Ordering principle for cluster expansions in the theory of quantum fluids, dense gases, and simple classical liquids

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A study is made of a series-expansion procedure which gives the leading terms of the n-particle distribution function $p^{(n)}(1,2,\ldots,n)$ as explicit functionals in the radial distribution function $g(r)$. The development of the series is based on the cluster-expansion formalism applied to the Abe form for $p^{(n)}$ expressed as a product of the generalized Kirkwood superposition approximation $p_K^{(n)}$ and a correction factor $\exp[A^{(n)}(1,2,\ldots,n)]$. An ordering parameter μ is introduced to determine $A^{(n)}$ and $p^{(n)}$ in the form of infinite power series in μ, and the postulate of minimal complexity is employed to eliminate an infinite number of possible classes of solutions in a sequential relation connecting $A^{(n-1)}$ and $A^{(n)}$. Derivation of the series for $p^{(n)}$ and many other algebraic manipulations involving a large number of cluster integrals are greatly simplified by the use of a scheme which groups together all cluster terms having, in a certain way, the same source term. In particular, the scheme is useful in demonstrating that the nature of the series structure of $p^{(3)}$ is such that its three-point Fourier transform $S^{(3)}(\vec{k}_1,\vec{k}_2,\vec{k}_3)$ has as a factor the product of the three liquid-structure functions $S(k_1)S(k_2)S(k_3)$. The results obtained to order μ^4 for $A^{(3)}$, $p^{(3)}$, and $S^{(3)}$ agree with those derived earlier in a more straightforward but tedious approach. The result for $p^{(4)}$ shows that the convolution approximation $p_c^{(4)}$, which contains μ^3 terms, must be supplemented by a correction of $O(\mu^3)$ in order to be accurate through third order. The μ-expansion approach is also examined for the cluster expansion of the correlation function in the Bijl-Dingle-Jastrow description of a many-boson system, and then compared with the number-density expansion formula by using the Gaussian model for $g(r)-1$ to evaluate cluster integrals. A testing procedure based on the requirement $p^{(3)}(1,2,2)=0$ is developed to study accuracy of the μ-ordered approximations for $p^{(3)}$. Numerical results obtained to orders μ^2, μ^3, and μ^4 with the Gaussian model indicate substantial improvements with each increase in the order of truncation in the power series of $p^{(3)}$. A brief discussion is presented concerning the asymptotic behavior of $g(r)$ in the context of equilibrium statistical mechanics.

I. INTRODUCTION

We are concerned primarily with a microscopic description of liquid ^{4}He under realistic conditions of density and strength of interaction.[1] The theoretical model consists of N interacting bosons confined to a cubical box of volume Ω. A normalized ground-state wave function

$$\Psi_0(\vec{r}_1,\vec{r}_2,\ldots,\vec{r}_N)\equiv\Psi_0(1,2,\ldots,N)\equiv\Psi_0$$

is generated by the Schrödinger equation and the conventional periodic boundary condition.

The n-particle distribution functions for the ground state are defined by

$$p^{(n)}(1,2,\ldots,n)=N(N-1)\cdots(N-n+1)\times\int\Psi_0^2 d\vec{r}_{n+1,n+2,\ldots,N}\,. \tag{1}$$

Successive distribution functions are connected by the limiting relation

$$p^{(n)}(1,2,\ldots,n)=\rho p^{(n-1)}(1,2,\ldots,n-1)\times[1+O(1/N)] \tag{2}$$

for $r_{nj}>>\rho^{-1/3}$, $j=1,2,\ldots,n-1$, and by the sequential statement

$$p^{(n-1)}(1,2,\ldots,n-1)=\frac{1}{N-n+1}\int p^{(n)}(1,2,\ldots,n)d\vec{r}_n\,. \tag{3}$$

In Eq. (2), $\rho=N/\Omega$ is the number density. In this context the basic quantities are the radial distribution function $g(r)$ and the liquid-structure function $S(k)$[1]:

$$p^{(2)}(1,2)=\rho^2 g(r_{12})=N(N-1)\int \Psi_0^2 d\vec{r}_{34\cdots N}\ , \tag{4a}$$

$$S(k)=N^{-1}\int \Psi_0^2 |\rho_{\vec{k}}|^2 d\vec{r}_{12\cdots N},\quad k\neq 0$$
$$=1+\rho\int e^{i\vec{k}\cdot\vec{r}}[g(r)-1]d\vec{r}\ , \tag{4b}$$

$$g(r)=1+\frac{1}{(2\pi)^3\rho}\int e^{-i\vec{k}\cdot\vec{r}}[S(k)-1]d\vec{k}\ , \tag{4c}$$

$$\rho_{\vec{k}}=\int\sum_{j=1}^{N}\delta(\vec{r}-\vec{r}_j)e^{i\vec{k}\cdot\vec{r}}d\vec{r}$$
$$=\sum_{j=1}^{N}e^{i\vec{k}\cdot\vec{r}_j}\ , \tag{4d}$$

$$\lim_{k\to 0}\frac{S(k)}{k}=\frac{\hbar}{2mc},\quad \text{hence}\quad S(0)=0\ , \tag{4e}$$

$$\lim_{r\to\infty} r^4[g(r)-1]=-\frac{\hbar}{2\pi^2 m\rho c}\ , \tag{4f}$$

$$S(\infty)=g(\infty)=1\ . \tag{4g}$$

Here c is the velocity of sound at $T=0$ in the limit of long wavelengths ($k<<2\pi\rho^{1/3}$). The notation

$$h(r)\equiv g(r)-1\ , \tag{4h}$$

$$F(k)\equiv S(k)-1\ , \tag{4i}$$

$$p^{(n)}\equiv p^{(n)}(1,2,\ldots,n)\ , \tag{4j}$$

will be used throughout this paper to simplify the typography.

The distribution functions occur in the perturbation formalisms developed to compute the energy of excitations in the fluid and the ground-state energy of the system. Also the boson distribution functions occur in procedures for producing trial functions to describe the low states of fermion systems. Approximations available for numerical estimates include the generalized Kirkwood superposition form

$$p_K^{(n)}(1,2,\ldots,n)=\rho^n\prod_{(1\le i<j\le n)}g(r_{ij}) \tag{5}$$

and the convolution form[1,2] $p_c^{(n)}(1,2,\ldots,n)$. These forms are *complete* functionals in the radial distribution function $g(r)$ [or the liquid-structure function $S(k)$]. The term "complete" signifies that all quantities entering into the formulas for $p_K^{(n)}$ and $p_c^{(n)}$ are expressed completely and explicitly as functionals in $g(r)$ [or $S(k)$]. A third approximate form having the character of a complete functional was first introduced by Abe[3] and later generalized by Stell[4,5] in the context of the classical statistical mechanics of an imperfect gas. Following Abe and Stell we write

$$p^{(n)}(1,2,\ldots,n)=p_K^{(n)}(1,2,\ldots,n)\exp[A^{(n)}(1,2,\ldots,n)]$$
$$=\rho p^{(n-1)}(1,2,\ldots,n-1)\left[\prod_{j=1}^{n-1}g(r_{jn})\right]\exp[A^{(n)}(1,2,\ldots,n)-A^{(n-1)}(1,2,\ldots,n)]\ . \tag{6}$$

In the second line the occurrence of $p^{(n-1)}$ as a factor points to the use of the sequential relation to determine $A^{(n)}$ and $A^{(n-1)}$. Abe expressed $A^{(3)}$ as a *formal* cluster expansion in powers of ρ with the coefficients in terms of the radial distribution function and gave explicit forms for the first- and second-order terms in ρ. Generalization of such expressions for $p^{(n)}$ as functionals in g was later given by Stell[4,5] to all orders for $n\geq 3$. The qualification "formal" refers to the fact that the coefficient of ρ^m is a functional in g and hence a function of ρ. It is, however, clear and universally recognized that number density ρ is not a suitable small parameter in the theory of liquid helium under realistic conditions. The formulation and development of an alternative ordering and grouping of the cluster expansion for $p^{(n)}$ is the essential objective of this paper.

Clues to more suitable expansion procedures can be found in discussions by Woo,[6] Lai, Sim, and Woo,[7] Campbell,[8] Lee and Feenberg,[9] and Lee.[10] References 6 and 7 are concerned primarily with the grouping of diagrams in the perturbation series for an energy eigenvalue (the ground-state energy of liquid ^{3}He in Ref. 6 and the elementary excitation energy of liquid ^{4}He in Ref. 7). In Ref. 7 the ordering criterion is the number of independent loops formed by free-phonon lines in a perturbation diagram. References 8–10 develop simple diagrammatic cluster formulas for distribution functions. Chung[11] is responsible for a first definitive statement in which the essential point of the latter program is made completely explicit using the number of independent loops in a cluster diagram as the ordering criterion.[12]

In all these studies the elementary bond in a

cluster is $h(r_{ij})\equiv g(r_{ij})-1$. The cluster formalism of classical statistical mechanics is based on the elementary bond $\exp[-\beta v(r_{ij})]-1$; however, the utility of cluster expansions using $h(r_{ij})$ as the bond is already recognized in the context of classical statistical mechanics.[3-5,13]

We choose to develop the procedure in terms of a formal ordering parameter μ introduced by the substitutions

$$h(r)\rightarrow\mu h(r)\ , \tag{7a}$$

$$F(k)\rightarrow F(k)\ , \tag{7b}$$

$$\Omega\rightarrow\mu\Omega\ , \tag{7c}$$

$$\rho\rightarrow\rho/\mu\ . \tag{7d}$$

The point of associating μ with Ω and ρ^{-1} as well as with $h(r)$ is to secure the invariance of the integrals

$$\rho\int d\vec{r}_1=N\ , \tag{8a}$$

$$\rho\int e^{i\vec{k}\cdot\vec{r}_{12}}h(r_{12})d\vec{r}_2=F(k)\ . \tag{8b}$$

We are following a procedure used earlier in developing the uniform-limit formalism.[9,10] In fact, the uniform limit defined by

$$|g(r)-1|=|h(r)|<<1$$

is included as a limiting case in the μ-ordered analysis. What is different in the present study is (i) no restriction on the magnitude of $h(0)$ [the physical value $h(0)=-1$ in the helium problem is not excluded] and (ii) no artificial scaling of the coordinate and wave-vector variables.

