n-1}\}) \tag{27}$$

We can add and substract $P_*(\{\mathbf{r}_n\})$ on the left side of Eq. (27) to obtain

$$P_*(\{\mathbf{r}_n\}) + [\bar{P}_*(\{\mathbf{r}_n\}) - P_*(\{\mathbf{r}_n\})] = (K/V) P(\{\mathbf{r}_{n-1}\}) \tag{28}$$

Integrating both sides of this equation over the whole space of the n particles, we get

$$\begin{aligned} \int \cdots \int_V P_*(\{\mathbf{r}_n\})\, d\{\mathbf{r}_n\} + \int \cdots \int_V [\bar{P}_*(\{\mathbf{r}_n\}) - P_*(\{\mathbf{r}_n\})]\, d\{\mathbf{r}_n\} \\ = (K/V) \int \cdots \int_V P(\{\mathbf{r}_{n-1}\})\, d\{\mathbf{r}_n\} \end{aligned} \tag{29}$$

By Eq. (10), the first integral on the left is unity, and by the normalization of $P(\{\mathbf{r}_{n-1}\})$, the integral on the right is also unity. The second integral on the left is again of order $v/V = O(1/N)$ and can be ignored. Thus, Eq. (29) becomes[4]

$$1 = K \tag{30}$$

so that K is unity. With this, Eq. (26) becomes

$$1 = [P(\{\mathbf{r}_{n-1}\})/V]/\phi_n \bar{\phi}_{n-1} \bar{\phi}_{n-2} \cdots \bar{\phi}_1 \tag{31}$$

It is not necessary to have a bar over ϕ_n, because it does not depend on the coordinates of the nth particle. Thus, we achieve our final result:

$$\phi_n = [P(\{\mathbf{r}_{n-1}\})/V]/\bar{\phi}_{n-1} \bar{\phi}_{n-2} \cdots \bar{\phi}_1 \tag{32}$$

4. INVESTIGATION OF SPECIAL CASES

We examine the conventional triplet case first. In this case, Eq. (32) becomes

$$\phi_{12} = (P_{12}/V)/\bar{\phi}_{23}\bar{\phi}_{13} \tag{33}$$

If we move particle 2 far from particle 1, $\bar{\phi}_{23}$ and $\bar{\phi}_{13}$ are unaffected, since particle 3 is already far from both particles 1 and 2 in each of them, respectively. Furthermore, it is well known that

$$\bar{P}_{12} = 1/V^2 \tag{34}$$

Thus, by moving particles 1 and 2 in Eq. (33) far apart, we obtain

$$\bar{\phi}_{12} = (1/V^3)/\bar{\phi}_{23}\bar{\phi}_{13} \tag{35}$$

or

$$\bar{\phi}_{12}\bar{\phi}_{23}\bar{\phi}_{13} = 1/V^3 \tag{36}$$

Since $\bar{\phi}_{12}$, $\bar{\phi}_{23}$, and $\bar{\phi}_{13}$ must be equal by symmetry, this equation implies that

$$\bar{\phi}_{23} = \bar{\phi}_{13} = 1/V \tag{37}$$

Substitution in Eq. (33) yields

$$\phi_{12} = VP_{12} \tag{38}$$

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[4] Since $K = 1$ implies $\alpha = 0$, this would appear to make *each* term in Eq. (16) vanish independently. This is a nonsense result which stems from the fact that Eq. (30) is approximate to the extent that terms of $O(1/N)$ have been neglected.

so that

$$P^*_{123} = \phi_{12}\phi_{23}\phi_{13} = V^3 P_{12} P_{23} P_{13} \tag{39}$$

which is the standard Kirkwood superposition approximation.

Thus, the optimum values of ϕ_{12}, etc., relative to the variation principle, are indeed given by the pair specific distribution functions P_{12}, etc. These are the values of the ϕ's which provide the most nearly correct system free energy subject to the constraint of superposition. Thus, the Kirkwood superposition approximation cannot be improved on by any other pairwise decomposition of the triplet function.

