A Diagrammatic Formulation of the Kinetic Theory of Fluctuations in Equilibrium Classical Fluids. I. The Fluctuation Basis and the Cluster Properties of Associated Functions†

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This is the first of a series of papers that presents a kinetic theory of fluctuations in equilibrium classical fluids that makes extensive use of diagrammatic techniques in its development and that will facilitate the use of diagrammatic techniques in the derivation of approximate kinetic theories. We develop the theory for atomic liquids, but the results are easily generalizable to molecular liquids. The fundamental fluctuating quantity in the theory is $f(\mathbf{R},\mathbf{P})$, the density of particles (atoms) at a point in single-particle phase space. The time correlation function for fluctuations of this quantity from its average, $\langle \delta f(\mathbf{R},\mathbf{P};t)\delta f(\mathbf{R}',\mathbf{P}';t')\rangle$, is the most basic correlation function of concern in generalized kinetic theories of fluctuations in liquids. In this paper, we investigate the properties of a basis set of vectors for the Hilbert space of classical dynamical variables that was suggested by Gross, Boley, and Lindenfeld. In later papers, this basis set, which we call the "fluctuation basis", will be used to construct a diagrammatic theory for this correlation function, its generalizations, and its memory function.

1. Introduction

The kinetic theory of fluctuations in equilibrium systems is an important component of the much larger subject of the kinetic theory of liquids. Much of its importance derives from the fact that transport coefficients for a system close to equilibrium can be expressed in terms of equilibrium time correlation functions for fluctuations; moreover, the scattering cross sections for such experiments as neutron scattering and light scattering can be expressed in terms of such time correlation functions.

Several theoretical techniques used to investigate and describe fluctuations were introduced by Mori.[1,2] These include the Hilbert space of dynamical variables for a classical system, memory functions, projection operators, and the continued fraction representation of correlation functions. Much of the subsequent work on the dynamics of fluctuations in liquids made extensive use of Mori's ideas.[3−5] In particular, the use of memory functions as a theoretical construct to describe fluctuations was adopted even in work that did not make use of the other techniques.

One line of development was the derivation of exact and approximate memory functions for the kinetic equation that describes the time evolution of the correlation function of the density in single-particle phase space. This density is often called $f(\mathbf{R},\mathbf{P};t)$, and the correlation function of interest is $\langle f(\mathbf{R},\mathbf{P};t)f(\mathbf{R}',\mathbf{P}';0)\rangle$, where the angular brackets denote an appropriate equilibrium ensemble average. Akcasu and Duderstadt[6] derived a weak coupling kinetic equation for this correlation function using projection operator techniques. ("Weak coupling" refers to use of low-order perturbation theory in the interparticle interaction.) They also derived the linearized Boltzmann equation for long wavelength fluctuations at low densities. Forster and Martin[7] obtained a weak coupling expression for the memory function without the use of projection operators.

The most extensive formal development along this line was that of Mazenko.[8,9] Using Hilbert space ideas and the language of memory functions (but not projection operators), he derived formal expressions for the memory function of the phase space density correlation function of an atomic fluid in terms of a function G, which is a function of four positions and momenta and one time interval. He also derived formally exact expressions to describe the time evolution of G. One of Mazenko's goals, which he achieved, was to express the dynamics in a "fully renormalized" form, which in this context means that the quantities in the kinetic equation are expressed in terms of static correlation functions of the fluid without direct reference to the interparticle potential.

The formal development of Mazenko's theory is straightforward but complex. However, the a priori motivation for some of the procedures is far from obvious. Boley[10] and Lindenfeld[11] subsequently showed that much of the structure of Mazenko's theory can be obtained by introducing a specific set of basis functions for the Hilbert space of dynamical variables and constructing projection operators using this basis set. The procedures that they presented are generalizations of that of Akcasu and Duderstadt.[6] Boley and Lindenfeld clarified the relationship between the work of Mazenko and that of Gross,[12−14] who had developed another version of a renormalized kinetic theory. They also clarified the relationship between the dynamics of Mazenko's G function and the type of dynamics that appears in projection operator theories. The kinetic theory of Sjögren[15] uses the same basis functions but relies on a time-dependent projection operator technique.

Mazenko's formally exact results are the foundation for a number of subsequent developments of approximate theories. Mazenko[8] derived a generalized Enskog equation and a mode-coupling approximation. Sjögren and Sjölander[15−20] used Mazenko's work to develop a theory that described binary collisions and mode coupling. This theory is the microscopic basis for

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the development of extended mode-coupling theory by Götze and Sjögren.[21] (Mode-coupling theories have also been derived in various other ways, mostly using projection operators.[22])

In a series of papers, of which this is the first, we will show how the basic insights of Gross, Boley, and Lindenfeld about the choice of a basis set for the Hilbert space can be used to derive a formally exact kinetic theory of fluctuations in liquids that has the features of Mazenko's theory but has the additional advantage that each correlation function and memory function of interest is expressed in terms of a diagrammatic series. The diagrams are similar to the Mayer cluster diagrams that appear in the equilibrium theory of fluids. The diagrammatic formulation is a special case of a more general diagrammatic method for classical systems developed by the author[23] based on the ideas of Martin, Siggia, and Rose.[24] Because of the simplifying features of the Gross/Boley/Lindenfeld basis, the full formal apparatus of this method is not needed. All of the results needed are derived independently in the present paper.

A diagrammatic formulation such as this one has several advantages. The dynamics of both the correlation functions and the memory functions are expressed in terms of the same types of diagrams. The assumptions underlying the various versions of mode-coupling theory are specific types of relationships between the two types of functions, and the diagrammatic theory is of a form that will facilitate the derivation of mode-coupling approximations and corrections to mode-coupling theories. Another important feature is that the diagrammatic language provides a convenient way of defining and manipulating functions with many arguments, just as Mayer's cluster theory provides a unified description of correlation functions involving any number of positions in space. Moreover, the diagrams provide a convenient language for discussing the cluster properties of functions of several arguments, just as Mayer's cluster theory provides a convenient language for discussing cumulants.

Section 2 defines what we call the "fluctuation basis", the definition of which is a minor variation of definitions used by Gross, by Boley and by Lindenfeld. It also discusses the process by which the basis is constructed, a process that assumes the existence of certain functions related to equilibrium static correlation functions of the density and the existence of their inverse functions. In section 3, we prove several theorems about the existence of these functions and about their cluster properties. These theorems are basic to the development of the cluster theory to be presented in later papers of this series. In particular, in the development of the cluster theory, the existence and properties of these functions must be known for correlations among arbitrarily many points in single-particle phase space.

2. The Fluctuation Basis

The physical system of interest is an equilibrium fluid of point particles with a pairwise additive interparticle potential in a volume V at temperature T with chemical potential μ. In this section, we state the basic equations for the system, define a Hilbert space of dynamical variables, and then construct the fluctuation basis for that Hilbert space.

2.1. The System of Interest. The mechanical state of the system is specified by the number of particles N and the positions and momenta $(\mathbf{r}^N,\mathbf{p}^N)$ of the particles. Here $\mathbf{r}^N$ denotes the set of positions $\mathbf{r}_1,\ldots,\mathbf{r}_N$ of each of the N particles, with a similar meaning for $\mathbf{p}^N$. The Hamiltonian is

$$H_N(\mathbf{r}^N,\mathbf{p}^N) = \sum_{i=1}^{N} \mathbf{p}_i\cdot\mathbf{p}_i/(2m) + \sum_{i<j=1}^{N} u(|\mathbf{r}_i - \mathbf{r}_j|)$$

Any function of N, $\mathbf{r}^N$, and $\mathbf{p}^N$ will be called a dynamical variable. If $A(N,\mathbf{r}^N,\mathbf{p}^N)$ is a dynamical variable, its grand canonical ensemble average is

$$\langle A\rangle \equiv \sum_{N=0}^{\infty} \int_V d\mathbf{r}^N \int_{-\infty}^{\infty} d\mathbf{p}^N\, P(N,\mathbf{r}^N,\mathbf{p}^N) A(N,\mathbf{r}^N,\mathbf{p}^N) \quad (1)$$

where

$$P(N,\mathbf{r}^N,\mathbf{p}^N) \propto \frac{1}{N!}\, e^{N\mu/(kT)} \exp(-H_N(\mathbf{r}^N,\mathbf{p}^N)/(kT))$$

The Liouville operator L is given by

$$iL = \sum_{i=1}^{N} ([\nabla_{\mathbf{p}_i} H]\cdot\nabla_{\mathbf{r}_i} - [\nabla_{\mathbf{r}_i} H]\cdot\nabla_{\mathbf{p}_i})$$

The set of all dynamical variables (i.e., the set of all functions of N, $\mathbf{r}^N$, $\mathbf{p}^N$) can be regarded as a vector space. We use Dirac's bra-ket notation, so the dynamical variable $A(N,\mathbf{r}^N,\mathbf{p}^N)$ corresponds to the vector $|A\rangle$. We define an inner product of two vectors $|B\rangle$ and $|A\rangle$ as

$$\langle B|A\rangle \equiv \langle BA\rangle$$

The brackets on the left are a bra and a ket, whereas on the right the brackets denote an ensemble average defined using eq 1. With this choice of inner product, the Liouville operator is Hermitian.