Physical quantities are expressed as power series in μ and the adequacy of a polynomial approximation can be tested by comparing coefficients of successive powers of μ. In the final numerical evaluation μ is replaced by unity. The possibility of converting polynomial approximations into Padé approximants is always available and deserves careful study when numerical results become available.

The general rule for determining the factor μ^m associated with a cluster diagram is

$$m=n_h-n_{\vec{r}}+n_{\vec{k}}\ , \tag{9}$$

where n_h is the number of h factors, $n_{\vec{r}}$ is the number of $\rho\int d\vec{r}$ operations, and $n_{\vec{k}}$ is the number of $[1/(2\pi)^3\rho]\int d\vec{k}$ operations. As an example, we consider the cluster integral

$$J(r_{12})=\rho^2\int\int d\vec{r}_3 d\vec{r}_4 h(r_{13})h(r_{14})h(r_{23})h(r_{24})h(r_{34})$$
$$=\frac{1}{(2\pi)^9\rho^3}\int\int\int d\vec{k}_1 d\vec{k}_2 d\vec{k}_3 e^{-i\vec{k}_1\cdot\vec{r}_{12}}F(k_{12})F(k_{13})F(k_2)F(k_3)F(k_{23})\ .$$

The first expression of $J(r_{12})$ has five h factors, two $\rho\int d\vec{r}$ operations, and no $(2\pi)^{-3}\rho^{-1}\int d\vec{k}$ operation, and hence, $m=5-2+0=3$. On the other hand, the second (wave-vector) representation involves three $(2\pi)^{-3}\rho^{-1}\int d\vec{k}$ operations with no h factor and no $\rho\int d\vec{r}$ operation, giving $m=0-0+3=3$. Thus, both of the expressions yield the same result that $J(r_{12})$ is $O(\mu^3)$.

The μ ordering groups together all cluster integrals with the same value of m, in effect applying the qualitative working hypothesis that the operation $\int d\vec{r}$ leaves the order of magnitude of the cluster integrand unchanged. The coefficient of μ^m in the power-series formula for $p^{(n)}$ is then found to have simple properties which are useful in the physical applications. The simplicity becomes strikingly apparent in the n-point structure functions $S^{(n)}(\vec{k}_1,\vec{k}_2,\ldots,\vec{k}_n)$ related to the Fourier transforms of $p^{(n)}(1,2,\ldots,n)$. Unless there could be any ambiguities, we will often use the notation

$$S^{(n)}\equiv S^{(n)}(\vec{k}_1,\vec{k}_2,\ldots,\vec{k}_n)\ .$$

Section II contains the derivation of μ-ordered approximations for $A^{(n)}$ and $p^{(n)}$ with emphasis on $n=3$ and Sec. III deals with the corresponding derivations of $S^{(n)}$. In Sec. IV the μ-ordered form for $p^{(3)}$ is used to derive the hypernetted chain (HNC) formula and improvements thereon. Procedures for testing the accuracy of μ-ordered forms of $p^{(3)}$ are developed in Sec. V. Questions related to the asymptotic behavior of $g(r)$ in the context of equilibrium statistical mechanics are taken up in Sec. VI. Appendices A and B give explicit expressions for various cluster integrals obtained analytically with the use of the Gaussian model for $h(r)$.

II. DERIVATION OF μ ORDERED $A^{(n)}$ AND $p^{(n)}$

To derive series expansions for $A^{(n)}$ and $p^{(n)}$ in powers of μ, it must be remembered that the parameter μ is introduced in the analysis through the substitution scheme given by Eqs. (7). In par-

ticular, it is clear that with substitution (7a), Eq. (5) takes the form

$$p_K^{(n)} \rightarrow \left[\frac{\rho}{\mu}\right]^n \sum_{m=0}^{n(n-1)/2} \mu^m P_{Km}^{(n)}(1,2,\ldots,n) . \tag{10a}$$

We now assume that under the substitution of Eqs. (7) the μ-ordered form of $A^{(n)}$ can be written as

$$A^{(n)} \rightarrow \sum_{m=2}^{\infty} \mu^m A_m^{(n)}(1,2,\ldots,n) . \tag{10b}$$

The trivial solutions $A_m^{(n)}=0$ $(m \leq 1)$ assumed in Eq. (10b) can easily be obtained under the solution scheme to be employed in this section. Substituting Eqs. (10a) and (10b) into the first line of Eq. (6), we see that $p^{(n)}$ has the form

$$p^{(n)} \rightarrow \left[\frac{\rho}{\mu}\right]^n \sum_{m=0}^{\infty} \mu^m P_m^{(n)}(1,2,\ldots,n) . \tag{10c}$$

In Eqs. (10a)–(10c) arrows simply indicate the fact that expressions on the right-hand side of the arrows are consequences of the substitutions given by Eqs. (7).

We now consider the μ expansions (10a)–(10c) in the sequential relation of Eq. (3). Using the second line of Eq. (6), a factor $p^{(n-1)}(1,2,\ldots,n-1)$ can be extracted from both members of the sequential relation, leaving the formula

$$N-n+1=\frac{\rho}{\mu}\int d\vec{r}_n \prod_{j=1}^{n-1}[1+\mu h(r_{jn})] \exp\left[\sum_{m=2}^{\infty} \mu^m [A_m^{(n)}(1,2,\ldots,n)-A_m^{(n-1)}(1,2,\ldots,n-1)]\right] . \tag{11}$$

Equation (11) can be written in the form

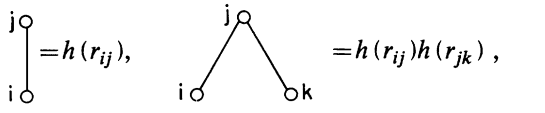
$$\sum_{m=0}^{\infty} \mu^m B_m^{(n)}(1,2,\ldots,n-1)=0 . \tag{12}$$

We treat μ as a genuinely variable quantity and obtain from Eq. (12) the sequence of conditions

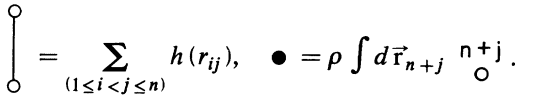
$$B_m^{(n)}(1,2,\ldots,n-1)=0, \quad m=0,1,2,\ldots , \tag{13}$$

which can be used to determine possible explicit forms for $A_m^{(n)}(1,2,\ldots,n)$.

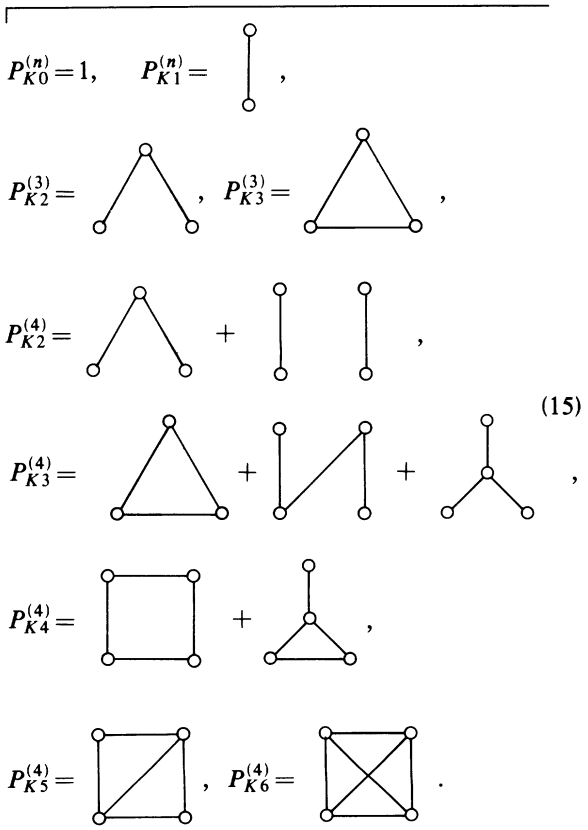
A conventional diagrammatic notation is helpful in working out the consequences of Eq. (13):



(14)

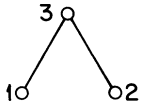


In the usual terminology an open circle is a *root point* and an open circle attached to a cluster by a single line is a *terminal* root point. An unlabeled cluster diagram represents the sum of all distinct clusters obtained by permuting the root indices $1,2,\ldots,n$. In terms of diagrams

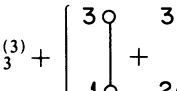


(15)

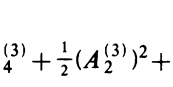
Here each diagram represents a sum of all distinct clusters with the coordinate indices ranging from 1 to n. For $n=3$, Eq. (11) yields

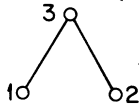
$$\rho\int\left[A_2^{(3)}(1,2,3)+\right.$$


$$\left.\right]d\vec{r}_3=0\,, \qquad (16a)$$

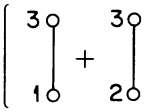
$$\rho\int\left[A_3^{(3)}+\left[\right.\right.$$


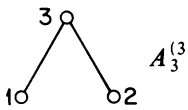
$$\left.\left.\right]A_2^{(3)}\right]d\vec{r}_3=0\,, \qquad (16b)$$

$$\rho\int\left[A_4^{(3)}+\tfrac{1}{2}(A_2^{(3)})^2+\left[\right.\right.$$


$$\left.\left.\right]A_3^{(3)}+\right.$$


$$\left.A_2^{(3)}\right]d\vec{r}_3=0\,, \qquad (16c)$$

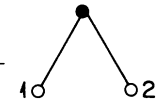
$$\rho\int\left[A_5^{(3)}+A_2^{(3)}A_3^{(3)}\right.$$


$$[A_4^{(3)}+\tfrac{1}{2}(A_2^{(3)})^2]$$


$$\left.A_3^{(3)}\right]d\vec{r}_3=0\,. \qquad (16d)$$

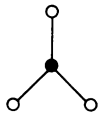
In solving these integral equations we seek *minimal* solutions using the *postulate of minimal complexity,* which asserts that the solutions are symmetrical functions involving the simplest possible clusters in h and the smallest number of clusters consistent with necessary physical conditions. One essential condition is that no direct bonds connecting any two or more points in $\vec{r}_1,\vec{r}_2,\ldots,\vec{r}_n$ occur in the cluster expansion of $A^{(n)}$, meaning that there are no $h(r_{ij})$ factors with both i and j in the set $\{1,2,\ldots,n\}$; any path linking two open circles passes through at least one solid circle. Thus $A^{(n)}$ involves only a type of indirect coupling which is entirely lacking in $p_K^{(n)}$. Meeron[13] formulated the condition as part of a general rule characterizing $A^{(n)}$ in his development of the HNC formalism for classical statistical mechanics of an imperfect gas. We need the condition just to exclude the (unphysical) solution generated by setting the integrands in symmetrized versions of Eq. (16) equal to zero. References 4, 5, and 10 give an appropriately strengthened version of the general rule formulated in the context of classical statistical mechanics of an interacting gas or the equivalent context of Bijl-Dingle-Jastrow (BDJ)-type trial functions describing an interacting boson system: (1) solid circles are directly connected among themselves and (2) open circles are not directly connected among themselves (implying that each open circle is connected to at least one solid circle).