It is of interest to see what Eq. (32) requires of the superposition approximation to the quadruplet distribution function. In this case,

$$P^*_{1234} = \phi_{123}\phi_{124}\phi_{134}\phi_{234} \tag{40}$$

and by Eq. (32),

$$\phi_{123} = (P_{123}/V)/\bar{\phi}_{12,4}\bar{\phi}_{13,4}\bar{\phi}_{23,4} \tag{41}$$

The bars in Eq. (41) indicate that particle 4 is far from the others, a fact which we have singled out by prefacing the subscript 4 by a comma. If we now move particle 3 far from the others, we obtain from Eq. (41)

$$\bar{\phi}_{12,3} = (P_{12}/V^2)/\bar{\phi}_{12,4}\bar{\bar{\phi}}_{1,34}\bar{\bar{\phi}}_{2,34} \tag{42}$$

where the double bar indicates that two particles have been moved beyond the correlation distance, and the two particles moved are again prefaced by a comma. In the double-barred symbols, we have exhausted all possibilities of moving particles out of range—all three particles have been moved out of range of each other. Thus, when we now move particle 2 out of range in Eq. (42), we obtain

$$\bar{\bar{\phi}}_{1,23} = (1/V^4)/\bar{\bar{\phi}}_{1,24}\bar{\bar{\phi}}_{1,34}\bar{\bar{\phi}}_{2,34} \tag{43}$$

nothing happening to the last two factors in the denominator of Eq. (42).

Rearranging Eq. (43), we get

$$\bar{\bar{\phi}}_{1,23}\bar{\bar{\phi}}_{1,24}\bar{\bar{\phi}}_{1,34}\bar{\bar{\phi}}_{2.34} = 1/V^4 \tag{44}$$

and since all the factors on the left must be identical by symmetry,

$$\bar{\bar{\phi}}_{1.23} = \bar{\bar{\phi}}_{1.24} = \bar{\bar{\phi}}_{1.34} = \bar{\bar{\phi}}_{2,34} = 1/V \tag{45}$$

Substitution of Eq. (45) into (42) gives

$$\bar{\phi}_{12.3}\bar{\phi}_{12.4} = P_{12} \tag{46}$$

and, again, by symmetry,

$$\bar{\phi}_{12,3} = \bar{\phi}_{12,4} = P_{12}^{1/2} \tag{47}$$

Substitution of Eq. (47) and its analogs into Eq. (41) gives

$$\phi_{123} = (1/V)P_{123}/(P_{12}P_{23}P_{13})^{1/2} \tag{48}$$

Substituting this, and the similar equations for ϕ_{124}, ϕ_{134}, and ϕ_{234}, into Eq. (40) yields

$$P_{1234}^{*} = (1/V^4)\, P_{123}P_{124}P_{134}P_{234}/P_{12}P_{23}P_{13}P_{14}P_{24}P_{34} \tag{49}$$

This is the superposition function which yields a system free energy closest to the true free energy in the case of the quadruplet distribution. It is interesting that Eq. (49) agrees with the form proposed by Fisher and Kopeliovich⁽²⁾ through consideration of the limit of low density, and a process of educated guessing.

By a more lengthy, but exactly similar, procedure, we can find the approximation to the fifth-order distribution function. This proves to be

$$P_{12345}^{*} = \frac{V^5(P_{1234}P_{1235}P_{1245}P_{1345}P_{2345})(P_{12}P_{13}P_{23}P_{14}P_{24}P_{31}P_{15}P_{25}P_{35})}{P_{123}P_{124}P_{134}P_{125}P_{135}P_{145}P_{235}P_{245}P_{345}} \tag{50}$$

The typical ϕ function in this case is

$$\phi_{1234} = VP_{1234}(P_{12}P_{13}P_{25}P_{14}P_{24}P_{34})^{1/3}/(P_{123}P_{124}P_{134}P_{234})^{1/2} \tag{51}$$

Clearly, Eq. (32) is capable of dealing with the approximation at any order.

It is to be noted especially [see Eqs. (33), (48), and (51)] that the ϕ's, beyond the pair function ϕ_{12}, are not simply the corresponding specific distribution functions, but, rather, far more complicated combinations of distribution functions of the same and lower orders. They are, however, well-defined functions of distribution functions and are uniquely specified by the variation principle.

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