Most of the dynamical variables in the current theory are labeled by points in single-particle phase space. For example, the density in single-particle phase space is

$$f(\mathbf{R}_1,\mathbf{P}_1;N,\mathbf{r}^N,\mathbf{p}^N) \equiv \sum_{i=1}^{N} \delta(\mathbf{R}_1 - \mathbf{r}_i)\delta(\mathbf{P}_1 - \mathbf{p}_i) \quad (2)$$

The pair of quantities $\mathbf{R}_1$ and $\mathbf{P}_1$ will be called a phase-point variable or phase point. We shall use the convention that an integer is used as an abbreviation for a phase point. Thus, for example, 1 is an abbreviation for $(\mathbf{R}_1,\mathbf{P}_1)$, and $2'$ is an abbreviation for $(\mathbf{R}_2',\mathbf{P}_2')$, etc. Similarly, a lower case letter is used as an abbreviation for the position and momentum of a particle; for example, i is an abbreviation for $(\mathbf{r}_i,\mathbf{p}_i)$. Thus, eq 2 will be written more compactly as

$$f(1;N,\mathbf{r}^N,\mathbf{p}^N) = \sum_{i=1}^{N} \delta(1;i)$$

where $\delta(1;i) \equiv \delta(\mathbf{R}_1 - \mathbf{r}_i)\delta(\mathbf{P}_1 - \mathbf{p}_i)$.

We use f to construct more complicated functions that are products of f factors.

$$\psi_2(12) = f(1)f(2) \quad (3)$$

$$\psi_n(12\ldots n) = f(1)f(2)\ldots f(n) \quad (4)$$

and so on. For completeness, we also define

$$\psi_0 = 1 \quad (5)$$

$$\psi_1(1) = f(1) \quad (6)$$

Note that each of these functions is a dynamical variable, that is, a function of N, $\mathbf{r}^N$, and $\mathbf{p}^N$, but that dependence is not indicated explicitly. (The subscript on a function indicates the number of phase-point arguments. Commas between phase-point arguments will often be omitted if there is no possibility of confusion.)

2.2. Construction of the Fluctuation Basis. We will define the fluctuation basis by a procedure that is a small modification of those used by Gross, Boley, and Lindenfeld. It is similar to the Schmidt orthogonalization procedure often used in quantum mechanics. The basis is a set of vectors, $|\phi_0\rangle$, $|\phi_1(1)\rangle$, $|\phi_2(12)\rangle$, etc., with the same numbers and types of arguments as the $|\psi\rangle$ vectors. The defining procedure is a recursive one.

First, consider $n = 0$. We define $|\phi_0\rangle \equiv |\psi_0\rangle$. We define $\mathcal{S}_0$ to be the subspace of the Hilbert space that is spanned by $|\phi_0\rangle$. Let P_0 be the projection operator onto that subspace. (An explicit representation of this projection operator can easily be constructed, so the existence of P_0 is assured.)

Next, consider $n = 1$. We define $|\phi_1(1)\rangle$ to be the projection of $|\psi_1(1)\rangle$ orthogonal to $\mathcal{S}_0$.

$$|\phi_1(1)\rangle \equiv (1 - P_0)|\psi_1(1)\rangle$$

Let $\mathcal{S}_1$ be the subspace spanned by the $|\phi_1(1)\rangle$ vectors for all values of the argument. Let P_1 be the projection operator onto $\mathcal{S}_1$. (This projection operator can also be constructed explicitly, using the direct correlation function of the fluid.)

Now, consider a general value of $n \geq 2$. Assume that subspaces $\mathcal{S}_m$ and projection operators P_m have been defined for $0 \leq m \leq n - 1$. We define $|\phi_n(1...n)\rangle$ to be the projection of $|\psi_n(1...n)\rangle$ orthogonal to $\mathcal{S}_0$, $\mathcal{S}_1$, ..., $\mathcal{S}_{n-1}$.

$$|\phi_n(1...n)\rangle \equiv (1 - P_0 - P_1 - ... - P_{n-1})|\psi_n(1...n)\rangle \quad (7)$$

Let $\mathcal{S}_n$ be subspace spanned by the $|\phi_n(1...n)\rangle$ vectors for all values of its n arguments. Let P_n be the projection operator onto $\mathcal{S}_n$. (The existence of these projection operators for $n \geq 2$ will be discussed more fully below.) This provides a recursive definition of $|\phi_n\rangle$, $\mathcal{S}_n$, and P_n for all $n \geq 0$.

Before proceeding further, a few comments about this procedure and the functions it generates are in order. (1) The various subspaces $\mathcal{S}_n$ are orthogonal to one another. (2) The basis set is not a complete basis for the Hilbert space. However, it spans a large enough subspace of the entire Hilbert space that it is adequate for the calculation of time correlation functions of the ψ functions. (3) We do not need to be concerned with the possibility that the recursive procedure might terminate.[25] (4) The ensemble average $\langle\phi_n\rangle$ of each basis function is zero, because $0 = \langle\phi_0|\phi_n(1...n)\rangle = \langle\phi_0\phi_n(1...n)\rangle = \langle\phi_n\rangle$ for $n \geq 1$. A nonzero value of ϕ_n ($n \geq 1$) represents a fluctuation of the multipoint densities from their ensemble average values. Hence, we call this the fluctuation basis. We shall refer to the subscript n as the fluctuation index. (5) These functions represent the fluctuations in a very specific way. The function $f(1) - \langle f(1)\rangle = f(\mathbf{R}_1,\mathbf{P}_1) - \langle f(\mathbf{R}_1,\mathbf{P}_1)\rangle = \phi_1(1)$ is usually used to represent density fluctuations at a point in single-particle phase space. As a result of the construction,

$$f(1)f(2) - \langle f(1)f(2)\rangle = (\text{a vector in } \mathcal{S}_1) + \phi_2(12)$$

The left side is the fluctuation of the two-point density from its average. The first term on the right is a linear combination of single-point fluctuations. Thus, in a sense, ϕ_2 describes the part of the fluctuation of the two-point density that cannot be accounted for by (i.e., that is not in the subspace spanned by) the set of all single-point density fluctuations. A similar interpretation can be given to the other basis functions. For example,

$$f(1)f(2)f(3) - \langle f(1)f(2)f(3)\rangle = (\text{a vector in } \mathcal{S}_1) + (\text{a vector in } \mathcal{S}_2) + \phi_3(123)$$

2.3. Existence and Construction of the Projection Operators. We now address the question of the existence and construction of the projection operators. Because the inner product of ϕ_0 with itself is unity, $\langle\phi_0|\phi_0\rangle = \langle 1\rangle = 1$, the projection operator that projects onto S_0 can clearly be represented as

$$P_0 = |\phi_0\rangle\langle\phi_0| \quad (8)$$

For $n \geq 1$, the matrix of inner products of the $|\phi_n\rangle$ vectors will be called F_n.

$$F_n(1...n;1'...n') = \langle\phi_n(1...n)|\phi_n(1'...n')\rangle \quad (9)$$

For functions similar to $F_n(1...n;1'...n')$, with two sets of arguments, the first set (here, $1...n$) will be called the "left arguments" and the second set (here, $1'...n'$) will be called the "right arguments'. Because the ψ_n vectors are symmetric under permutation of their arguments, the same holds for the ϕ_n vectors. It follows that the F_n function defined in eq 9 is a symmetric function of its left arguments and a symmetric function of its right arguments. It is also clearly symmetric under interchange of left and right arguments, because the basis functions are real. For brevity, we shall refer to these three types of symmetry as left symmetry, right symmetry, and left−right symmetry.

A possible representation of the projection operator that projects onto the subspace spanned by a set of vectors $|\phi_n\rangle$ is[26]

$$P_n = \frac{1}{(n!)^2}\int \mathrm{d}1''...\mathrm{d}n''\ \mathrm{d}1'''...\mathrm{d}n'''\ |\phi_n(1''...n'')\rangle K_n(1''...n'';1'''...n''')\langle\phi_n(1'''...n''')| \quad (10)$$

It is straightforward to show that this operator is a projection operator and it projects onto $\mathcal{S}_n$ if K_n is a function that is a matrix inverse of F_n in the sense that

$$\frac{1}{n!}\int \mathrm{d}1''...\mathrm{d}n''\ K_n(1...n;1''...n'')F_n(1''...n'';1'...n') = \mathbf{I}(1...n;1'...n') \quad (11)$$

Here

$$\mathbf{I}(1...n;1'...n') = \sum_{\mathscr{P}(1'...n')} \mathscr{P}[\delta(1,1')\delta(2,2')...\delta(n,n')] \quad (12)$$

where $\mathscr{P}(1'...n')$ is a permutation of the arguments $1'...n'$ and the sum is over all $n!$ distinct permutations.

The existence of such an inverse is not a trivial matter and should not be assumed without careful consideration. For example, there exist functions closely related to the F_n for which inverses do not exist.[27] Moreover, the inverse K_1 exists when the basis functions are defined in the grand canonical ensemble for finite V but not when defined in the canonical ensemble.[28] The existence of an inverse for a function, in the sense defined above, is closely related to the cluster properties of a function. By "cluster properties", we mean the behavior of a function of several variables when some or all of the position variables are far from the others. Thus, the question of the existence of K_n

for each n is associated with the question of the existence of F_n for the same value of n (and smaller values) and with the cluster properties of F_n.