The requirement of a symmetrical solution excludes the possibility of simply equating the integrand in Eq. (16a) to zero. An equivalent form for Eq. (16a) is

$$\rho\int A_2^{(3)}(1,2,3)d\vec{r}_3=-$$


$$\,, \qquad (17)$$

with the minimal solution

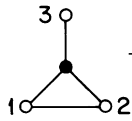
$$A_2^{(3)}(1,2,3)=$$

$$\,. \qquad (18)$$

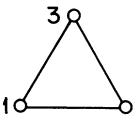
Here we use the basic integral condition on h,

$$\rho^{-1}\int[p^{(2)}(1,2)-\rho^2]d\vec{r}_1=\rho\int h(r_{12})d\vec{r}_1 =-1\,, \qquad (19)$$

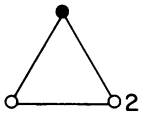
which is simply a statement of the sequential relation for $n=2$ and does not involve the asymptotic behavior of the radial distribution function or the value of $S(k)$ at the origin.

A symmetrical integrand is obtained in Eq. (16b) by adding the integral

$$\rho\int\left[\right.$$


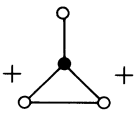
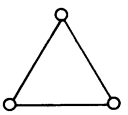
$$+$$


$$\left.\right]d\vec{r}_3$$

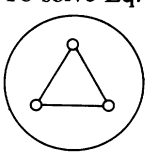
$$=\left[\rho\int h(r_{34})d\vec{r}_3+1\right]$$


$$=0\,. \qquad (20)$$

Now Eq. (16b) can be written in an explicit diagrammatic form

$$\rho\int\left[A_3^{(3)}(1,2,3)+\right.$$

$$+$$

$$\left.\right]d\vec{r}_3=0\,, \qquad (21)$$

in which the integrand is symmetrical in $\vec{r}_1$, $\vec{r}_2$, $\vec{r}_3$. To solve Eq. (21), consider the cluster function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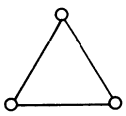
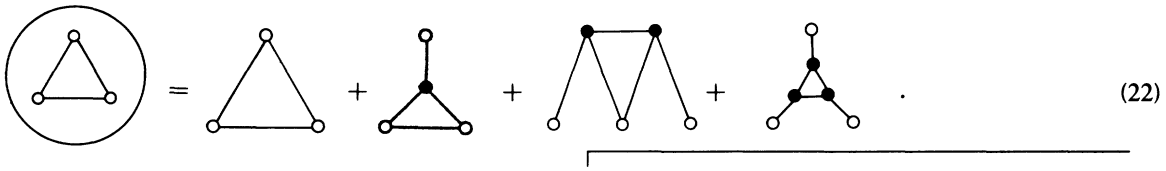


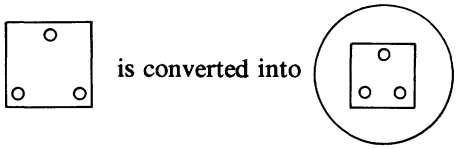
by substituting for $(j=1,2,3)$ in all possible distinct combinations from zero to three:



(22)

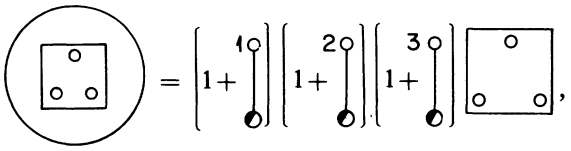
Equation (22) introduces a generalization of a construction used effectively by Wu and Chien[2] in a paper on the structure and properties of the general convolution form $p_c^{(n)}$. We define a *primitive cluster*

as one with no terminal root points. Some of the examples are those given by Eq. (29) and the first cluster on the right-hand side of Eq. (22). The general primitive cluster symbol



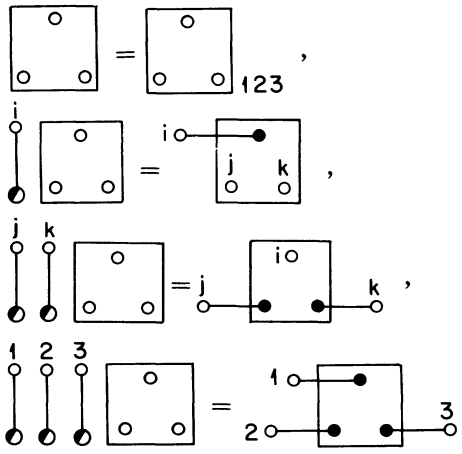
is converted into

by the construction on which Eq. (22) is based, i.e., attaching terminal root points in all possible distinct ways. Symbolically, for $n=3$,



(23)

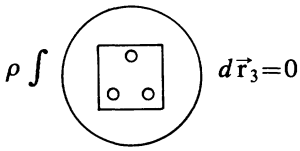
assuming the distributive law and the interpretation



(24)

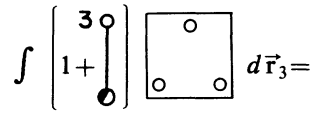
where (i,j,k) is a permutation of (1,2,3).

The equation



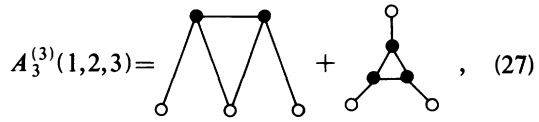
(25)

is easily verified as a consequence of Eq. (19) in the form



(26)

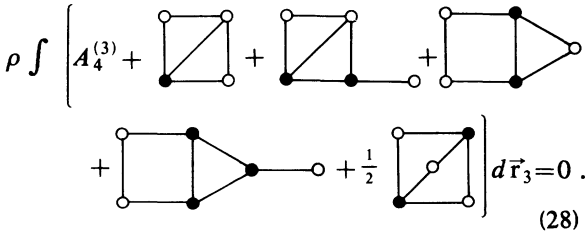
The comparison of Eqs. (21), (22), and (25) immediately yields the minimal solution



(27)

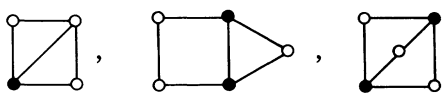
a result originally found in a study of the Abe problem using the uniform-limit formalism.[1,10]

Equation (16c) is easily solved by essentially the same procedure. Again a symmetrical integrand is obtained by adding an unsymmetrical set of diagrams which integrate to zero. The resulting form for Eq. (16c) is



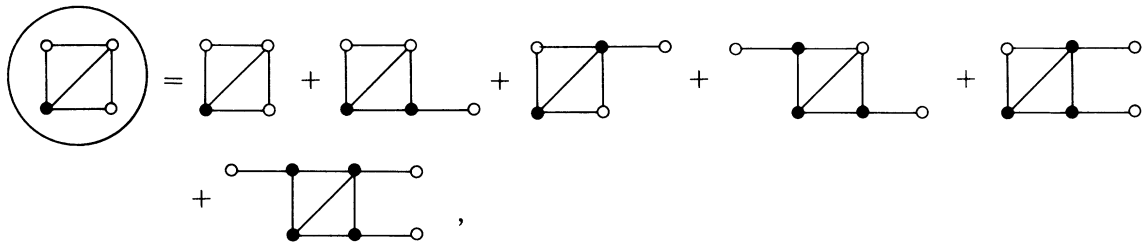
(28)

Three primitive clusters

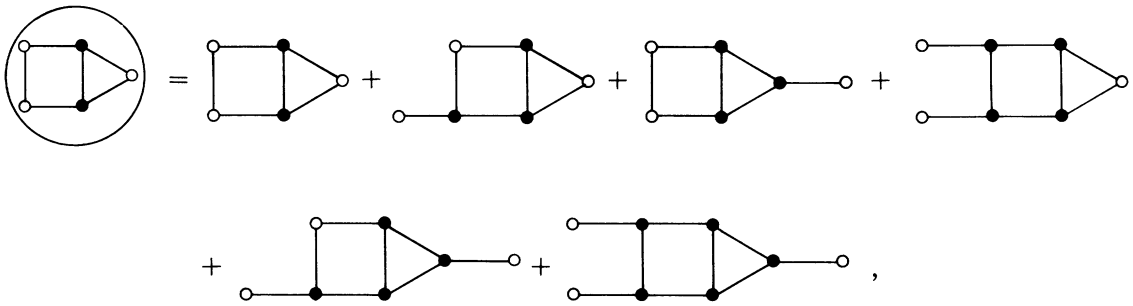


(29)

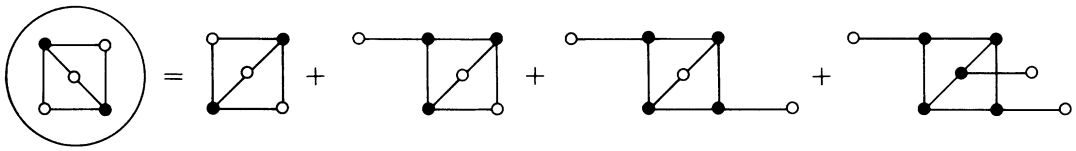
are used to generate



(30a)

