Basis of the Functional Assumption in the Theory of the Boltzmann Equation

M. S. GREEN AND R. A. PICCIRELLI
National Bureau of Standards, Washington, D. C.
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The long-time behavior of the n-particle probability densities for a large, dilute system of point particles interacting with short-range repulsive forces is studied. The main result is an exact series for the n-particle density which consists of two parts. The first part is a time-independent functional of the singlet density which is expressed as a functional power series and which is a direct analog of the equilibrium density series. The second part is also a functional power series in the singlet density but the coefficients depend on time and on the initial correlations. The coefficients of both series are given explicitly in terms of operators which are determined by the dynamics of isolated groups of particles. It is demonstrated that these operators vanish for phase points corresponding to motions during which there are two or more groups of particles which either are statistically and dynamically independent or are such that each of them is dynamically connected to the rest by no more than one particle. It is argued that all the terms of the exact series are finite and that the terms of second part (the error) decrease with increasing time so that the first part is the asymptotic form proposed by Bogoliubov. The relevance of the results for the Boltzmann equation is indicated. A form of the Boltzmann collision integral which is valid in the steady state and to all orders of the density is described.

I. INTRODUCTION

ONE of the most important contributions to the understanding of the statistical mechanical basis of the Boltzmann equation has been the conjecture or assumption, made by Bogoliubov, that, in a dilute gas, the n-particle probability densities are functionals of the singlet probability density.[1] The application of this idea to the derivation of the higher density corrections to the Boltzmann equation has been investigated by a number of authors.[2–4]

It is obvious, however, that the functional assumption cannot be valid under all circumstances since, subject to rather weak conditions, the probability densities are quite arbitrary and one can conceive of possible n-particle probability densities which are not compatible with any particular functional relationship to the singlet probability density. Thus, it is of some importance, for the foundations of the Boltzmann equation and even for the general statistical mechanical theory of nonequilibrium phenomena, to establish from fundamentals *whether* the n-particle probability densities in an imperfect gas become, in some sense, functionals of the one-particle probability density. The features of the system and the initial conditions which allow such a result are of some interest in themselves. Furthermore, in view of the general import of this result for the foundations of statistical mechanics, insights into the *manner* in which the probability densities evolve toward the functional form are especially valuable.

In addition to these questions of principle are some more practical questions; namely, what is the explicit form of the functionals and how are they related to the known equilibrium expansions of the n-particle densities? What is the form of the Boltzmann equation to all orders in the number density?

In a previous publication, one of us, (MSG), has given a partial answer to some of these questions.[5] The first two terms of a series expansion of the functional representing the pair density were derived under the following assumptions: (a) The interparticle forces are purely repulsive and short ranged, and (b) the probability densities satisfy the product condition at the initial instant with a correlation length of the order of the range of forces. The time for approach to the functional form was estimated to be of the order of the ratio of the correlation length to a representative particle velocity. In addition, it was shown that, if the equilibrium singlet density is substituted in the functional expression, the result coincides up to third order in density with the known density expansion of the equilibrium pair density.

The present work generalizes and completes paper II by addressing itself to the proof of a general theorem; namely, supposing certain restrictions on the nature of the system and on the initial values, *if the time is large enough, the time-dependent n-particle probability densities are certain time-independent functionals of the one-particle probability density.* More symbolically, let any integer, say, α, denote the position and momentum (the "phase") of the αth particle and let $f_n(1,\cdots,n;t)$ denote the n-particle probability density considered as a function of these one-particle phase points. The theorem

[1] N. N. Bogoliubov, J. Phys. (U.S.S.R.) **10**, 265 (1946). See also, "Problems of a Dynamical Theory of Statistical Physics," translated by E. Gora, Providence College, 1959 (unpublished).

[2] S. T. Choh and G. E. Uhlenbeck, "The Kinetic Theory of Phenomena in Dense Gases," University of Michigan, 1958 (unpublished).