In the construction of a kinetic theory using these basis functions, more must be known about the functions K_n and F_n than their existence and cluster properties. Their symmetry properties (e.g., left, right, and left−right) are also worthwhile knowing. Also, it is worthwhile knowing whether K_n is a left inverse of F_n as well as a right inverse; that is, whether the following equation holds.

$$\frac{1}{n!}\int \mathrm{d}1''...\mathrm{d}n''\, F_n(1...n;1''...n'')K_n(1''...n'';1'...n') = \mathbf{I}(1...n;1'...n') \quad (13)$$

For finite dimensional matrices, a left inverse is also a right inverse, but the same statement does not always apply to "matrices" with continuous "indices". Finally, for a theory of the generality that we seek to develop, we need to know these properties of F_n and K_n for arbitrary values of n.

The rest of this paper is devoted to an exploration of these questions of the existence, cluster properties, and symmetry properties of functions such as F_n and K_n that are used when applying the fluctuation basis to the development of a kinetic theory. The arguments that we present will be inductive, because, for example, the existence and properties of K_n depend on the existence and properties of F_n, which cannot be defined without P_{n-1}, the existence of which depends on the existence and properties of K_{n-1}. Diagrammatic methods will be especially useful for stating and deriving the conclusions that we reach for arbitrary n.

3. Theorems on the Existence and Properties of Various Functions

3.1. Introduction. In this section, we prove the existence of, as well derive some properties of, the F_n and K_n functions and other functions that arise from the fluctuation basis, including their cluster properties and some of their symmetry properties. The discussion makes extensive use diagrammatic methods. See Appendix A for a summary of the diagrammatic terminology that we use.

The basis for proving the cluster properties of these functions is the corresponding properties of the fundamental moments of the density. Let

$$m_n(1...n) \equiv \langle \psi_n(1...n)\rangle = \langle f(1)...f(n)\rangle$$

Each of these moments can be expressed in terms of Maxwell−Boltzmann distribution function for the momentum arguments and equilibrium moments of the density in position space. The cumulants for these moments will be denoted $m_n^{(c)}(1...n)$. The definition of the cumulants is recursive.

$m_n^{(c)}(1...n) \equiv m_n(1...n) -$ (the sum of all topologically different diagrams with (i) n roots labeled $1...n$ and (ii) two or more $m_p^{(c)}$ vertices)

which implies that

$m_n(1...n) =$ the sum of all topologically different diagrams with (i) n roots labeled $1...n$ and (ii) $m_p^{(c)}$ vertices (14)

(See Figure 1). These cumulants have what we shall call "the cluster property". A function of two or more field points is said to have the cluster property if the function is zero when one (or more) of the arguments is (are) separated from all of the other arguments by a distance that is many multiples of the correlation length of the fluid. Only the position components (not the momentum components) of the arguments are relevant for this definition.

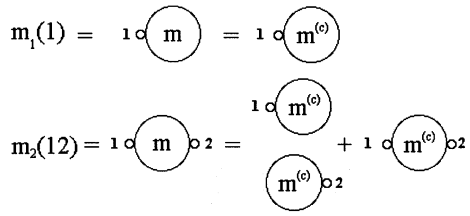

Figure 1. Cumulant representation of the first two density moments, $m_1(1)$ and $m_1(12)$ (see eq 14). Each small open circle is a root point. These diagrams have no free points. Each large circle with a symbol inside is a vertex. The name of the vertex is indicated by the symbols inside, and the subscripts can be inferred from the number of points. For example, the first vertex in the first equation is $m_1(1)$. The last vertex in the second equation is $m_2^{(c)}(12)$. This is one of the few diagrammatic expressions in this paper that do not need to make a distinction between left and right points.

3.2. Preliminary Discussion. The discussion will be highly inductive, in the sense that the important theorem will be based on inductive reasoning and many of the theorems leading up to it involve multiple layers of inductive logic. It will help to develop symbols for various types of assertions. The first of these is the following.

Definition. Let $\mathcal{A}_\phi(M)$ be the assertion that the projection operators P_n exist for $0 \le n \le M-1$ and that hence the vectors $|\phi_n\rangle$ exist in the Hilbert space for $0 \le n \le M$. In this subsection, we shall assume that this assertion is correct for some value of M and explore the consequences. This will lead to a theorem that is stated at the end of this subsection.

The first consequence is that we can define the following inner products of the basis functions,

$$F_n(1...n;1...n) \equiv \langle \phi_n(1...n)|\phi_n(1'...n')\rangle$$

and

$$J_{nm}(1...n;1'...m') \equiv \langle \phi_n(1...n)|\psi_m(1'...m')\rangle$$

for all $0 \le n \le M$ and $0 \le m < \infty$. Moreover,

$$J_{nm}(1...n;1'...m') = 0 \quad \text{for} \quad n > m \quad (15)$$

$$F_n(1...n;1'...n') = J_{nn}(1...n;1'...n') \quad (16)$$

These two results follow from the orthogonality properties that the construction process builds into the ϕ functions. The symmetry of ψ_n under permutation of its arguments implies that the ϕ_n are also symmetric. It follows that F_n and J_{nm} are left symmetric and right symmetric. Moreover, F_n is left−right symmetric, because the basis functions are real.

Then, we can define cluster functions (analogous to cumulants) associated with J. The cluster functions will be called $J_{nm}^{(c)}$ and will be defined recursively for $0 \le n \le M$, $m \ge 0$, and $n + m \ge 1$.

$J_{nm}^{(c)}(1...n;1'...m') \equiv J_{nm}(1...n;1'...m') -$ (the sum of all topologically different matrix diagrams with (i) n left roots labeled $1...n$, (ii) m right roots labeled $1'...m'$, and (iii) two or more $J_{pq}^{(c)}$ vertices for $p + q \ge 1$) (17)

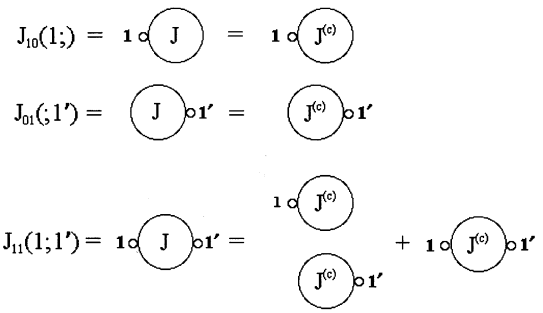

Figure 2. Cluster representation of the first few J matrix elements, $J_{10}(1;)$, $J_{01}(;1')$, and $J_{11}(1;1')$ (see eq 18). Note that these diagrams are drawn with the left roots on the left of the diagram and the right roots at the right.

It follows that

$J_{nm}(1...n;1'...m') =$ the sum of all topologically different matrix diagrams with (i) n left roots labeled $1...n$, (ii) m right roots labeled $1'...m'$, and (iii) $J^{(c)}_{pq}$ vertices (18)

(See Figure 2).

Lemma. $\mathcal{A}_\phi(M)$ implies that

$$J^{(c)}_{nm}(1...n;1'...m') = 0 \quad \text{for} \quad 0 \le m < n \le M \tag{19}$$

[This follows by induction from the corresponding property of the J_{nm}; see eq 15. The proof (details omitted) is based on the fact that the definition of $J^{(c)}_{nm}$ contains $J^{(c)}_{pq}$ but only for $p + q < n + m$ and $p \le n$.]

We can apply the cluster representation above to J_{nn}. Recognizing that this is equal to F_n, we have, for $1 \le n \le M$,

$F_n(1...n;1'...n') =$ the sum of all topologically different matrix diagrams with (i) n left roots labeled $1...n$, (ii) n right roots labeled $1'...n'$, and (iii) $J^{(c)}_{pq}$ vertices

Any diagram in this series containing a $J^{(c)}_{pq}$ with $p < q$ must contain another $J^{(c)}_{rc}$ with $r > s$ and hence the diagram would be zero, according to eq 19. Hence, the only nonzero diagrams are those in which all of the $J^{(c)}_{pq}$ have $p = q$. Let us define $F^{(c)}_n(1...n;1'...n') = J^{(c)}_{nn}(1...n;1'...n')$. Then, we have, for $1 \le n \le M$,

$F_n(1...n;1'...n') =$ the sum of all topologically different matrix diagrams with (i) n left roots labeled $1...n$, (ii) n right roots labeled $1'...n'$, and (iii) $F^{(c)}_p$ vertices (20)

This is a cumulant-like expression for the F_n. It can be used to obtain a recursion formula for the $F^{(c)}_n$ in terms of the F_n and to prove the following lemma.

Lemma. $\mathcal{A}_\phi(M)$ implies that $F^{(c)}_n$ has left symmetry, right symmetry, and left−right symmetry. We summarize the results of this subsection in the following theorem.

Theorem. $\mathcal{A}_\phi(M)$ implies that (i) the functions F_n and J_{nm} exist for $0 \le n \le M$, (ii) the function $F^{(c)}_n$ exists for $1 \le n \le M$, (iii) the function $J^{(c)}_{nm}$ exists for $0 \le n \le M$ and $n + m \ge 1$, (iv) each of these functions has left symmetry and right symmetry, and (v) the functions with one subscript in their names have left−right symmetry.

3.3. Additional Definitions. *Definition.* Let $\mathcal{A}_{Jc}(M)$ be assertion $\mathcal{A}_\phi(M)$ plus the assertion that $J^{(c)}_{nm}$ has the cluster property for $0 \le n \le M$ and $n + m \ge 2$.