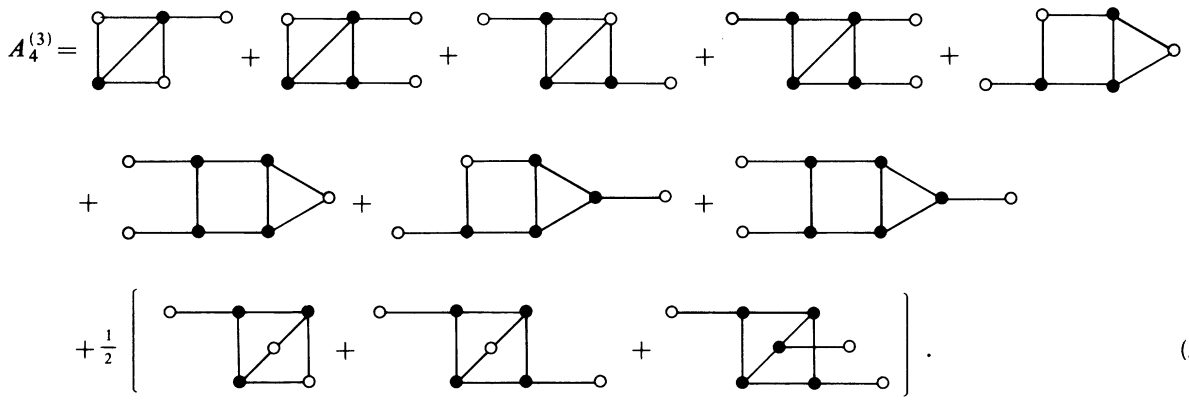


(30b)



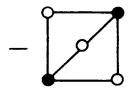
(30c)

By construction these functions are solutions of Eq. (25); consequently, the minimal solution of Eq. (28) is



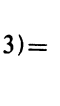
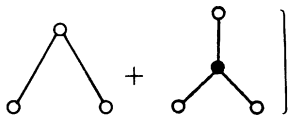
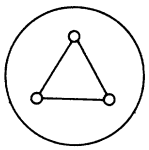
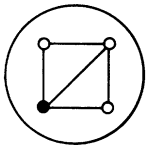
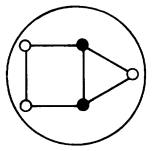
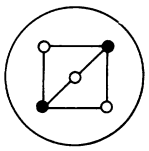
(31)

Here rule 1 enters for the first time to require the replacement of



by the sum of three clusters inside the parentheses. Equation (31) agrees with the result found by Lee[10] and by Chung[11] in the context of the Abe problem. (Our h function corresponds to $-G$ in Ref. 10.)

Equations (6), (10), (18), (27), and (31) combine to yield

$p^{(3)}(1,2,3)=\left[\frac{\rho}{\mu}\right]^3\Bigg[1+\mu$  $+\mu^2\Bigg[$  $+\mu^3$  $+\mu^4\Bigg[$  $+$  $+\frac{1}{2}$  $\Bigg]+O(\mu^5)\Bigg]$, (32)

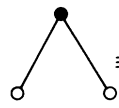
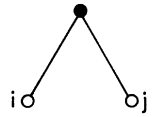
again in agreement with Chung[11] for $\mu=1$. The distinguishing feature of Eq. (32) is that the sequential relation connecting $n=2$ and 3 is satisfied exactly by the first part containing only zeroth-, first-, and second-order terms, which is just the convolution form[1,2,14,15] $p_c^{(3)}$ given by Eq. (39a). Third- and higher-order terms in μ make no contribution to the sequential relation, since they consist of only circled primitive clusters [which integrate to zero as shown by Eq. (25)].

For $n>3$, Eqs. (11)–(13) require

$$\int\left[A_2^{(n)}(1,2,\ldots,n)-A_2^{(n-1)}(1,2,\ldots,n-1)+\sum_{(1\le i<j\le n-1)}h(r_{in})h(r_{nj})\right]d\vec{r}_n=0\,, \tag{33a}$$

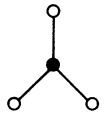
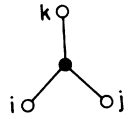
$$\int\Bigg[A_3^{(n)}(1,2,\ldots,n)-A_3^{(n-1)}(1,2,\ldots,n-1)+[A_2^{(n)}(1,2,\ldots,n)-A_2^{(n-1)}(1,2,\ldots,n-1)]\sum_{i=1}^{n-1}h(r_{in})+\sum_{(1\le i<j<k\le n-1)}h(r_{in})h(r_{jn})h(r_{kn})\Bigg]d\vec{r}_n=0\,. \tag{33b}$$

Corresponding diagrammatic forms are

$\rho\int[A_2^{(n)}(1,2,\ldots,n)-A_2^{(n-1)}(1,2,\ldots,n-1)]d\vec{r}_n=-$  $\equiv-\sum_{(1\le i<j\le n-1)}$ 

, (34)

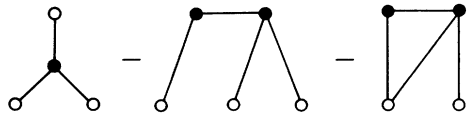
with the minimal solution

$A_2^{(n)}(1,2,\ldots,n)=$  $\equiv\sum_{(1\le i<j<k\le n)}$ 

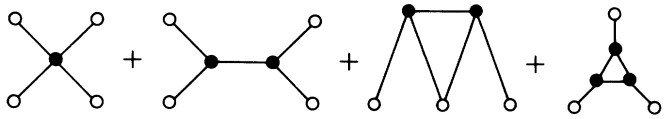
, (35)

and

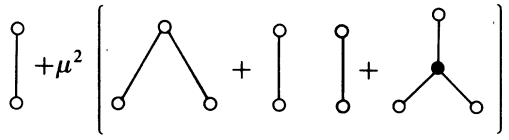
$\rho\int[A_3^{(n)}(1,2,\ldots,n)-A_3^{(n-1)}(1,2,\ldots,n-1)]d\vec{r}_n$

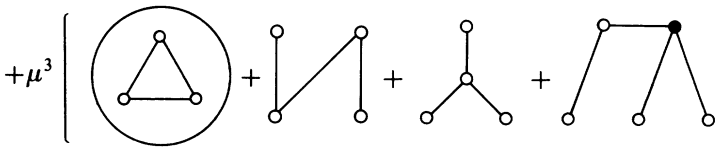
$=-$  , (36)

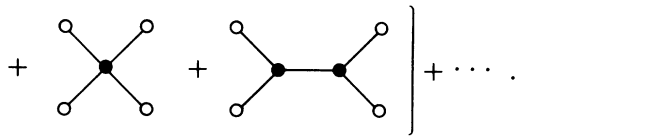
with the minimal solution

$A_3^{(n)}(1,2,\ldots,n)=$  (37)

For $n=4$ these results check the derivation presented in Ref. 10, where $A_4^{(4)}$ is also given.
Specializing to $n=4$, Eqs. (6), (15), (35), and (37) yield

$\left[\frac{\mu}{\rho}\right]^4 p^{(4)}(1,2,3,4)=1+\mu$ 

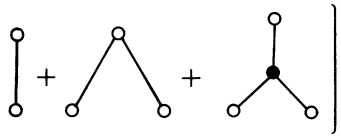


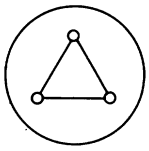
 (38)

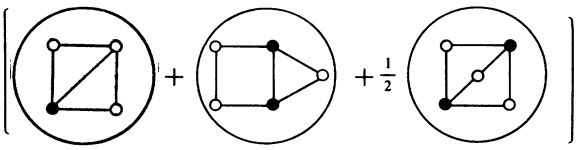
In testing and applying these results, it is convenient to label truncated forms for $p^{(n)}$ with the degree in μ. Thus, we write (for $\mu=1$),

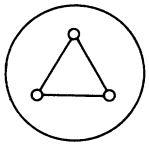
$$p_m^{(n)}=\sum_{l=0}^{m} P_l^{(n)} .$$

Then

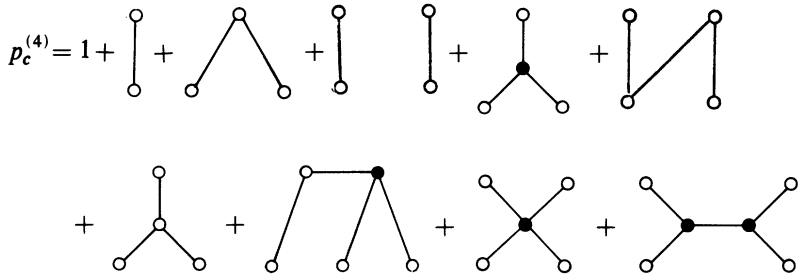
$p_2^{(3)}=p_c^{(3)}=\rho^3\left[1+\right.$  , (39a)

$p_3^{(3)}=p_c^{(3)}+\rho^3$  , (39b)

$p_4^{(3)}=p_3^{(3)}+\rho^3\left[\right.$  , (39c)

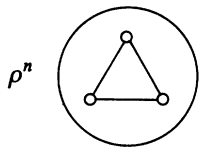
$p_3^{(4)}=p_c^{(4)}+\rho^4$  , (40a)

where

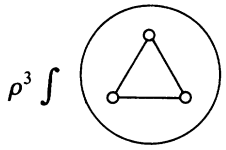


(40b)

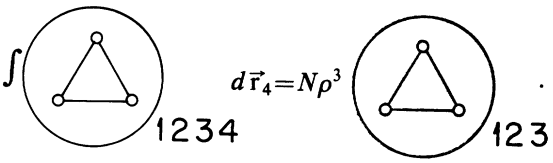
is the convolution form for $n=4$.[1,2,15,16] Equations (39b) and (40a) show that for $n=3$ and 4 the convolution form $p_c^{(n)}$ is supplemented by the term



to give $p_3^{(n)}$. The circled symbol in Eq. (39b) is given by Eq. (22) for a set of indices (1,2,3), but the same symbol in Eq. (40a) signifies a sum over all four sets of three indices: (1,2,3), (1,2,4), (1,3,4), and (2,3,4). Consequently, using Eq. (25), we find



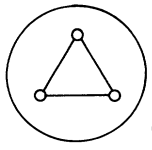
$$\rho^3 \int \quad d\vec{r}_3 = 0 \,, \tag{41}$$



$$\rho^4 \int \quad d\vec{r}_4 = N\rho^3 \quad . \tag{42}$$

Observe that while the cubic polynomial form $p_3^{(3)}$ of Eq. (39b) satisfies the sequential relation (3) accurately, $p_3^{(4)}$ given by Eq. (40a) is not quite adequate in the sequential relation to generate $p_3^{(3)}$ exactly; instead it yields

$$\frac{1}{N-3}\int p_3^{(4)}(1,2,3,4)\,d\vec{r}_4 = p_3^{(3)}(1,2,3) + \frac{3}{N-3}\rho^3$$



, (43)

where the error term is of order μ^3 but vanishes in the limit $N\to\infty$. In Ref. 10, it is pointed out that in the sequential relation $p_4^{(4)}$ generates $p_4^{(3)}$ with the error term of order μ^4.