[3] E. G. D. Cohen, lectures in *Fundamental Problems in Statistical Mechanics*, edited by E. G. D. Cohen (North-Holland Publishing Company, Amsterdam, 1962).

[4] M. S. Green, J. Chem. Phys. **25**, 836 (1956), hereafter referred to as Paper I.

[5] M. S. Green, Physica **24**, 393 (1958), hereafter referred to as Paper II.

says that there is a certain functional, $f_n(1,\cdots,n|f_1(t))$, which depends on the time only through $f_1(t)$, and which differs negligibly from the n-particle probability density if the time is large enough; that is, the error defined by

$$\mathcal{E}_n(1,\cdots,n;t) \equiv f_n(1,\cdots,n;t)-f_n(1,\cdots,n|f_1(t)) \quad (1.1)$$

vanishes asymptotically in the time, t.[6]

This work also makes contact with paper I particularly by discussing some of the consequences of the theorem for the existence of a "generalized" Boltzmann equation.

We propose to establish this asymptotic theorem under the following restrictions on the system and on the initial state: *The probability densities describe a very large, dilute, system consisting of particles interacting with short-range, repulsive forces.* Thus, the probability densities satisfy the *infinite* Bogoliubov, Born, Green, Kirkwood, and Yvon hierarchy and, moreover, their expansions in powers of the density are meaningful. The *initial values of the probability densities satisfy the product condition* which was proposed and discussed in paper I. Essentially, $f_{n+m}([n]+[m];0)$ satisfies this condition if it becomes the product $f_n([n];0)f_m([m];0)$ when the phase points, $[n]$ and $[m]$, are sufficiently separated.[7]

An *exact* series expression for the n-particle probability density will be established. It will be shown that, under the restrictions just stated, this result lends itself rather naturally to a demonstration of the theorem and an understanding of the questions posed above. The expression is composed of two parts each given as a functional Taylor series in the one-particle density, $f_1(t)$; one part is just the asymptotic functional which has been denoted by $f_n([n]|f_1(t))$ and the other is the error $\mathcal{E}_n$ comprising the system's memory of its initial condition.

The asymptotic functional has a special form which is a generalization of the result for $f_2(12|f_1)$ given in paper II. It is a functional power series in $f_1(t)$ with time-independent coefficients; that is, it may be expressed as[8]

$$f([n]|f_1) = \sum_{l\geq 0}\frac{1}{l!}\int d([l])\tau_l^{(n)}([n];[l])\prod_{\alpha\in[n]+[l]} f_1(\alpha;t), \quad (1.2)$$

where the "coefficient-operator" $\tau_l^{(n)}$ is a sum of products of time-independent substitution operators each of which uniquely maps any given phase point into another one.[9] The error $\mathcal{E}_n$ is similarly expressed[10] but the coefficients, which are generalizations of the $\tau_l^{(n)}$, depend explicitly on time and on the initial correlations. The coefficients of both the asymptotic functional and the error are analogous to the irreducible clusters of equilibrium theory in that they are sums of not-more-than-singly-connected products of operators characteristic of the dynamics of isolated groups of particles. Roughly speaking, the terms of the asymptotic functional correspond to motions for which, at the initial time, the particles are moving freely and are uncorrelated while the error term corresponds to motions of initially correlated or interacting particles.

An important feature of the exact series expression is that both kinds of coefficients vanish for phase points such that one or more subgroups of particles are "singly connected." Singly connected groups are so called because of their analogy to singly connected clusters in the equilibrium theory of dense gases; essentially, they are groups whose motion is statistically and dynamically independent of the motion of all other particles except (possibly) for one "connecting particle." This property of the coefficients has several consequences for the exact series expansion: It implies that the only contributions to the error arise from points for which there are interactions and correlations present at the initial instant and for which there are no singly connected groups. In addition, this property guarantees that all the integrals have finite values at any finite time and that the terms in the error decrease with increasing time. The leading term in the two-particle probability density, for example, is proportional to t^{-1}.