Definition. Let $\mathcal{A}_{Fc}(M)$ be assertion $\mathcal{A}_\phi(M)$ plus the assertion that $F^{(c)}_n$ has the cluster property for $1 \le n \le M$.

Definition. Let $\mathcal{A}_K(M)$ be assertion $\mathcal{A}_\phi(M)$ plus the assertion that for $0 \le n \le M$ the following are true: (i) $K_n(1...n;1'...n')$ exists; (ii) K_n is an inverse of F_n in the sense of eq 11 (i.e., it is a left inverse); (iii) $K_n(1...n;1'...n')$ also is a right inverse in the sense of eq 13; (iv) K_n is left−right symmetric and (for $n > 1$) is left symmetric and right symmetric.

Suppose that $\mathcal{A}_K(M)$ is correct for some $M \ge 1$. We define cluster functions $K^{(c)}$ that have the same relationship to K as $F^{(c)}$ has to F. Namely, for $1 \le n \le M$, we define

$K^{(c)}_n(1...n;1'...n') = K_n(1...n;1'...n') -$ the sum of all topologically different matrix diagrams with (i) n left roots labeled $1...n$, (ii) n right roots labeled $1'...n'$, and (iii) two or more $K^{(c)}_p$ vertices

It follows that

$K_n(1...n;1'...n') =$ the sum of all topologically different matrix diagrams with (i) n left roots labeled $1...n$, (ii) n right roots labeled $1'...n'$, and (iii) $K^{(c)}_p$ vertices (21)

Lemma. $\mathcal{A}_K(M)$ implies that, for $1 \le n \le M$, $K^{(c)}_n$ exists and has left symmetry, right symmetry, and left−right symmetry. (This follows from the previous discussion.)

Definition. Let $\mathcal{A}_{Kc}(M)$ be the assertion $\mathcal{A}_K(M)$ plus the assertion that $K^{(c)}_n$ has the cluster property for all $1 \le n \le M$.

3.4. Main Theorem. With these definitions in place, we can now proceed to the major theorem of this paper.

Lemma. $\mathcal{A}_K(M-1)$ implies $\mathcal{A}_\phi(M)$. (This is straightforward, because $\mathcal{A}_K(M-1)$ implies the existence of all the K functions and projection operators needed to define ϕ_M.)

Lemma. $\mathcal{A}_{Jc}(M)$ implies $\mathcal{A}_{Fc}(M)$. (This follows from eq 16.)

Theorem. For $M \ge 2$, $\mathcal{A}_{Fc}(M)$ and $\mathcal{A}_{Kc}(M-1)$ imply $\mathcal{A}_{Kc}(M)$. (See Appendix B.2 for the rather lengthy proof.)

Theorem. For $M \ge 2$, $\mathcal{A}_{Jc}(M-1)$ and $\mathcal{A}_{Kc}(M-1)$ imply $\mathcal{A}_{Jc}(M)$. (See Appendix B.3 for the rather lengthy proof.)

Lemma. $\mathcal{A}_\phi(1)$, $\mathcal{A}_{Jc}(1)$, $\mathcal{A}_{Fc}(1)$, $\mathcal{A}_K(1)$, and $\mathcal{A}_{Kc}(1)$ are true. (See Appendix B.4 for the proof.)

Theorem. $\mathcal{A}_\phi(M)$, $\mathcal{A}_{Jc}(M)$, $\mathcal{A}_{Fc}(M)$, and $\mathcal{A}_{Kc}(M)$ are true for all M. (This follows by induction from the theorems and lemmas above.)

This last theorem is the major result of this paper. Its implications are summarized below.

4. Summary and Conclusions

We can summarize the main theorem and some of the associated results in the following statements, which hold for *all* values of n (unless otherwise noted). (i) The fluctuation basis vectors $|\phi_n(1...n)\rangle$ exist. (ii) The following functions exist: $J_{nm} \equiv \langle\phi_n|\psi_m\rangle$; $F_n \equiv \langle\phi_n|\phi_n\rangle$; and K_n, the inverse function to F_n. Moreover, $J_{nn} = F_n$. Also, $J_{nm} = 0$ for $n > m$. (iii) The cluster functions associated with these functions also exist: $J^{(c)}_{nm}$ for $n + m \ge 1$; and $F^{(c)}_n$ and $K^{(c)}_n$ for $n \ge 1$. The relationships between the cluster functions and the functions from which they are derived are eqs 18, 20, and 21. Moreover $J^{(c)}_{nm} = 0$ for $n > m$. (iv) Each of the functions is left symmetric and right

symmetric. (v) Each of the functions with one subscript is left−right symmetric. (vi) Each function with a superscript (c) in its name has the cluster property. (vii) K_n is both a left inverse and a right inverse of F_n, in the sense of eqs 11 and 13. (viii) The projection operator P_n exists.

Several specific instances of these results for various small values of n have previously been obtained by Gross,[12] Boley,[10] Lindenfeld,[11] and Mazenko,[8,9] but these general results and the graphical method used to obtain them are new. This paper lays the groundwork for a kinetic theory of fluctuations in fluids that exploits the insights of Gross, Boley, and Lindenfeld about the usefulness of the fluctuation basis and that utilizes graphical techniques extensively in the formal development. We have shown that the members of the fluctuation basis are well defined and have derived symmetry properties as well as the cluster properties of several functions that are inner products involving the fluctuation basis. The proofs apply to all values of the fluctuation index n rather than being restricted to small n. In the next paper in this series, we will derive the equations of motion for the time-dependent versions of the fluctuation basis vectors and show the close relationship of the resulting theory to the fully renormalized kinetic theory of Mazenko. In the third paper, we will exploit graphical techniques to perform a cluster analysis of the renormalized interactions a develop a form of the theory that is useful for the graphical derivation of mode-coupling approximations.

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Appendix A. Summary of Diagrammatic Terminology

The diagrammatic formulation that we will use is closely analogous to those used in the equilibrium cluster theory of classical fluids.[5,29−33] (It is related to diagrammatic formulations often used to discuss critical phenomena in classical statistical mechanics.[34−36]) The diagrams have rooted points (which we shall call "roots"), represented by open circles that are labeled with variables, free points, represented by closed circles that are not labeled, vertices with one or more points on them, represented by closed geometric figures (often a large circle) with points on its boundary, which correspond to functions of the variables associated with their points, and bonds between pairs of points, represented by lines, which correspond to functions of the variables assigned to their points. (Bonds are not needed in this first paper, but they will be used in later papers in the series.) Each diagram has a value that is a function of the labels on the root points. In evaluating a diagram, dummy variables are assigned to the free points and these variables are integrated over.

An important characteristic of a vertex is the topological equivalence, or lack thereof, of its points, which is a reflection of the symmetry of the function that corresponds to the vertex. Some vertices, such as m_n vertices, represent functions, for example, $m_n(1...n)$, that are totally symmetric under permutation of their n arguments. Accordingly, all n points are regarded as topologically equivalent. Some vertices, such as J_{nm}, represent functions with left and right arguments, for example $J_{nm}(1...n;1'...m')$. In this case, the function is both left symmetric and right symmetric. To take this into account, we always draw a J_{nm} vertex with n points on the left for the left arguments and m points on the right for the right arguments. The left points are topologically equivalent to one another, and the right points are topologically equivalent to one another.[37]

Many of the graphical series of concern here represent functions with left arguments or right arguments or both and contain vertices and bonds with left points or right points or both. In these cases, the graph is usually required to satisfy the following conditions: (i) Each left root is attached to a left point of a vertex or bond and nothing else. (ii) Each right root is attached to the right point of a vertex or bond and nothing else. (iii) Each free point is attached to the right point of one vertex or bond and the left point of another vertex or bond and nothing else. When the variable associated with a free point is integrated over in evaluating the diagram, we get a structure that is much like matrix multiplication. Thus, a "matrix diagram" will be defined as one that satisfies all three of these restrictions.

We now summarize the graph theoretic definitions and conventions used throughout this paper. They are consistent with those used by several previous authors,[30−33] although different authors use slightly different terms. (1) In the diagrams, each root is labeled with a set of variables. The nature of the set depends on the specific type of graph. In the present paper, the label is always a phase point, that is, a combination of a position **R** and a momentum **P**. This is often abbreviated as an integer; for example, 1 is an abbreviation for $\mathbf{R}_1\mathbf{P}_1$, and $2'$ is an abbreviation for $\mathbf{R}_2'\mathbf{P}_2'$. (In later papers in this series, the label is a combination of a phase-point variable and a time variable, t, or a combination of several phase-point variables and a time variable.) The free points are unlabeled. Such diagrams are conventionally referred to as "unlabeled diagrams" (to indicate that the free points are unlabeled). (2) The procedure for evaluating an unlabeled diagram involves assigning a set of dummy variables to the free points. The variables that are assigned should be distinct from each other and from the labels attached to roots. A diagram with labels assigned to the free points is referred to as a "labeled diagram". (Labeled diagrams are introduced only as a device for defining topological equivalence and evaluating unlabeled diagrams.) (3) Two labeled diagrams are topologically equivalent if there is a one-to-one correspondence between the points, bonds, and vertices of the two diagrams such that corresponding points have the same labels, corresponding vertices are of the same type and have points with the same labels attached, and corresponding bonds have the same type and have points with the same labels attached. (4) The symmetry number of an unlabeled diagram is obtained by converting it to a labeled diagram and counting the number of ways of permuting the labels on the free points in such a way as to give a topologically equivalent labeled diagram. (5) The rules for evaluating an unlabeled diagram are the standard ones: convert it to a labeled graph, construct a summand/integrand containing the functions corresponding to all of the bonds and vertices in the diagram, integrate and sum over the dummy variables assigned to the free points, and divide by the symmetry number of the diagram. (6) Two unlabeled graphs are topologically equivalent if there is some way of assigning the same set of labels to the free points of each so that the resulting labeled diagrams are topologically equivalent. (7) Two unlabeled graphs are topologically different if they are not topologically equivalent.