III. EXPLICIT FORMS FOR μ ORDERED $S^{(3)}(\vec{k}_1,\vec{k}_2,\vec{k}_3)$

In applications the distribution functions may occur as generators of structure functions. These are essentially n-point Fourier transforms defined by

$$S^{(n)}(\vec{k}_1,\vec{k}_2,\ldots,\vec{k}_n) = N^{-1}\int \Psi_0^2 \rho_{\vec{k}_1}\rho_{\vec{k}_2}\cdots\rho_{\vec{k}_n} d\vec{r}_{12\cdots N} \tag{44}$$

with

$$\vec{k}_1+\vec{k}_2+\cdots+\vec{k}_n=0 \tag{45}$$

plus the restriction that no proper subset of the $\vec{k}$'s adds up to zero.[1] For $n=3$, it is convenient to define the Fourier transform of a symmetrical function $f(\vec{r}_1,\vec{r}_2,\vec{r}_3)$:

$$\Delta(\vec{k}_1,\vec{k}_2,\vec{k}_3\,|\,f) = \frac{1}{N}\rho^3 \int e^{i(\vec{k}_1\cdot\vec{r}_1+\vec{k}_2\cdot\vec{r}_2+\vec{k}_3\cdot\vec{r}_3)} \times f(\vec{r}_1,\vec{r}_2,\vec{r}_3)\,d\vec{r}_{123} \,. \tag{46}$$

Equation (44) for $n=3$ then becomes

$$S^{(3)}(\vec{k}_1,\vec{k}_2,\vec{k}_3) = -2+S(k_1)+S(k_2)+S(k_3) + \Delta(\vec{k}_1,\vec{k}_2,\vec{k}_3\,|\,p^{(3)}/\rho^3) \,. \tag{47}$$

The result of $S^{(3)}$ derived with $p_m^{(3)}$ for $p^{(3)}$ will be denoted by

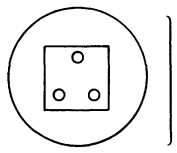
$$S_m^{(3)} \equiv S_m^{(3)}(\vec{k}_1,\vec{k}_2,\vec{k}_3) \,.$$

For $m=2$, 3, and 4, $S_m^{(3)}$ can be obtained using Eqs. (39a)–(39c). Evaluation of the convolution form simply yields[1,13,14]

$$S_2^{(3)}(\vec{k}_1,\vec{k}_2,\vec{k}_3) \equiv S_c^{(3)}(\vec{k}_1,\vec{k}_2,\vec{k}_3) = S(k_1)S(k_2)S(k_3) \,. \tag{48}$$

The involved algebra for $m=3$ and 4 is equally

straightforward but becomes extremely complicated if the circled diagrams in Eqs. (39b) and (39c) are replaced with the explicit expressions given by Eqs. (22) and (30). This is the procedure used in Ref. 17 for $S_3^{(3)}$ and in Ref. 11 for $S_3^{(3)}$ and $S_4^{(3)}$. Fortunately, it can be shown that Eqs. (23) and (24) permit a very direct and quite simple derivation of the Fourier transform of a general circled diagram

$\Delta\left[\vec{k}_1,\vec{k}_2,\vec{k}_3\,|\right.$  $\left.\right]$

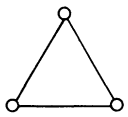
$$=\frac{1}{N}\rho^3\left[\prod_{j=1}^{3}\left[\int d\vec{r}_j e^{i\vec{k}_j\cdot\vec{r}_j}+\int d\vec{r}_{j+3}e^{i\vec{k}_j\cdot\vec{r}_{j+3}}\rho\int d\vec{r}_j e^{i\vec{k}_j\cdot\vec{r}_{j,j+3}}h(r_{j,j+3})\right]\right]$$ 

$$=\frac{1}{N}\rho^3\left[\prod_{j=1}^{3}\int d\vec{r}_j e^{i\vec{k}_j\cdot\vec{r}_j}[1+F(k_j)]\right]$$ 

$$=S(k_1)S(k_2)S(k_3)\Delta\left[\vec{k}_1,\vec{k}_2,\vec{k}_3\,|\right.$$  $\left.\right]$. (49)

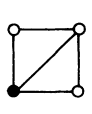
Using Eqs. (40) and (41), we verify the Chung formula[11] for $S_3^{(3)}$ and $S_4^{(3)}$:

$$S_3^{(3)}(\vec{k}_1,\vec{k}_2,\vec{k}_3)=S(k_1)S(k_2)S(k_3)[1+\Delta_3(\vec{k}_1,\vec{k}_2,\vec{k}_2)]\,, \tag{50}$$

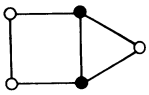
$$\Delta_3(\vec{k}_1,\vec{k}_2,\vec{k}_3)=\Delta\left[\vec{k}_1,\vec{k}_2,\vec{k}_3\,|\right.$$  $\left.\right]$

$$=\frac{1}{(2\pi)^3\rho}\int F(k)F(|\vec{k}+\vec{k}_2|)F(|\vec{k}-\vec{k}_3|)d\vec{k}\,, \tag{51}$$

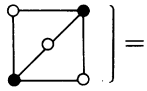
$$S_4^{(3)}(\vec{k}_1,\vec{k}_2,\vec{k}_3)=S(k_1)S(k_2)S(k_3)[1+\Delta_3(\vec{k}_1,\vec{k}_2,\vec{k}_3)+\Delta_{4a}(\vec{k}_1,\vec{k}_2,\vec{k}_3)$$
$$+\Delta_{4b}(\vec{k}_1,\vec{k}_2,\vec{k}_3)+\tfrac{1}{2}\Delta_{4c}(\vec{k}_1,\vec{k}_2,\vec{k}_3)]\,, \tag{52}$$

$$\Delta_{4a}(\vec{k}_1,\vec{k}_2,\vec{k}_3)=\Delta\left[\vec{k}_1,\vec{k}_2,\vec{k}_3\,|\right.$$  $\left.\right]$

$$=\frac{1}{(2\pi)^6\rho^2}\int[F(|\vec{k}+\vec{k}_1|)F(|\vec{k}'+\vec{k}_2|)+F(|\vec{k}+\vec{k}_2|)F(|\vec{k}'+\vec{k}_3|)$$
$$+F(|\vec{k}+\vec{k}_3|)F(|\vec{k}'+\vec{k}_1|)]F(k)F(k')F(|\vec{k}+\vec{k}'|)d\vec{k}\,d\vec{k}'\,, \tag{53}$$

$$\Delta_{4b}(\vec{k}_1,\vec{k}_2,\vec{k}_3)=\Delta\left[\vec{k}_1,\vec{k}_2,\vec{k}_3\,|\right.$$  $$\left.\right]$$

$$=\frac{1}{(2\pi)^6\rho^2}\int[F(|\vec{k}+\vec{k}_1|)F(|\vec{k}'+\vec{k}_2|)F(|\vec{k}'+\vec{k}_2+\vec{k}_3|)$$
$$+F(|\vec{k}+\vec{k}_2|)F(|\vec{k}'+\vec{k}_3|)F(\vec{k}'+\vec{k}_3+\vec{k}_1|)$$
$$+F(|\vec{k}+\vec{k}_3|)F(|\vec{k}'+\vec{k}_1|)F(|\vec{k}'+\vec{k}_1+\vec{k}_2|)]$$
$$\times F(k)F(k')F(|\vec{k}+\vec{k}'|)d\vec{k}\,d\vec{k}'\,, \tag{54}$$

$$\Delta_{4c}(\vec{k}_1,\vec{k}_2,\vec{k}_3)=\Delta\left[\vec{k}_1,\vec{k}_2,\vec{k}_3\,|\right.$$  $$\left.\right]=\frac{1}{(2\pi)^6\rho^2}\int F(|\vec{k}+\vec{k}_1|)F(|\vec{k}'+\vec{k}_2|)F(|\vec{k}+\vec{k}'-\vec{k}_3|)$$

$$\times F(k)F(k')F(|\vec{k}+\vec{k}'|)d\vec{k}\,d\vec{k}'\,. \tag{55}$$

In the expressions for Δ_3 and Δ_{4c} the actual symmetry in $\vec{k}_1$, $\vec{k}_2$, $\vec{k}_3$ is not explicit, but symmetry is easily demonstrated by suitable displacements and reflections in the space of the integration variables. The n-point structure function $S^{(n)}$ for general n has been evaluated by Wu[18] under the convolution approximation $p_c^{(n)}$.

The fact that the *minimal* form for $A^{(3)}$ in the context of the ground-state solution Ψ_0 coincides with the corresponding form generated by the BDJ trial function Φ_0 is at first welcome because it indicates in still another way a close correspondence between Ψ_0 and the optimum Φ_0. However, we must recognize that *minimal* does not necessarily mean *correct.* It seems likely that an exact cluster formula for $p^{(3)}$ generated by Ψ_0 must contain many cluster integrals which do not occur in the *minimal* solution. At present, very little is known about such terms and no procedure is available for generating or estimating them. Fortunately independent tests are available which are capable of yielding some quantitative information on the absolute and relative accuracy of approximate trial forms. Such a test is discussed in Sec. V.