The procedure to be used in establishing the exact series expression and its consequences can be outlined in the following way. Rather than assuming the detailed form of the coefficients and then proving its validity, we will give arguments which lead to their structure in a natural way. Thus, while we assume that the asymptotic functional has the structure indicated by Eq. (1.2), the coefficient operators $\tau_l^{(n)}$ are specified only as being time-independent substitutuion operators (i.e., we assume only that we are dealing with a time-independent functional). To develop an expression for the error, we use the formal series solution of the infinite hierarchy for the f_n which expresses them in terms of the solutions of isolated n-particle problems. This result, which is derived in paper I,[11] is briefly recapitulated in Sec. II. Then, by assuming the general structure of the asymptotic functional, this exact solution is used to develop a form for the error which involves only the $\tau_l^{(n)}$ and the solutions of n-particle Liouville equations. Thus, to discuss the long-time behavior of the error,

[6] Note that the theorem is easily restated for the time-dependent analogs of the Ursell-Mayer functions of equilibrium theory by using the relation (algebraic) between these and the n-particle densities.

[7] Here and in the following, the symbol $[n]$ denotes a set of n integers, say, $\{\alpha_1,\cdots,\alpha_n\}$.

[8] The product symbol is the usual one while the symbol "$\int d([l])$" has the same meaning as "$\int d\beta_1 d\beta_2\cdots d\beta_l$," where $\beta_i\in[l]$.

[9] This expression for the asymptotic functional (see Sec. IV for definition) of $\tau_l^{(n)}$ has also been derived for $n=2$ by E. G. D. Cohen, J. Math. Phys. 4, 183 (1963).

[10] See Eq. (5.1).

[11] Actually what is given there is the analogous series for Ursell functions. Our result is simply obtained from this by using the well-known relation between Ursell functions and densities.

the asymptotic behavior of the solution of Liouville's equation for, say, n particles is needed and this is discussed in Sec. III.

In Sec. IV, we show that this asymptotic behavior together with the supposition that $f([n]|f_1)$ is *the* asymptotic form leads to a full determination of the $\tau_l^{(n)}$. A similar argument, given in Sec. V, leads to a useful and more transparent expression for the error in terms of f_1 and coefficient operators which are natural generalizations of the $\tau_l^{(n)}$ and which include all the effects of initial conditions. In this manner, we arrive at the exact series representation of the n-particle probability density in a form which can be used to analyze long-time behavior. Sections IV and V also contain statements of the properties of the coefficient operators along with some discussion of their implications.

Finally, in the last section, various properties and consequences of this exact functional series for f_n are given. In particular, we argue that the error terms decrease in time and also discuss the consequences of the theorem for the generalized Boltzmann equation.

II. THE ERROR IN TERMS OF LIOUVILLE FUNCTIONS

In order to determine the form of the coefficient operators $\tau_l^{(n)}$ and to analyze the error $\mathcal{E}_n$ a suitable exact expression for the n-particle densities is needed. We choose for this a formally exact series solution of the infinite hierarchy which is given in terms of a sequence of functions ξ_m, each of which is a solution of Liouville's equation for some number m of isolated particles. For convenience, we will refer to such functions as "Liouville functions" in the sequel.[12] In this section, we will review this result and some of its properties and then use it to derive an expression for the error in terms of Liouville functions.

A. A Series Solution of the Hierarchy

A series solution of the initial value problem for the infinite hierarchy obeyed by the f_n has been given in Paper I.[11] The result is that for densities which satisfy the product condition initially, one has that

$$f([n];t)=\sum_{l\geq 0}\frac{1}{l!}\int d([l])\psi^{(n)}([n];[l];t). \quad (2.1)$$

The functions $\psi_l^{(n)}$ are defined in terms of Liouville functions ξ_m in the same manner as the "modified Ursell functions" of equilibrium theory are defined in terms of the Boltzmann factors; that is, for example, by the recursion relation:

$$\xi([n]+[l])=\sum_{[h]+[k]=[l]}\psi^{(n)}([n];[h])\xi([k]), \quad (2.2)$$

where the summation is over all partitions of $[l]$ into two disjoint parts, $[h]$ and $[k]$, either of which may be empty.[13] The explicit solution of Eq. (2.2) for $\psi_l^{(n)}$ is given in Appendix A, Eq. (A3). The functions $\psi_l^{(1)}$ defined by Eq. (2.2) are the same as the ψ_{1+l} of Paper I (except for factors of the number density).