In the present work, we use a few additional conventions and definitions. (1) The statement that a graph contains certain objects, for example, "free points" or $K^{(c)}$ vertices, means that it contains "zero or more" of the object. (2) Every point on a vertex (or bond) must be attached to a free point or a root. (3)

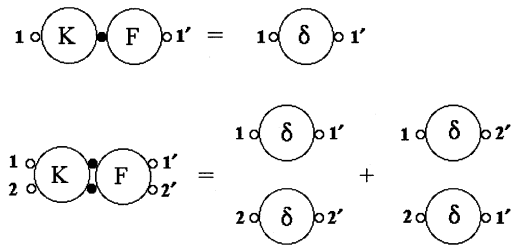

Figure 3. Graphical interpretation of the left inverse relationship between K_n and F_n for $n = 1, 2$ (see eq 22). The small filled circles are free points. The symmetry number of the diagram on the left of the second equation is 2.

The most basic topological relationship between two objects that is represented in a diagram is the attachment of a point to a vertex or bond, and this is the only type of attachment. Such attachment is what is implied when we say that a root point or field point is "on" a vertex or bond. We say that two vertices that are attached to the same point are connected (rather than attached) to one another. More generally, two objects *a* and *b* (which could be roots, free points, vertices, or bonds) in a diagram are connected if there is a sequence of objects starting with *a* and ending with *b* such that each object in the interior of the sequence is attached to the immediately preceding object and the immediately following object. (4) A diagram is connected if every object in the diagram is connected to every other object. (5) A disconnected diagram consists of two or more subgraphs. Each subgraph is connected, consists of a subset of the objects in the original graph, and contains all of the objects in the original graph to which its objects are connected.

The lemmas of Morita and Hiroike[30] and their straightforward generalizations greatly facilitate the derivation of relationships among graphical series.[32,33]

Appendix B. Proofs of Two Theorems and a Lemma

The proof of the final theorem in section 3.4 relied on two theorems and one lemma the proofs of which are given here, preceded by some lemmas.

B.1. Graphical Meaning of the Inverse Relationship. The left side of the inverse relationship eq 11 can be interpreted as the value of a graph, and the right side represents the sum of the values of a set of graphs. Thus, eq 11 is equivalent to the following graphical equation.

(the matrix diagram with (i) n left points labeled 1...n, (ii) n right points labeled 1′...n′, (iii) n free points, and (iv) one K_n and one F_n vertex such that (i) each left root is on the K_n, (ii) each right root is on the F_n, and (iii) each free point is on both the F_n and the K_n vertices) = (the sum of all topologically different matrix diagrams with (i) n left points labeled 1...n, (ii) n right points labeled 1′...n′, and (iii) n δ vertices) (22)

Because of the left symmetry and right symmetry of the F_n and K_n functions, the symmetry number of the graph on the left-hand side of this relationship is $n!$, and the left side of eq 11 thus corresponds to the value of this graph (see Figure 3).

The assertions $\mathscr{A}_{Fc}(M)$ and $\mathscr{A}_{Kc}(M)$ imply some useful consequences of this graphical equation. If we assume these assertions are true for some $M \geq 1$, then on the left side of the equation (for each value of $n \leq M$), we replace each F_n and K_n by its cluster representation. The result can be expressed conveniently in terms of diagrammatic series, using the theorems of Morita and Hiroike[30] and their straightforward generalizations. The result leads to the following lemma.

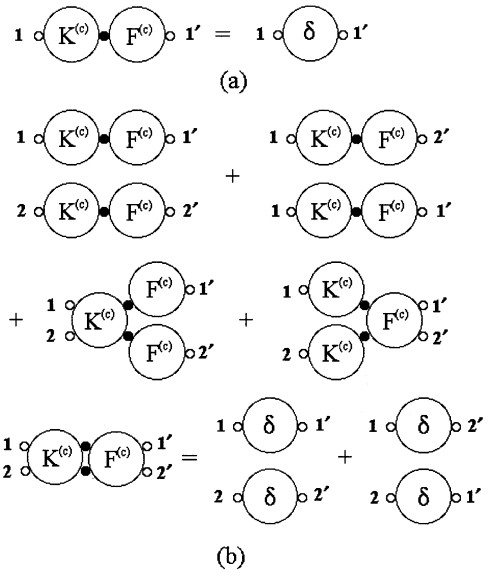

Figure 4. The left inverse relationship of Figure 3 expressed in terms of cluster functions (see eq 23). Note that when the equation in part a is used to evaluate the first term on the left-hand side of the equation in part b, the result is equal to the first term on the right-hand side of part b. Similarly, the second term on the left-hand side of part b is equal to the second term on the right-hand side of part b. Therefore, the sum of the remaining terms on the left of part b must equal zero. This gives the graphical equality in Figure 5. This is a special case of a more general result, eq 25, that is derived in a different way in the text.

Lemma. $\mathscr{A}_{Kc}(M)$ and $\mathscr{A}_{Fc}(M)$ imply, for $1 \leq n \leq M$,

(the sum of all topologically different matrix diagrams with (i) n left points labeled 1...n, (ii) n right points labeled 1′...n′, (iii) n free points, and (iv) $K_m^{(c)}$ vertices and $F_m^{(c)}$ vertices such that (i) each left root is on a $K_m^{(c)}$, (ii) each right root is on an $F_m^{(c)}$, (iii) each free point is on a $K_m^{(c)}$ and an $F_m^{(c)}$) = (the sum of all topologically different diagrams with (i) n left points labeled 1...n, (ii) n right points labeled 1′...n′, and (iii) n δ vertices) (23)

(See Figure 4).

Each of the types of vertices ($K_m^{(c)}$, $F_m^{(c)}$, and δ) that appear in this relationship has the cluster property. It follows that the cluster properties of the graphs are directly related to their connectivity. Any graph in which each root is connected to every other root represents a function that has the cluster property. If a graph is not connected, its value is a product of the values of each of its subgraphs and hence the value of a graph that is not connected is independent of the relative positions of the various sets of connected roots. Each of the subgraphs contains equal numbers of left and right roots.

It follows that for each type of connectivity among the roots, the sum of the diagrams on the left with that connectivity must equal the sum of the diagrams on the right with that connectivity. This leads to the following theorem.

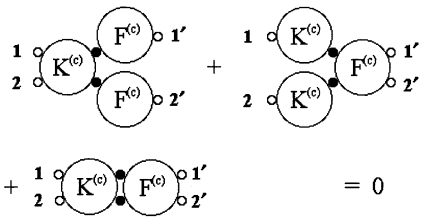

Figure 5. A consequence of the graphical version of the left inverse relationships of Figure 4 (see eq 25).

Theorem. $\mathscr{A}_{Fc}(M)$ and $\mathscr{A}_{Kc}(M)$ imply

(the matrix diagram with (i) one left point labeled 1, (ii) one right point labeled $1'...n'$, (iii) one free point, and (iv) one $K_1^{(c)}$ vertex and one $F_1^{(c)}$ vertex such that (i) the left root is on the $K_1^{(c)}$ and (ii) the right root is on the $F_1^{(c)}$) = $\delta(1;1')$ (24)

and, for $2 \leq n \leq M$,

(the sum of all topologically different connected matrix diagrams with (i) n left points labeled $1...n$, (ii) n right points labeled $1'...n'$, (iii) n free points, and (iv) $K_m^{(c)}$ vertices and $F_m^{(c)}$ vertices such that (i) each left root is on a $K_m^{(c)}$, (ii) each right root is on an $F_m^{(c)}$, and (iii) each free point is a right point on a $K_m^{(c)}$ and a left point on an $F_m^{(c)}$) = 0 (25)

(See Figure 5).

B.2. First Theorem to be Proven. *Theorem.* For $M \geq 2$, $\mathscr{A}_{Fc}(M)$ and $\mathscr{A}_{Kc}(M-1)$ imply $\mathscr{A}_{Kc}(M)$.

Outline of the Proof. We are given that F_n and $F_n^{(c)}$ exist and that $F_n^{(c)}$ has the cluster property for $1 \leq n \leq M$. We are also given that K_n exists as a left and right inverse of F_n, that $K_n^{(c)}$ exists and has the cluster property, and that both functions have left symmetry, right symmetry, and left−right symmetry, for $1 \leq n \leq M-1$. What has to be proven is that K_M exists and is a left and right inverse of F_M, that $K_M^{(c)}$ has the cluster property, and that both K_M and $K_M^{(c)}$ have left symmetry, right symmetry, and left−right symmetry.

The method of the proof will be to start with the left inverse relationship equation that K_M must satisfy, insert a trial solution of the form of the cluster expansion for K_M, and obtain an equation that $K_M^{(c)}$ must satisfy. Then, we will solve this equation and show that the solution has left symmetry and right symmetry and the cluster property. This will establish that K_M exists as a left inverse that is both left symmetric and right symmetric. Then, we go through the same procedure starting with the right inverse relationship. This will show that we can construct a version of K_M that is a right inverse and that is both left symmetric and right symmetric. It is elementary to prove that the left and right inverses are the same. Then, it follows from the left−right symmetry of F that K_M has left−right symmetry, and from this, it follows that $K_M^{(c)}$ has left−right symmetry.