IV. AN IMPROVED HYPERNETTED CHAIN RELATION

The results stated in this section are not new, but the earlier derivation[9,10,15] in the context of the uniform-limit formalism involved the explicit assumption $|g(r)-1|=|h(r)|\ll 1$ and an artificial scaling of the coordinate and wave vector variables. The same relations can be developed using the μ-ordered expansion for $p^{(3)}$. The analysis starts from the BDJ approximation for Ψ_0:

$$\Phi_0(1,2,\ldots,N)=\prod_{(1\le i<j\le N)}e^{u(r_{ij})/2}\Bigg/\left[\int\prod_{(1\le m<n\le N)}e^{u(r_{mn})}d\vec{r}_{1,2,\ldots,N}\right]^{1/2}, \tag{56}$$

and an inhomogeneous, linear, integral equation of a familiar type [the first equation of a Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy]:

$$\nabla_1 u(r_{12})=\nabla_1\ln g(r_{12})-\int K(1,2;3)\nabla_1 u(r_{13})d\vec{r}_3\,, \tag{57}$$

which determines $u(r)$ as an explicit functional in $g(r)$ or $S(k)$, if the kernel

$$K(1,2;3)=[p^{(3)}(1,2,3)-\rho^3 g(r_{12})g(r_{13})]/\rho^3 g(r_{12}) \tag{58}$$

is given as an explicit functional in $g(r)$ or $S(k)$. Under the μ-ordering scheme discussed in Secs. I–III, the kernel in the integral equation (57) enters as a power series in μ and the integral equation is solved by expressing $u(r)$ as a power series in μ in the form

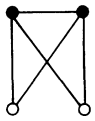
$$u(r)=\ln[1+\mu h(r)]-\sum_{m=1}^{\infty}\mu^m u_m(r)\ . \tag{59}$$

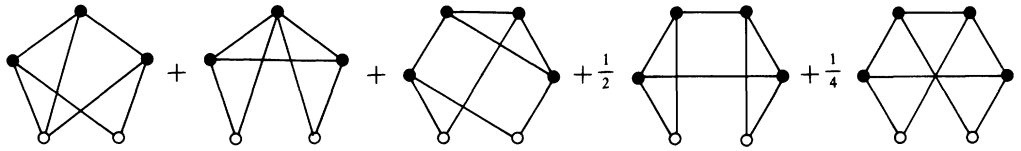
With $\mu<1$, the assumption that $u(r)$ and $u_m(r)$ possess Fourier transforms involves no difficulty and facilitates the development of an explicit solution. The final step of taking the limit $\mu\to 1$ is applied to a form in which the Fourier transforms of excessively singular functions do not occur. For details which are easily translated into the μ-ordered notation, the reader is referred to Ref. 10.

The final result is

$$u_1(r)=\frac{1}{(2\pi)^3\rho}\int e^{i\vec{k}\cdot\vec{r}}\frac{[S(k)-1]^2}{S(k)}d\vec{k}\ , \tag{60a}$$

$$u_2(r)=0\ , \tag{60b}$$

$u_3(r_{12})=\frac{1}{2}$  , (60c)

$u_4(r_{12})=$  (60d)

For $\mu=1$, Eq. (59) with $u_m(r)=0$ for $m>2$, i.e.,

$$u(r)=\lng(r)-u_1(r)$$

is the HNC formula.

An earlier derivation of a closely related general formula appears in the work by van Leeuwen, Groeneveld, and de Boer[19]:

$$w(r_{12})=\rho\int[g(r_{13})-1]\times[g(r_{32})-1-w(r_{32})]d\vec{r}_3\ , \tag{61}$$

where

$$w(r)=\lng(r)-u(r)-\epsilon(r)\ , \tag{62}$$

and $\epsilon(r)$ is given as a sum of so-called *bridge* clusters. In wave-vector space Eq. (61) reduces to

$$\rho\int e^{i\vec{k}\cdot\vec{r}}w(r)d\vec{r}=[S(k)-1]^2/S(k)\ . \tag{63}$$

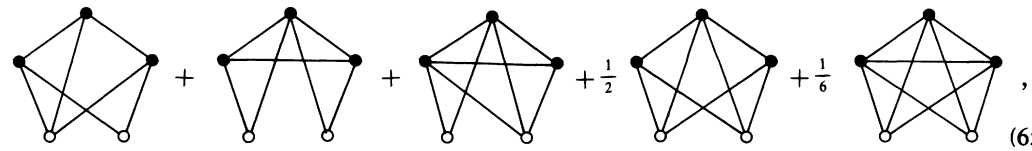
Consequently,

$$u(r)=\lng(r)-\frac{1}{(2\pi)^3\rho}\int e^{i\vec{k}\cdot\vec{r}}\frac{[S(k)-1]^2}{S(k)}d\vec{k}-\epsilon(r) \tag{64}$$

and $-\epsilon(r)$ is the correction to the HNC approximation. Smith[20] writes an explicit approximation for $\epsilon(r)$ as

$$\epsilon(r)=\epsilon_4(r)+\epsilon_5(r)+\cdots\ , \tag{65a}$$

$$\epsilon_4(r)=u_3(r)\ , \tag{65b}$$

$\epsilon_5(r_{12})=$  , (65c)

the subscripts of ϵ denoting the number of particles (sum of the number of open circles and number of solid circles) involved in the cluster formula. Both derivations yield the same leading correction.

However, u_4 and ϵ_5, each a linear combination of five clusters, have only two clusters in common; u_4 is a linear combination of five- and six-particle clusters involving seven and eight bonds, respec-

tively, while ϵ_5 contains only five-particle clusters and the number of bonds range from seven to nine meaning that ϵ_5 includes terms belonging to the coefficients of μ^4, μ^5, and μ^6. The difference expresses the fact that the μ expansion is based on a rational ordering principle whereas the grouping of all clusters generated by a given number of particles is essentially reverting to the use of number density as an expansion parameters.

No positive information is available at present as to which form, u_4 or ϵ_5, represents the better approximation under realistic conditions for liquid helium. On the other hand, the Gaussian model for the function $h(r)$ [given by Eq. (75)] is useful in estimating the relative magnitudes of the cluster integrals appearing in Eqs. (60d) and (65c). The obtained results are presented in Appendix A, where the norms and average values of the cluster integral functions are also given. Although no definite conclusion can be drawn from the comparison of the numerical results, they are not unfavorable to the view that the six-particle cluster terms in $u_4(r)$ may generally contribute more than the μ^5 and μ^6 terms in $\epsilon_5(r)$.

V. A PROCEDURE FOR TESTING THE ACCURACY OF $p_m^{(3)}$

A realistic description of liquid ^{4}He requires that all distribution functions ($n \geq 2$) vanish if two points coincide. This reflects the occurence of strong repulsive interactions when the K-shell regions of two He atoms begin to overlap. The condition is identically satisfied by the Kirkwood form $p_K^{(n)}$ and hence by the Abe form $p^{(n)}$ defined by the first line of Eq. (6); however, for the convolution approximation $p_c^{(n)}$ and the μ-ordered polynomial form $p_m^{(n)}$ it fails except at isolated points. Note that this failure by $p_m^{(n)}$ is order by order and is the consequence of the artificial truncation of Eq. (6) induced by the μ-ordered expansion. A point of interest is that

$$\rho \int p_m^{(3)}(1,2,2)d\vec{r}_1=0 , \tag{66}$$

an immediate consequence of the sequential relation. Thus the convolution and μ-ordered forms satisfy the condition $p_m^{(3)}(1,2,2)=0$ in the mean.

The Fourier transform is a useful tool in the study of the magnitude and general behavior of $p_m^{(3)}(1,2,2)$. Let

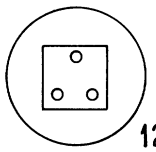
$$Q_m(k)=\rho^{-2} \int e^{i\vec{k}\cdot\vec{r}_{12}} p_m^{(3)}(1,2,2)d\vec{r}_1 . \tag{67}$$

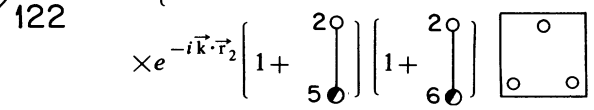
An immediate consequence of Eq. (66) is $Q_m(0)=0$. Substitution of Eqs. (39) into Eq. (67) yields

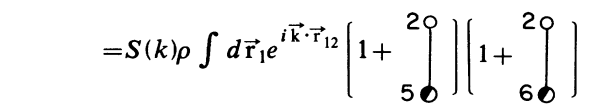
$$\frac{Q_2(k)}{S(k)}=\frac{1}{(2\pi)^3\rho} \int F(|\vec{k}-\vec{k}'|)F(k')d\vec{k}' , \tag{68a}$$

$$\frac{Q_3(k)}{S(k)}=\frac{1}{(2\pi)^6\rho^2} \int [2+F(|\vec{k}-\vec{k}''|)]F(k')F(k'')F(|\vec{k}-\vec{k}'|)F(\vec{k}'-\vec{k}''|)d\vec{k}'d\vec{k}'' . \tag{68b}$$

Using a procedure similar to that employed in Eq. (49), it can be shown that Eqs. (23) and (24) allow a general proof that $S(k)$ occurs as a factor in $Q_m(k)$:

$\rho \int e^{i\vec{k}\cdot\vec{r}_{12}}$  $d\vec{r}_1=\rho\left[\int d\vec{r}_1 e^{i\vec{k}\cdot\vec{r}_1}+\int d\vec{r}_4 e^{i\vec{k}\cdot\vec{r}_4}\rho\int d\vec{r}_1 e^{i\vec{k}\cdot\vec{r}_{14}}h(r_{14})\right]$

$\times e^{-i\vec{k}\cdot\vec{r}_2}$ 

$=S(k)\rho \int d\vec{r}_1 e^{i\vec{k}\cdot\vec{r}_{12}}$  . (69)

Observe that

$$\frac{1}{(2\pi)^3\rho}\int Q_m(k)d\vec{k}=\frac{1}{\rho^3}p_m^{(3)}(1,1,1)\,, \tag{70}$$

expressing the integral of $Q_m(k)$ directly in terms of $p_m^{(3)}$ for $r_{12}=r_{23}=r_{31}=0$. In contrast the quantity

$$W_m=\frac{1}{(2\pi)^3\rho}\int\frac{Q_m(k)}{S(k)}d\vec{k} \tag{71}$$

gives a measure of $p_m^{(3)}(1,2,2)$ for intermediate values of r_{12}. In terms of the direct correlation function

$$C(r)=\frac{1}{(2\pi)^3\rho}\int e^{i\vec{k}\cdot\vec{r}}\left[\frac{1}{S(k)}-1\right]d\vec{k}\,, \tag{72}$$

Eq. (71) transforms into

$$W_m=\rho^{-2}\int C(r_{12})p_m^{(3)}(1,2,2)d\vec{r}_1+p_m^{(3)}(2,2,2)/\rho^3\,. \tag{73}$$

Equations (68) and (71) yield

$$W_2=1\,, \tag{74a}$$

$$W_3=\frac{1}{(2\pi)^6\rho^2}\int d\vec{k}'d\vec{k}''F(k')F(k'')F(|\vec{k}'-\vec{k}''|)\left[-2+\frac{1}{(2\pi)^3\rho}\int d\vec{k}F(|\vec{k}-\vec{k}'|)F(|\vec{k}-\vec{k}''|)\right]. \tag{74b}$$