Since the Liouville function ξ_m is by definition the solution of Liouville's equation for m isolated particles, the formal solution of the initial value problem may be written as

$$\xi([m];t)=\xi([m]_{-t};0)\equiv S_{-t}([m])\xi([m];0), \quad (2.3)$$

where the phase point $[m]_{-t}$ is the particular point which evolves into $[m]$ during an interval t under the natural motion of the m particles. The operator $S_{-t}([m])$, defined by this equation, has been introduced for notational convenience. It is the time-dependent substitution operator which projects the point $[m]$ into its "image" t seconds earlier; that is, as the time t increases, it generates the *prior* trajectory of the point $[m]$.[14]

Using Eq. (2.1), $\psi_l^{(n)}$ and hence f_n at time t are determined by the initial values of the sequence of Liouville functions. These initial values should be thought of as determined for a given sequence, $f_1(t=0)$, $f_2(t=0)$, $\cdots$, through Eqs. (2.1) and (2.2) evaluated at the initial time. As has been shown elsewhere, since the $f_n(t=0)$ satisfy the product condition, so do the $\xi_m(t=0)$ and, as a consequence, the $\psi_l^{(n)}$ have a cluster property.

It is significant that the lth term in the series for f_n involves the initial data and the dynamics of not more than $(n+l)$ isolated particles. However, probably the most important feature of this formal solution has already been discussed in papers I and II; namely, that each term in the series is asymptotic to a power of the time so that the later terms in the series grow faster with time than the earlier terms.[15] Hence, any number of terms of the series is a useful approximation to f_n only for *short* enough times (essentially for times small compared to the mean time *between* collisions). With this fact in mind, it will be helpful to consider as our goal the discovery of a transformation of Eq. (2.1) to a form which is valid for *large* times; i.e., we want,

[12] The ξ_m have a physical meaning in themselves. They are the probability densities at $[m]$ under the condition that a large enough region surrounding $[m]$ is empty of particles. For further discussion refer to the lectures by M. S. Green, in *Lectures in Theoretical Physics*, edited by W. E. Britten, B. W. Downs, and Joanne Downs (Interscience Publishers, Inc., New York, 1961).

[13] Thus, in this summation $[h]$ runs over every distinct (permutations of a given set are not distinct) subset, proper or not, of $[l]$; in particular, if $[h]=[l']$ and $[k]=[l'']$ is one term, $[h]=[l'']$ and $[k]=[l']$ is another. Also note that here and elsewhere time dependence is suppressed unless needed for emphasis or clarity. Similarly, variables (sets) of summation are suppressed whenever there is no ambiguity.

[14] Thus, $S_{-t}([m])$ is equivalent to $\exp(-itL_m)$ where L_m is the m-particle Liouville operator.

[15] To see this requires knowledge of the behavior of Liouville functions for long times and we will return to this point briefly in Sec. IV.

somehow, to use short-time information to deduce long-time information.

B. An Expression for the Error

The solution given by Eq. (2.1) can be used to express the error $\mathcal{E}_n$ in terms of the $\psi_l^{(n)}$ and eventually in terms of the Liouville functions.