Proof. The first step is to construct a formula for a K_M that is a left inverse of F_M. We construct a trial solution for K_M using the cluster formula, eq 21, in which we make use of the known $K_n^{(c)}$ for $n < M$ and regard $K_M^{(c)}$ as an unknown function to be obtained. Without loss of generality, we assume that the unknown $K_M^{(c)}$ is right symmetric. (In the left inverse relationship, $K_M^{(c)}$ is multiplied on the right by $F_M^{(c)}$, which is left symmetric. Thus, if we had a $K_M^{(c)}$ that implied a K_M that is a left inverse of F_M and if the $K_M^{(c)}$ were symmetrized with regard to its *right* arguments, the new K_M would also be a left inverse of F_M.) However, we make no assumptions that $K_M^{(c)}$ has other symmetries or has the cluster property.

Substitute the trial solution for K_M into the diagrammatic form, eq 22, for the (left) inverse relationship between K_M and F_M. (Because the $K_M^{(c)}$ is right symmetric, it can be represented as a vertex in which the right points are all topologically equivalent.) The result is an equation for $K_M^{(c)}$ that is of the same form as eq 23.

In the resulting equation, some of the diagrams are not connected. The diagrams that are not connected do not contain the unknown function $K_M^{(c)}$; instead, they contain only the known $K_M^{(c)}$ for $m < M$. If we group together the terms of a given connectivity, the sum is a product of factors with one factor for each of the connected sets of roots. Each of these factors is of the form of the left side of eq 24) or 25. As a result, all of the disconnected diagrams cancel on both sides of the equation, leaving the following equation for $K_M^{(c)}$.

(the sum of all topologically different connected matrix diagrams with (i) M left roots labeled $1...n$, (ii) M right roots labeled $1'...n'$, (iii) M free points, and (iv) $F^{(c)}$ and $K^{(c)}$ vertices such that (i) each left root is on a $K^{(c)}$, (ii) each right root is on an $F^{(c)}$ and (iii) each free point is a left point on a $K^{(c)}$ and a right point on an $F^{(c)}$) = 0 (26)

Let's categorize the terms that appear here. (i) Some diagrams have no $K_M^{(c)}$. The sum of these is a function that has the cluster property and that has left symmetry and right symmetry. Let's call this sum $L_M(1...M;1'...M')$. (ii) Some diagrams have one $K_M^{(c)}$ vertex but all of the $F^{(c)}$ vertices are $F_1^{(c)}$ vertices. (iii) Some have $K_M^{(c)}$ and at least one $F_m^{(c)}$ with $m > 1$.

Let us define $F_M^{(D)}(1...n;1'...n')$ to be the sum of terms in the cluster series for F_M that have only $F_1^{(c)}$ vertices. This is the completely "disconnected" part of F_M. We also define $K_M^{(D)}$ to be the sum of terms in the cluster series for $K_M^{(D)}$ that have only $K_1^{(c)}$ vertices. It is straightforward to show, using eq 24 and the left−right symmetry of $F_1^{(c)}$ and $K_1^{(c)}$, that $K_M^{(D)}$ is both a left inverse and a right inverse of $F_M^{(D)}$.

Then, the second set of terms above is equal to

$$\frac{1}{M!}\int \mathrm{d}1''...\mathrm{d}M''\, K_M^{(c)}(1...M;1''...M'')F_M^{(D)}(1''...M'';1'...M') \tag{27}$$

Similarly, we define $F_M^{(P)}$ to be the sum of the terms in the cluster series for F_M that have at least one $F_M^{(c)}$ for $m > 1$. This is the "at least partially connected" part of F_M. Equation 26 then becomes

$$L_M + K_M^{(c)}F_M^{(D)} + K_M^{(c)}F_M^{(P)} = 0$$

Here, for brevity, we use a matrix notation with the matrix product being defined as in eq 27.

Multiply on the right by $K_M^{(D)}$, the inverse of $F_M^{(D)}$. The result becomes

$$K_M^{(c)} = -L_M K_M^{(D)} - K_M^{(c)} F_M^{(P)} K_M^{(D)}$$

This is an integral equation for $K_M^{(c)}$. Note that all of the functions in this equation other than $K_M^{(c)}$ are defined in terms of finite series of diagrams containing functions that ultimately are defined in terms of the moments, m_n, and cumulants, $m_n^{(c)}$. If we could prove the existence of a solution of this equation and demonstrate the needed symmetry and cluster properties, we would thereby complete the proof of the theorem.

The equation can be solved formally by iteration. The result is an infinite series of the form

$$K_M^{(c)} = -L_M K_M^{(D)} + L_M K_M^{(D)} F_M^{(P)} K_M^{(D)} - L_M K_M^{(D)} F_M^{(P)} K_M^{(D)} F_M^{(P)} K_M^{(D)} + \ldots$$

This could be expressed in diagrammatic language, but this will not be necessary. The result has the cluster property. (This follows because L_M has the cluster property and appears in each term and every other function in this expression consists of factors that are connected, in a graphical sense, to the L_M.) The result is also left symmetric (because L_M appears on the left of each term and it is left symmetric) and right symmetric (because $K_M^{(D)}$ appears on the right of each term and it is right symmetric).

We have found a solution for $K_M^{(c)}$ that has the cluster property. Using this, we have a form for K_M that is a left inverse of F_M and that has left symmetry and right symmetry.

Now we can go through the same procedure to produce a right inverse. Most of the terms will be the same as in the left inverse because they are based on the previous $K^{(c)}$ functions. At this stage, we have not established that the last term, the $K_M^{(c)}$ term, is the same in both functions. Let's call these functions K_{Ml} and K_{Mr} for the left and right inverse. Elementary arguments (details omitted) then can be used to show that these two inverses are the same function and that they have left−right symmetry. It follows that $K_M^{(c)}$ has left−right symmetry. This establishes all of the results needed to complete the theorem.

B.3. Second Theorem to be Proven. *Theorem.* $\mathscr{A}_{Jc}(M-1)$ and $\mathscr{A}_{Kc}(M-1)$ imply $\mathscr{A}_{Jc}(M)$.

Outline of the Proof. We are given that the $J_{nm}^{(c)}$ functions have the cluster property for $0 \le n \le M-1$ and $n+m \ge 2$ and that the inverses K_n and their cluster functions $K_n^{(c)}$ exist with all their properties as described above for $0 \le n \le M-1$. We want to prove that $J_{Mm}^{(c)}$ has the cluster property.

Because the K_n functions exist for $n \le M-1$, eq 7 is a valid representation for ϕ_n for $n \le M$. This will be used to obtain an expression for J_{nm} in terms of $J_{pq}^{(c)}$ and $K_{pq}^{(c)}$ for $p < n$. This expression will be compared with the cluster expansion, eq 18, which expresses J_{nm} in terms of $J_{pq}^{(c)}$ for $p \le n$. On the basis of this comparison, we will obtain a graphical expression for $J_{nm}^{(c)}$ in terms of $J_{pq}^{(c)}$ and $K_{pq}^{(c)}$ for $p < n$ that is valid for $n \le M$. This result for J_{Mm} allows us to conclude that J_{Mn} has the cluster property.

Proof. Using eq 7 and the representation of the projection operators in eq 10, we have, for $0 < n \le M$,

$$|\phi_n(1\ldots n)\rangle = |\psi_n(1\ldots n)\rangle - \sum_{p=0}^{n-1} \frac{1}{(p!)^2} \int \mathrm{d}1'''\ldots\mathrm{d}p'''\, \mathrm{d}1''\ldots\mathrm{d}p''\, |\phi_p(1'''\ldots p''')\rangle K_p(1'''\ldots p''';1''\ldots p'')\langle\phi_p(1''\ldots p'')|\psi_n(1\ldots n)\rangle$$

If we operate on both sides of this equation with $\langle\psi_m(1'\ldots m')|$ and take the transpose of both sides, we get

$$J_{nm}(1\ldots n;1'\ldots m') = m_{n+m}(1\ldots n1'\ldots m') - \sum_{p=0}^{n-1} \frac{1}{(p!)^2} \int \mathrm{d}1'''\ldots\mathrm{d}p'''\, \mathrm{d}1''\ldots\mathrm{d}p''\, J_{np}^{\mathrm{T}}(1\ldots n;1''\ldots p'') K_p(1''\ldots p'';1'''\ldots p''')J_{pm}(1'''\ldots p''';1'\ldots m') \quad (28)$$

where

$$J_{nm}^{\mathrm{T}}(1\ldots n;1'\ldots m') \equiv J_{mn}(1'\ldots m';1\ldots n) \quad (29)$$

Equation 28 is valid for $n \le M$ and for all m. The $p = 0$ term here should be regarded as $-J_{n0}^{\mathrm{T}}(1\ldots n;)J_{0m}(;1'\ldots m')$.