We now turn to the numerical evaluation of various quantities, using the Gaussian functions (the Gaussian model)

$$h(r)=-e^{-s^2},\quad s\equiv\sqrt{\pi}\rho^{1/3}r\,, \tag{75}$$

$$F(k)=-e^{-q^2},\quad q\equiv k/(2\sqrt{\pi}\rho^{1/3})\,, \tag{76}$$

which have the virture of yielding explicit formulas for cluster integrals.[1,10,12] Note that Eqs. (75) and (76) are related to each other by the Fourier transform expressed by Eq. (4b) or (4c). We use Eqs. (75) and (76) to get some information on the general behavior and magnitude of the cluster integrals occurring in the μ-ordered formulas for $p_m^{(3)}$ and Q_m. This information may be moderately reliable in problems where $h(r)$ is a monotonic function (as in the problem of the charged boson gas).[9,10] Explicit results for the cluster integrals appearing in $p_m^{(3)}(1,2,3)$ are given in Appendix B for $m=2$, 3, and 4. The results for $p_m^{(3)}(1,2,2)/\rho^3$ and $Q_m(k)/S(k)$ are

$$p_2^{(3)}(1,2,2)/\rho^3=e^{-2s_{12}^2}-(\tfrac{1}{3})^{3/2}e^{-2s_{12}^2/3}\,, \tag{77a}$$

$$p_3^{(3)}(1,2,2)/\rho^3=2(\tfrac{1}{3})^{3/2}e^{-5s_{12}^2/3}+\tfrac{1}{64}e^{-s_{12}^2/2}-(\tfrac{1}{8})^{3/2}(e^{-s_{12}^2}+2e^{-5s_{12}^2/8})\,, \tag{77b}$$

$$Q_2(k)/S(k)=(\tfrac{1}{2})^{3/2}e^{-q^2/2}\,, \tag{78a}$$

$$Q_3(k)/S(k)=2(\tfrac{1}{5})^{3/2}e^{-3q^2/5}-(\tfrac{1}{8})^{3/2}e^{-q^2}\,. \tag{78b}$$

We have also obtained the expressions for $m=4$, but they are not given in Eqs. (77) and (78) since they are quite lengthy. Comparison of the results for $p_m^{(3)}(1,2,2)/\rho^3$ and $Q_m(k)/S(k)$ for $m=2$, 3, and 4 illustrates nicely the reduction in computational complexity resulting from the shift to wave-vector space. Figures 1(a) and 1(b) display numerical results for $p_m^{(3)}(1,2,2)/\rho^3$ as function of s_{12}^2 and Fig. 2 shows results of $Q_m(k)/S(k)$ plotted against q^2, each for $m=2$, 3, and 4. Clearly, at least for $m=2$ and 3, $p_{m+1}^{(3)}$ and Q_{m+1} represent substantial improvements over $p_m^{(3)}$ and Q_m, respectively, in approaching the requirement that $p^{(3)}(1,2,3)=0$ if two points coincide.

Substitution of Eqs. (78) into Eq. (71) yields analytic results for W_m, whose numerical values are

$$W_2=1\,,\quad W_3=0.3407\,,\quad W_4=0.048\,32\,. \tag{79}$$

Another scalar quantity that can serve as an average measure of $|p_m^{(3)}(1,2,2)|/\rho^3$ is the square root of the norm

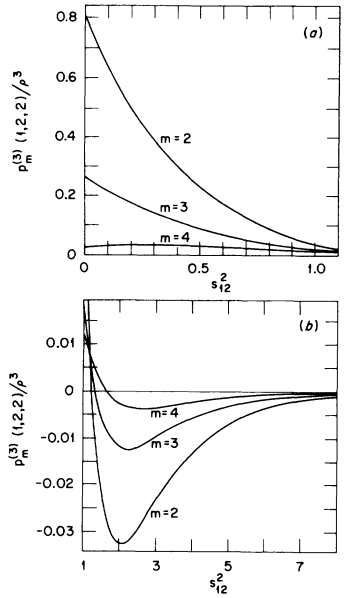


FIG. 1. Plots of $p_m^{(3)}(1,2,2)/\rho^3$ as functions of $s_{12}^2=\pi\rho^{2/3}r_{12}^2$ for $m=2$, 3, and 4.

$$U_m=\left[\rho\int[p_m^{(3)}(1,2,2)/\rho^3]^2 d\vec{r}_1\right]^{1/2}, \tag{80}$$

which can also be evaluated analytically with Eqs. (77). The numerical results for $m=2$, 3, and 4 are

$$\begin{aligned} U_2&=0.2463\ , \\ U_3&=0.088\,99\ , \\ U_4&=0.023\,75\ . \end{aligned} \tag{81}$$

Equations (79) and (81) also demonstrate that the requirement of $p^{(3)}(1,2,2)=0$ is more accurately

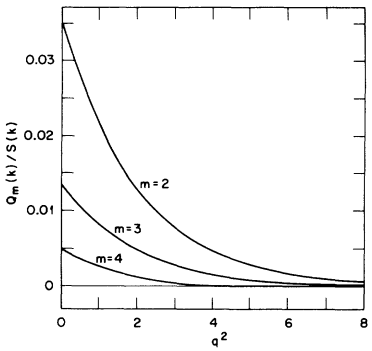


FIG. 2. Plots of $Q_m(k)/S(k)$ as functions of $q^2=k^2/(4\pi\rho^{2/3})$ for $m=2$, 3, and 4.

satisfied by $p_{m+1}^{(3)}(1,2,3)$ than by $p_m^{(3)}(1,2,3)$ for $m=2$ and 3 in the Gaussian model given by Eqs. (75) and (76).

We close this section by pointing out that a somewhat related form for $p^{(3)}(1,2,3)$ is studied in Ref. 21, where the approximation is given by

$$\hat{p}_m^{(3)}(1,2,3)=p_K^{(3)}(1,2,3)\times\exp\left[\sum_{l=2}^{m}A_l^{(3)}(1,2,3)\right]. \tag{82}$$

Note that Eq. (82) for $m=\infty$ is identical to Eq. (6) for $n=3$. While $\hat{p}_m^{(3)}$ appears more general and possibly better than $p_m^{(3)}$, the analysis of this approximation scheme in the wave-vector space is practically impossible. Moreover, the Gaussian function of Eq. (75) does not permit explicit evaluation of integrals involving $\hat{p}_m^{(3)}(1,2,3)$ or $\hat{p}_m^{(3)}(1,2,2)$.

VI. μ ORDERING IN EQUILIBRIUM STATISTICAL MECHANICS

Consider a system of N identical particles interacting in pairs. In the classical formulation of statistical mechanics the N-particle distribution function is

$$p^{(N)}(1,2,\ldots,N)=N!\frac{\exp[-\beta V(1,2,\ldots,N)]}{\int\exp[-\beta V(1,2,\ldots,N)]d\vec{r}_{1,2,\ldots,N}}, \tag{83}$$

where

$$V(1,2,\ldots,N)=\sum_{(1\le i<j\le N)}v(r_{ij})\ . \tag{84}$$

The sequential relation given by Eq. (3) is used to generate $p^{(n)}(1,2,\ldots,n)$, leading finally to

$$p^{(2)}(1,2)=\rho^2 g(r_{12})=N(N-1)\frac{\int\exp[-\beta V(1,2,\ldots,N)]d\vec{r}_{3,4,\ldots,N}}{\int\exp[-\beta V(1,2,\ldots,N)]d\vec{r}_{1,2,\ldots,N}} \tag{85}$$

and

$$p^{(1)}(1)=N/\Omega=\rho\ . \tag{86}$$

Notice that

$$\frac{\rho^2}{N}\int\int[g(r_{12})-1]d\vec{r}_1 d\vec{r}_2=\rho\int[g(r)-g(\infty)]d\vec{r}+N[g(\infty)-1]=-1\ . \tag{87}$$

Now

$$S(k)=1+\rho\int e^{i\vec{k}\cdot\vec{r}}[g(r)-g(\infty)]d\vec{r}\ , \tag{88}$$

$$S(0)=N[1-g(\infty)]$$
$$=k_BT/mc^2$$
(Ornstein-Zernicke relation) . (89)

The function $h(r)=g(r)-1$ has a constant long-range tail which is needed to meet the requirements of the sequential relation as stated in Eqs. (3), (11), and (87). In these relations the range of integration is the fundamental cube and not infinite space; the quantity

$$4\pi\rho\int_0^\infty[g(r)-g(\infty)]r^2dr=-1+S(0) \tag{90}$$

never occurs in a proper statement of the sequential relation.

The conventional procedures used to transform $p^{(n)}$ and $A^{(n)}$ from functionals in $\exp[-\beta v(r)]-1$ into functionals in $h(r)$ appear to treat $h(r)$ as a short-range function. In effect $h(r)$ is replaced by a sequence of short-range functions with $h(r)$ as the (nonuniform) limit

$$\lim_{m\to\infty}h_m(r)=h(r)\ , \tag{91}$$

$$\lim_{r\to\infty}r^3h_m(r)=0\ , \tag{92}$$

$$\rho\int h_m(r_{12})d\vec{r}_2=-1\ . \tag{93}$$

These complications do not surface in the μ-ordering formalism. In general, μ ordering can serve as a useful supplementary procedure. The *minimal* requirement is not expected to yield unique solutions in all problems; but, when it does, the solution should agree with that generated by the conventional inversion procedures.

ACKNOWLEDGMENTS

One of the authors (D.K.L.) is grateful to Professor George Stell for pointing out that the generalization of Abe's work in Ref. 3 was carried out to all orders in Ref. 4. This research was supported in part by the National Science Foundation under Grant No. DMR76-14929 and in part by the U. S. Department of Energy under Contract No. W-7405-eng-26 with the Union Carbide Corporation.