This is simply achieved by noticing that, according to Eq. (1.1), if we assume the general form for the asymptotic functional given by Eq. (1.2), the error $\mathcal{E}_n$ is thereby expressed in terms of f_n and f_1. Then, if we evaluate this result by substituting for f_n and f_1 their expressions in terms of the $\psi_l^{(n)}$ and $\psi_k^{(1)}$, respectively, and collecting terms involving the same numbers of integration variables, we obtain (see Appendix A) an expression for $\mathcal{E}_n$ in terms of the $\psi_l^{(n)}$ and $\psi_k^{(1)}$. Since, however, it is the time dependence of the Liouville functions rather than that of the $\psi_l^{(n)}$ which is most easily understood, it is useful to go one step further and make the dependence of the error on the Liouville functions fully explicit. To accomplish this we have only to express $\psi_l^{(n)}$ and $\psi_k^{(1)}$ (in the result to which we have just referred) in terms of the ξ_m using the definition, Eq. (2.2).

The full transformation of Eq. (1.1) to the form explicit in the Liouville functions is carried out in Appendix A we find that

$$\mathcal{E}([n];t)=\sum_{l\geq 0}\frac{1}{l!}\int d([l]) \times \sum_{[h]+[k]=[l]} \mathfrak{D}^{(n)}([n];[h])\vartheta_0([k]), \quad (2.4)$$

where the summation has the same meaning as in Eq. (2.2). The factor $\mathfrak{D}_h^{(n)}$ is defined by

$$\mathfrak{D}^{(n)}([n];[h])\equiv\xi([n]+[h]) - \sum_{[q]+[r]=[h]} \tau^{(n)}([n];[q])\mathfrak{A}([n]+[q];[r]), \quad (2.5)$$

where the object $\mathfrak{A}([u];[v])$ is a sum of products of the ξ_m. It is defined by

$$\mathfrak{A}([u];[v]) \equiv \sum_{[v_0]+\Sigma_\alpha[v_\alpha]=[v]} \prod_{\alpha\in[u]} \xi(\alpha,[v_\alpha])\vartheta_{u-2}([v_0]), \quad (2.6)$$

where the sum is over all partitions of $[v]$ into $(u+1)$ parts, $[v_0]$ and the $[v_\alpha]$, some of which may be empty.

For convenience, we have introduced in Eq. (2.6) the sum $\vartheta_\sigma([v])$ (for the case $\sigma=u-2$). This is defined to be unity for $v=0$ and for $v\geqslant 1$ it is defined by

$$\vartheta_\sigma([v])\equiv\sum_{p=1}^{v}(-1)^p\frac{(\sigma+p)!}{\sigma!}\sum_{\mathcal{P}:[v]\text{ into }p}\prod_{\beta=1}^{p}\xi([v_\beta]), \quad (2.7)$$

where the summation is over all *distinct* partitions of $[v]$ into p, *nonempty* disjoint parts, $[v_\beta]$. Thus, $\mathfrak{A}([u];[v])$ is a sum of products of ξ_m whose arguments are disjoint subsets of $[u]+[v]$ with u of them containing a single member of $[u]$ (i.e., these are "hooked on" to $[u]$). Finally, the factor ϑ_0 appearing in Eq. (2.4) is determined by Eq. (2.7) for the case $\sigma=0$.

Equation (2.4) together with the Eqs. (2.5), (2.6), and (2.7) which define $\mathfrak{D}_l^{(n)}$ and ϑ_σ in terms of the ξ_m is the desired expression for $\mathcal{E}_n$ in terms of Liouville functions.

III. PROPERTIES OF LIOUVILLE FUNCTIONS

Having achieved an expression for the error in terms of Liouville functions, the significance of their asymptotic behavior for the properties of the error is manifest. It is already clear from the examples in Papers I and II that if an m-particle Liouville function satisfies the product condition initially, it eventually becomes determined by Liouville functions of lower order; that is, $\xi([m];t)$, say, asymptotically approaches a particular product of Liouville functions of lower order evaluated at time t and at points which are definite functions of $[m]$.

Our purpose here is to state and discuss these asymptotic forms, reiterating and generalizing those already given in Papers I and II and also quoting some additional ones which will be needed. A new notation is used which sharpens the results and seems easier to manipulate. Classical analogs of scattering operators arise naturally in the discussion and some of their properties, which are analogous to those of the ξ_m, will also be given.