Translating this formula into diagrams, we get

$J_{nm}(1\ldots n;1'\ldots m') = m_{n+m}(1\ldots n1'\ldots m')$ − (the sum of all topologically different matrix diagrams with (i) n left roots labeled $1\ldots n$, (ii) m right roots labeled $1'\ldots m'$, (iii) $2p$ free points, where $0 \le p \le n-1$, and (iv) a J_{np}^{T} vertex, a K_p vertex, and a J_{pm} vertex such that (i) the left roots are on the J_{np}^{T}, (ii) the right roots are on the J_{pm}, (iii) one set of p free points are the right points on the J_{np}^{T} and the left points on the K_p, and (iv) one set of p free points are the right points of the K_p and the left points of the J_{pm})

Introduce the cumulant expansion of the m_{n+m} moments and the cluster representation of the K, J, and J^{T} vertices.

$J_{nm}(1\ldots n;1'\ldots m')$ = (the sum of all topologically different diagrams with (i) $n+m$ roots labeled $1\ldots n1'\ldots m'$ and (ii) $m^{(c)}$ vertices) − (the sum of all topologically different matrix diagrams with (i) n left roots labeled $1\ldots n$, (ii) m right roots labeled $1'\ldots m'$, (iii) $2p$ free points, where $0 \le p \le n-1$, and (iv) $J^{(c)\mathrm{T}}$, $K^{(c)}$, and $J^{(c)}$ vertices such that (i) the left roots are on the $J^{(c)\mathrm{T}}$ vertices, (ii) the right roots are on the $J^{(c)}$ vertices, (iii) one set of p free points are the right points on the $J^{(c)\mathrm{T}}$ and the left points on the $K^{(c)}$, and (iv) one set of p free points are the right points of the $K^{(c)}$ and the left points of the $J^{(c)}$) (30)

Here $J^{(c)\mathrm{T}}$, the transpose of $J^{(c)}$, is defined as in eq 29.

In each of these two series, there are diagrams that are not connected. Free points may appear in diagrams in the second series, and each free point is connected to a root. Thus, each diagram consists of one or more connected subgraphs. Each subgraph contains one or more roots, zero or more free points, and at least one vertex. Each graph corresponds to a partition of the $n+m$ root labels into one or more subsets, according to how the roots are connected in the diagram. If $\mathscr{D}(1\ldots n1'\ldots m')$

denotes a partition of the labels, then this equation can be expressed formally as

$$J_{nm}(1...n1'...m') = \sum_{\mathscr{P}(1...n1'...m')} (\text{the sum of all terms on the right of eq 30 that correspond to } \mathscr{P}(1...n1'...m')) \quad (31)$$

Comparing this with the cluster expansion for the same quantity in eq 18, we see that both equations are sums over partitions. In eq 18, each partition gives one contribution equal to the product the $J^{(c)}$ factors for each of the subsets in the partition. Equation 18 can be expressed as

$$J_{nm}(1...n1'...m') = \sum_{\mathscr{P}(1...n1'...m')} \prod_i J^{(c)}(\mathscr{P}_i(1...n1'...m')) \quad (32)$$

Here, $\mathscr{P}_i$ denotes the ith subset of arguments in the partition $\mathscr{P}$ and the product is over all subsets.

In eq 31, because of the connectivity of the diagrams and the cluster properties of the vertices that appear in them, we can conclude that the term for a specific $\mathscr{P}$ is zero unless for each subset $\mathscr{P}_i$ the arguments in that subset are all close together. This holds for all $n \leq M$. In eq 32, the same property holds, but at the moment, we know this only for $n < M$. (Note that we have not established and cannot assume that $J^{(c)}_{Mm}$ has the cluster property. This function does not appear in eq 31 for $n \leq M$ or in eq 32 for $n < M$, but it does appear in eq 32 for $n = M$.) Moreover, for both equations, the value of a term for specific $\mathscr{P}$ is invariant under translation of all of the position arguments in one subset relative to those in the other subsets. From these statements, it is straightforward to show that for each $\mathscr{P}$ the summand in eq 31 for $n < M$ is equal to the summand in eq 32 for the same n. Hence, for $n < M$,

$$(\text{the sum of all terms on the right of eq 30 that correspond to } \mathscr{P}) = (\text{the product of } J^{(c)} \text{ factors for the subsets in } \mathscr{P}) \quad (33)$$

In particular, for the special case that $\mathscr{P}$ contains a single subset, the diagrams must be connected and we get, for $n < M$,

$$J^{(c)}_{nm}(1...n1'...m') = \text{the sum of all the contributions to } J_{nm}(1...n1'...m') \text{ in eq 30 that are connected} \quad (34)$$

By a different line of reasoning, we will show that eqs 33 and 34 hold also for $n = M$. But first, we must establish some lemmas.

Lemma. For $n < M$,

(the sum of all topologically different connected matrix diagrams with (i) n left roots labeled $1...n$, (ii) m right roots labeled $1'...m'$, (iii) $2p$ free points, where $0 \leq p \leq n-1$, and (iv) $J^{(c)T}$ vertices, $K^{(c)}$ vertices, and $J^{(c)}$ vertices such that (i) the left roots are on the $J^{(c)T}$ vertices, (ii) the right roots are on the $J^{(c)}$ vertices, (iii) one set of p free points are the right points on the $J^{(c)T}$ and the left points on the $K^{(c)}$, and (iv) one set of p free points are the right points of the $K^{(c)}$ and the left points of the $J^{(c)}$) $= m^{(c)}_{n+m}(1...n1'...m') - J^{(c)}(1...n;1'...m')$ (35)

(This follows directly from eqs 34 and 30.)

Consider the diagrams with the same structure as in eq 35 except that $p = n$. For these diagrams, we have the following lemma.

Lemma. For $n < M$,

(the sum of all topologically different connected matrix diagrams with (i) n left roots labeled $1...n$, (ii) m right roots labeled $1'...m'$, (iii) $2n$ free points, and (iv) $J^{(c)T}$ vertices, $K^{(c)}$ vertices, and $J^{(c)}$ vertices such that (i) the left roots are on the $J^{(c)T}$ vertices, (ii) the right roots are on the $J^{(c)}$ vertices, (iii) one set of n free points are the right points on the $J^{(c)T}$ and the left points on the $K^{(c)}$, and (iv) one set of n free points are the right points of the $K^{(c)}$ and the left points of the $J^{(c)}$) $= J^{(c)}(1...n;1'...m')$ (36)

Proof. Because there are as many points that are right points of the $J^{(c)T}$ vertices as there are left points, in each nonzero diagram every $J^{(c)T}$ must have the same number of left points and right points. (This follows from eq 19.) But $J^{(c)T}_{nn} = F^{(c)}_n$. Hence, the value of the left-hand side of eq 36 is equal to

(the sum of all topologically different connected matrix diagrams with (i) n left roots labeled $1...n$, (ii) m right roots labeled $1'...m'$, (iii) $2n$ free points, and (iv) $F^{(c)}$, $K^{(c)}$, and $J^{(c)}$ vertices such that (i) the left roots are on the $F^{(c)}$, (ii) the right roots are on the $J^{(c)}$, (iii) one set of n free points are the right points on the $F^{(c)}$ and the left points on the $K^{(c)}$ and (iv) one set of n free points are the right points of the $K^{(c)}$ and the left points of the $J^{(c)}$ (37)

Some diagrams in this series have a single $J^{(c)}_{nm}$ vertex attached to all of the right roots and some do not. Let's consider these two sets of diagrams separately (see Figure 6).

For the diagrams that have a single $J^{(c)}_{nm}$ attached to all of the right roots, any connectivity among the left roots and the $2n$ free points provided by any set of $F^{(c)}$ and $K^{(c)}$ vertices will lead to an acceptable connected diagram that is in the series. The sum of all of these diagrams is simply $J^{(c)}_{nm}(1...n;1'...m')$, because the sum over all of the possibilities for the $F^{(c)}$ and $K^{(c)}$ is equal to the **I** function, with its left arguments being $1...n$ and its right arguments being the n free points that are the left points of the $J^{(c)}_{nm}$.

For the diagrams that have two or more $J^{(c)}$ vertices attached to the right roots, the set of $F^{(c)}$ and $K^{(c)}$ vertices must provide additional connectivity for the diagram to be connected, and only diagrams with that additional connectivity are included in the sum. It can be shown that each way of providing the needed connectivity leads to a result that sums to zero. The basic reason for this is that the total way of providing any particular type of needed connectivity has the structure of the transpose of the left side of eq 22, which is zero. A simple case of this is illustrated in Figure 6, and a straightforward analysis shows that the same reasoning applies to all diagrams with two or more $J^{(c)}$ attached to the roots. Hence, the sum of the diagrams in eq 37 with two or more $J^{(c)}$ attached to the roots is zero.