APPENDIX A: COMPARISON OF $u_4(r)$ AND $\epsilon_5(r)$

A brief discussion of the μ ordering and density expansion approaches for $u(r)$ presented in Sec. IV is examined in more detail in this appendix through numerical evaluation of the cluster integrals appearing in Eqs. (60c), (60d), and (65c). The Gaussian model for $h(r)$ defined by Eq. (75) allows analytic and explicit evaluation of the integrals. Their expressions resulting from integration are also Gaussian functions of the form

$$I_k(r_{12})=\frac{1}{D_k\,|\,D_k\,|^{1/2}}e^{-\alpha_k s_{12}^2},\quad s=\sqrt{\pi}\rho^{1/3}r\ . \tag{A1}$$

The relative magnitudes of the cluster integrals may be estimated from the norm

$$||I_k||=\rho\int I_k^2(r)d\vec{r}=\frac{1}{D_k^3(2\alpha_k)^{3/2}}\ . \tag{A2}$$

Another scalar quantity useful in the comparison is the expectation value of $\sum_{i<j}I_k(r_{ij})$ per particle

$$\langle I_k\rangle\equiv\frac{1}{N}\sum_{(1\le i<j\le N)}\int\Psi_0^2I_k(r_{ij})d\vec{r}_{1,2,\ldots,N}$$
$$=\tfrac{1}{2}\rho\int g(r)I_k(r)d\vec{r}$$
$$=\frac{1}{2D_k\,|\,D_k\,|^{1/2}}\left[\frac{1}{\alpha_k^{3/2}}-\frac{1}{(\alpha_k+1)^{3/2}}\right], \tag{A3}$$

where Ψ_0 is the normalized ground-state wave function.

TABLE I. Numerical values of D_k, α_k, $||I_k||$, and $\langle I_k \rangle$ for the cluster integrals I_k given by Eq. (A1).

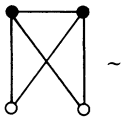
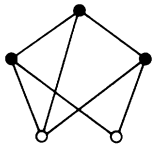
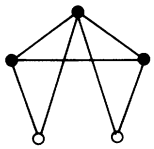
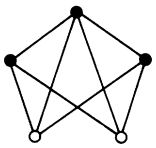
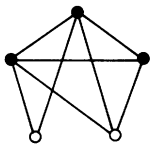
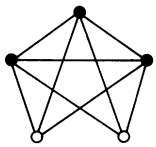
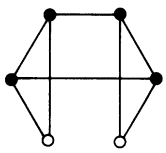
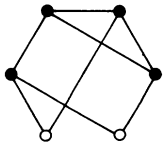
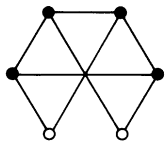
k	I_k	D_k	α_k	$\|\|I_k\|\|$	$\langle I_k \rangle$
1		-8	1	6.905×10^{-4}	1.4285
2		-21	$\frac{8}{7}$	3.125×10^{-5}	0.2596
3		-24	$\frac{7}{8}$	3.125×10^{-5}	0.3539
4		30	$\frac{3}{2}$	7.128×10^{-6}	0.0887
5		35	$\frac{8}{7}$	6.749×10^{-6}	0.1207
6		-50	$\frac{3}{2}$	1.540×10^{-6}	0.0412
7		45	$\frac{2}{3}$	7.128×10^{-6}	0.2273
8		40	$\frac{7}{8}$	6.749×10^{-6}	0.1645
9		45	$\frac{4}{5}$	5.422×10^{-6}	0.1629

TABLE II. Numerical values of D, α, β, and γ for the cluster integrals $I(1,2,3)$ given by Eq. (B1).

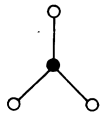
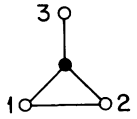
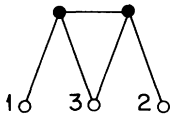
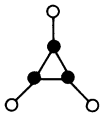
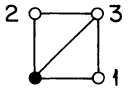
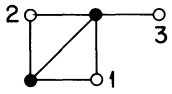
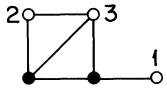
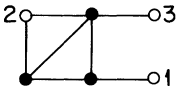
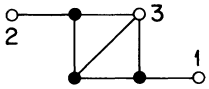
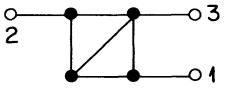
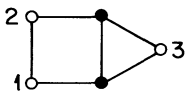
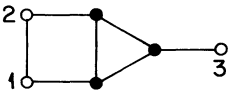
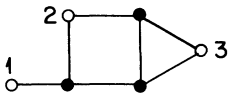
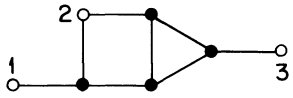
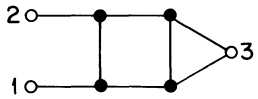
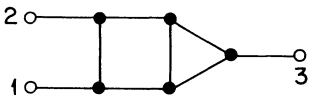
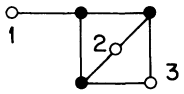
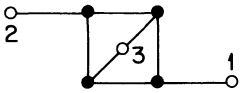
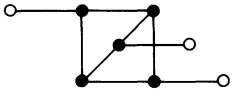
$I(1,2,3)$	D	α	β	γ
	-3	$\frac{1}{3}$	$\frac{1}{3}$	$\frac{1}{3}$
	3	$\frac{1}{3}$	$\frac{1}{3}$	$\frac{4}{3}$
	-8	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{8}$
	16	$\frac{1}{4}$	$\frac{1}{4}$	$\frac{1}{4}$
	-3	$\frac{4}{3}$	$\frac{4}{3}$	$\frac{1}{3}$
	11	$\frac{4}{11}$	$\frac{4}{11}$	$\frac{9}{11}$
	8	$\frac{3}{2}$	$\frac{1}{2}$	$\frac{1}{8}$
	-24	$\frac{1}{2}$	$\frac{1}{6}$	$\frac{3}{8}$
	-21	$\frac{4}{7}$	$\frac{4}{7}$	$\frac{1}{21}$
	45	$\frac{4}{15}$	$\frac{4}{15}$	$\frac{1}{5}$
	8	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{9}{8}$
	-16	$\frac{1}{4}$	$\frac{1}{4}$	$\frac{5}{4}$

TABLE II. (*Continued.*)

$I(1,2,3)$	D	α	β	γ
	-21	$\frac{5}{7}$	$\frac{4}{21}$	$\frac{3}{7}$
	40	$\frac{3}{8}$	$\frac{1}{10}$	$\frac{1}{2}$
	45	$\frac{1}{3}$	$\frac{1}{3}$	$\frac{1}{5}$
	-75	$\frac{1}{5}$	$\frac{1}{4}$	$\frac{4}{15}$
	9	$\frac{2}{3}$	$\frac{2}{3}$	$\frac{2}{3}$
	-21	$\frac{6}{7}$	$\frac{2}{7}$	$\frac{2}{7}$
	45	$\frac{2}{5}$	$\frac{2}{5}$	$\frac{2}{15}$
	-81	$\frac{2}{9}$	$\frac{2}{9}$	$\frac{2}{9}$

The numerical values of D_k, α_k, $||I_k||$, and $\langle I_k \rangle$ are listed in Table I. The question we would like to be able to answer from the numerical results is as follows: Which terms, I_4, I_5, and I_6, or I_7, I_8, and I_9, should be grouped together with I_2 and I_3 to form the leading correction to $u_3(r) = \epsilon_4(r) = \frac{1}{2}I_1(r)$? Unfortunately, the answer does not seem clear. The functions $I_4(r)$ and $I_5(r)$ are larger than $I_7(r)$, $I_8(r)$, and $I_9(r)$ at small values of r but smaller at large values of r. In particular, norms of I_4, I_5, I_7, I_8, and I_9 are of the same order of magnitude. However, the following several observations can be made that slightly favor the μ expansion scheme.

(1) Magnitudes of $\langle I_7 \rangle$, $\langle I_8 \rangle$, and $\langle I_9 \rangle$ are all larger than any of those of $\langle I_4 \rangle$, $\langle I_5 \rangle$, and $\langle I_6 \rangle$.

(2) While I_7, I_8, and I_9 all contribute nearly equally in many respects, the contributions from I_6 are consistently much smaller than those from I_4 and I_5. Therefore, validity of grouping I_6 as the same order as I_2, I_3, I_4, and I_5 appears questionable.

(3) If the n-particle correction term in the density expansion is written as

$$\epsilon_n(r) = \sum_{\lambda=\lambda_{\min}}^{\lambda_{\max}} E_{n\lambda}(r)\mu^{\lambda} , \tag{A4}$$

it can be shown that

$$\lambda_{\max}=\tfrac{1}{2}(n-1)(n-2)\ , \tag{A5}$$

$$\lambda_{\min}=\begin{cases}\tfrac{1}{2}(n+2), & n:\ \text{even}\\ \tfrac{1}{2}(n+3), & n:\ \text{odd}\end{cases}\ . \tag{A6}$$

For large n, $\lambda_{\max}-\lambda_{\min}$ varies as $\frac{1}{2}n^2$ and hence the difficulty mentioned in (2) will become progressively more serious as n becomes larger.

(4) If the λth-order correction term in the μ expansion is written as

$$u_\lambda(r)=\sum_{n=n_{\min}}^{n_{\max}} U_{\lambda n}(r)\ , \tag{A7}$$

with n representing the number of particles in the cluster diagrams, it is not difficult to show that

$$n_{\max}=2\lambda-2 \tag{A8}$$

and $n_{\min}$ is an integer such that

$$\tfrac{1}{2}[(8\lambda+1)^{1/2}+3]\le n_{\min}<\tfrac{1}{2}[(8\lambda+1)^{1/2}+5]\ . \tag{A9}$$

For $\lambda=4$, it is seen that the six-particle clusters I_7, I_8, and I_9 make smaller contributions than those from the five-particle clusters I_2 and I_3. This suggests a possibility that if λ becomes large, the ordering may suffer from a difficulty similar to that pointed out in (2) and (3). However, for large λ, $n_{\max}-n_{\min}\approx 2\lambda$ and therefore the problem would not be as serious as in the density expansion procedure.

APPENDIX B: CLUSTER INTEGRALS OF $p_m^{(3)}(1,2,3)$ IN THE GAUSSIAN MODEL

The cluster integrals appearing in the expressions of $A_m^{(3)}(1,2,3)$ and $p_m^{(3)}(1,2,3)$ can be evaluated analytically if the function $h(r)$ is given by the Gaussian form of Eq. (75). The results of integration are always given explicitly in the form of

$$I(1,2,3)\equiv\frac{1}{D\,|D|^{1/2}}\exp(-\alpha s_{23}^2-\beta s_{31}^2-\gamma s_{12}^2)\ . \tag{B1}$$

Table II lists numerical values of D, α, β, and γ for $m=2$, 3, and 4.

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