Thus, the sum of all of the diagrams in eq 37 is equal to the sum of those that have a $J^{(c)}_{nm}$, and we showed above that this sum is simply $J^{(c)}_{nm}$, which was to be demonstrated. We can now

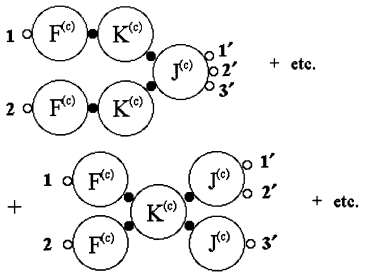

Figure 6. Illustration of the basic idea of the lemma that proves eq 36. These are some of the diagrams on the left-hand side of eq 37. The first diagram is an example of a diagram with a single $J^{(c)}$ attached to all of the right roots. Because the diagrams must be connected, we can see that the $F^{(c)}$ and $K^{(c)}$ must connect the left points of the $J^{(c)}$ to each of the left roots. The various sets of $F^{(c)}$ and $K^{(c)}$ that can accomplish this are precisely the transposes of the diagrams on the left-hand side of part b of Figure 4. These sum to give the simple delta functions on the right of part b. Using this sum to obtain the sum of all diagrams on the left of eq 37 that have a single $J^{(c)}$ attached to the left roots gives a result in which that single $J^{(c)}$ is now directly attached to the left roots. The second figure is an example of a diagram in which there are two $J^{(c)}$ on the right. Because the diagrams in this series must be connected and because the right roots are not connected by the $J^{(c)}$, the $F^{(c)}$ and $K^{(c)}$ vertices must not only connect the left points of the $J^{(c)}$ to left roots, as in the previous diagram, but must also connect the left roots together and connect the two left points on the two $J^{(c)}$ vertices together. The various sets of $F^{(c)}$ and $K^{(c)}$ that can accomplish this are precisely the transposes of the *connected* diagrams on the left of part b of Figure 4. But, as is seen in Figure 5, the sum of these connected diagrams is zero.

add the results for the previous two lemmas together to get another lemma.

Lemma. For $n < M$,

(the sum of all topologically different connected matrix diagrams with (i) n left roots labeled $1...n$, (ii) m right roots labeled $1'...m'$, (iii) $2p$ free points, where $0 \le p \le n$, and (iv) $J^{(c)T}$ and $K^{(c)}$ vertices and one $J^{(c)}$ vertex such that (i) the left roots are on the $J^{(c)T}$, (ii) the right roots are on the $J^{(c)}$, (iii) one set of p free points are the right points on the $J^{(c)T}$ and the left points on the $K^{(c)}$, and (iv) one set of p free points are the right points of the $K^{(c)}$ and the left points of the $J^{(c)}$) $= m^{(c)}_{n+m}(1...n1'...m')$ (38)

Lemma. For $n = M$, eq 33 holds for partitions $\mathscr{P}$ that contain two or more subsets.

Proof. In each graph in the second sum in eq 30, there are p free points connected to $J^{(c)T}$ vertices and an equal number of distinct free points connected to $J^{(c)}$ vertices. Any $J^{(c)T}$ vertex with more right points than left points is zero, as is any $J^{(c)}$ vertex with more left points than right points. It follows that in any nonzero diagram, half the number of free points (i.e., p) must be less than or equal to n and less than or equal to m. The diagrammatic series contains the explicit restriction that $p \le n - 1$. Thus, we replace the restriction that $p \le n - 1$ with the restriction that $p \ne n$.

Consider a partition $\mathscr{P}$ that contains two or more subsets and consider the sum of all terms on the right-hand side of eq 30 that correspond to $\mathscr{P}$. If the restriction that $p \ne n$ were not present in the second sum of eq 30, the sum of all terms that correspond to $\mathscr{P}$ would be zero. To see this, note that in the diagrams in the second sum, there would be (in the absence of the $p \ne n$ restriction) no relationship between the vertices and connections within one subgraph and within another. Therefore, the second sum would be a product of factors, one factor for each subset of arguments, and the factor for a subset is the sum of connected diagrams with the specifications given in the series. But this sum of connected diagrams is the same as the series in eq 38, and the result is just a factor of $m^{(c)}$ for the subset. The result for the second sum is precisely the one term in the first sum of eq 30 that corresponds to $\mathscr{P}$. The minus sign in front of the second term implies that the terms would cancel.

Because of this cancellation, it follows that the sum of all terms on the right-hand side of eq 30 that correspond to $\mathscr{P}$ can be obtained by simply evaluating the second sum with the added restriction that $p = n$ and with a plus sign. (In effect, we add and subtract the sum with the $p = n$ restriction to the right side of the equation, and the subtracted sum combines with the existing second sum to cancel the existing first sum.) Thus, we have

(the sum of all terms on the right of eq 30 that correspond to $\mathscr{P}$) = (the sum of all topologically different matrix diagrams with (i) n left roots labeled $1...n$, (ii) m right roots labeled $1'...m'$, (iii) $2n$ free points, and (iv) $J^{(c)T}$ and $K^{(c)}$ vertices and one $J^{(c)}$ vertex such that (i) the left roots are on the $J^{(c)T}$ vertices, (ii) the right roots are on the $J^{(c)}$ vertices, (iii) one set of n free points are the right points on the $J^{(c)T}$ and the left points on the $K^{(c)}$, (iv) one set of n free points are the right points of the $K^{(c)}$ and the left points of the $J^{(c)}$, and (v) the connectivity of the diagram corresponds to $\mathscr{P}$)

Because $\mathscr{P}$ contains two or more subsets, these diagrams are disconnected and each consists of one or more connected subgraphs. In each graph, the number of free points attached to the $J^{(c)T}$ vertices is equal to the number of left roots. This must be true for each disconnected subgraph as well if the diagram has a nonzero value. (If one subgraph has more or fewer free points attached to $J^{(c)T}$ vertices than it has left roots, then some other subgraph has more free points attached to $J^{(c)T}$ vertices than it has left roots. This implies that there would have to be at least one $J^{(c)T}$ attached to more free points than left roots, and such a $J^{(c)T}$ has the value of zero.) Then, the number of free points that are right points of the $K^{(c)}$ in each subgraph must also be equal to the number of left roots in the subgraph.

The sum on the right side of this latest result is a product of factors for each of the subsets in $\mathscr{P}$. Each of these subsets has fewer than n left roots and hence has fewer than M left roots. The factor for each subset is the same as the series in eq 35, with n and m in that series being replaced by the number of left and right roots in the subgraph. Hence, the factor for each subset is the $J^{(c)}$ for that subset, which was to be demonstrated.

Lemma. Equations 33 and 34 hold for $n = M$.

Proof. This result is an elementary consequence of the preceding lemma. The preceding lemma established that for $n = M$ eq 33 holds for all partitions containing two or more subsets. There is only one other partition, namely, the partition with one subset. Therefore, the corresponding summands in eqs 31 and 32 are equal for each of the $\mathscr{P}$ with two or more subsets. But the left sides of these equations are the same. Therefore, the summands for the partition with one subset must also be equal. This establishes eq 33 for $n = M$. Equation 34 for $n = M$ follows immediately, which was to be demonstrated.

Return to the Main Theorem. We can now complete the proof of the theorem. The previous lemma shows that J_{Mm} is given by eq 34. Every diagram on the right in that equation is connected, and every vertex that appears has the cluster property. Therefore, J_{Mm} has the cluster property, which was to be demonstrated.

B.4. Proof of a Lemma. *Lemma.* $\mathcal{A}_\phi(1)$, $\mathcal{A}_{Jc}(1)$, $\mathcal{A}_{Fc}(1)$, $\mathcal{A}_K(1)$, and $\mathcal{A}_{Kc}(1)$ are true.

Proof. P_0 is given by eq 8. Thus, $\mathcal{A}_\phi(1)$ is true. By direct evaluation, it is easily shown that

$$J^{(c)}_{0m}(;1'...m') = m^{(c)}_m(1'...m')$$

$$J^{(c)}_{1m}(1;1'...m') = m^{(c)}_{m+1}(1;1'...m')$$

$$F_1(1;1') = F^{(c)}_1(1;1') = m^{(c)}_2(11') = nM(\mathbf{P}_1)\delta(1;1') + nM(\mathbf{P}_1)nM(\mathbf{P}_2)h(\mathbf{R}_1,\mathbf{R}_1')$$

Here $n = \langle N\rangle/V$ is the particle density, $h(\mathbf{R}_1,\mathbf{R}_2) = g(\mathbf{R}_1,\mathbf{R}_2) - 1$, g is the usual radial distribution function of the fluid, and $M(\mathbf{P})$ is a normalized Maxwell−Boltzmann distribution function of momentum. An explicit formula for $K_1(1;1')$ in terms of the direct correlation function $c(\mathbf{R}_1,\mathbf{R}_2)$ is

$$K_1(1;1') = K^{(c)}_1(1;1') = \frac{1}{nM_m(\mathbf{P}_1)}\delta(1;1') - c(\mathbf{R}_1,\mathbf{R}_{1'}) \quad (39)$$

It follows straightforwardly from the Ornstein−Zernike relationship between c and h that this K_1 is both a left and a right inverse of F_1. The $m^{(c)}$ functions and the direct correlation function have the cluster property and are symmetric under permutation of their arguments. The lemma follows directly from these results.

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(25) If the procedure terminated, which is unlikely, such that, for example, ϕ_m for some m does not exist, the structure of the theory would not change; the only effect would be that the size of the part of Hilbert space that is relevant for the calculation of correlation functions would be somewhat smaller.
(26) Except for the factor of $1/(n!)^2$, which could be absorbed into the definition of K_n, this is a standard formula for a projection operator onto a subspace. We note that the present definition of the inverse function K_n is different from that of the analogous function that appears in the theory of Mazenko,[8] because of a different choice of the form of the "multiplication" on the left-hand side of eq 11 and a different choice of the "identity" **I** [see eq 12] that appears that appears on the right-hand side. The differences are small, that is, various factors of $n!$, but the definition presented here is more suitable for the graphical formulation that is the subject of this paper.
(27) For example, see the discussion after eq 20 and in footnote 27 of Mazenko.[8]
(28) See the discussion after eqs 11 and 12 of Boley.[10]
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