The detailed nature of the asymptotic form for a given point depends on certain structural (or topological) characteristics of its prior trajectory. In order to have a convenient visualization of such characteristics we will use diagrams which represent for a given point, say, $[m]$, the *projection* of the prior trajectory $[m]_{-t}$ onto ordinary configuration space. Of course, the diagrams do not represent every detail of the trajectory of a point but only what will be called its "collision history." The collision history of a point essentially[16] consists of the following information about it: (a) on its *entire* prior trajectory, *which collisions* have occurred and their order and, (b) on the portion of its trajectory occurring *at and before* the initial instant, *which correlations* have occurred (particles are understood to be correlated if they are within some correlation length of each other). Clearly, then, each diagram represents many points all having the same collision history.

As an example of such diagrams consider Fig. 1. The solid lines represent single-particle trajectories and circles represent collisions between the particles whose trajectories are enclosed. Circles at the initial instant

[16] The additional bit of information which is needed is described in Sec. IIIB.

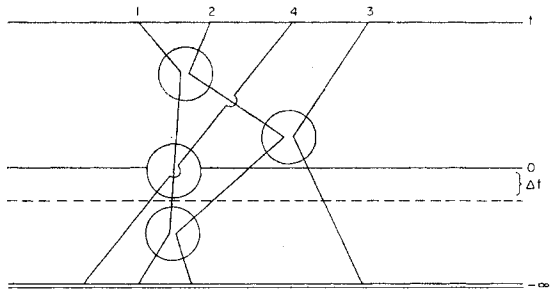

FIG. 1. Typical diagram of a collision history illustrating correlations and a collision at the initial time.

represent *either* collisions or correlations between the enclosed particles and it will not be necessary to explicitly indicate correlations prior to the initial instant. The order of collisions is obvious, while the double line at the bottom represents an infinite earlier time; i.e., the diagram says that no other collisions than the one between particles 1 and 2 occur for times more than t seconds earlier. It will be important to realize that, if one considers the history of the *same* point, say, Δt seconds later, the only effect on the diagram is to change the indicated position of the origin of time to a position Δt seconds earlier.[17]

In addition to diagrams representing collision histories we will also use diagrams which we will call "schematics" of collision histories. These simply represent whole classes of collision histories all having the same general character. For example, Fig. 6 is schematized by Fig. 2 (see also Fig. 5).

A. Statistical and Dynamical Independence

Because of the assumed finiteness of the correlation length and the range of force, one is rather naturally led to introduce concepts of statistical and dynamical independence. Since these concepts will be used extensively in the sequel, a brief elaboration will be given.

Several (disjoint) groups of particles are *dynamically independent* during an interval of time if there are no collisions between any particles belonging to different groups during the interval. Therefore, when we say that, for the point $[m]$ the groups $[m_\lambda]$ are dynamically independent during some time interval, the immediate consequence (or even the meaning) is that, for times within the interval,

$$S_{-t}([m])=\prod_\lambda S_{-t}([m_\lambda]), \tag{3.1}$$

where the phase points $[m_\lambda]$, appearing as arguments, are just the projections of $[m]$ onto the respective m_λ-particle phase spaces.

Similarly, several groups of particles are *statistically independent* at a given time if each particle of any one group is separated from every particle of any other group by a distance greater than the correlation length. Thus, when we say that the point $[m]$ is such that at the initial time, the groups $[m_\lambda]$, which partition $[m]$, are statistically independent, we mean that the phase points $[m_\lambda]_{-t}$, which are the image points of the groups $[m_\lambda]$, t seconds earlier,[18] are sufficiently separated so that the product condition is satisfied; that is, we can say for such points that

$$\xi([m]_{-t};0)=\prod_\lambda \xi([m_\lambda]_{-t};0)$$
$$\equiv S_{-t}([m])\prod_\lambda \xi([m_\lambda];0). \tag{3.2}$$

Furthermore, saying that statistical independence holds anywhere in the time interval $[0,-\infty]$ for a given point $[m]$ means that once Eq. (3.2) holds for some given time it holds for all longer times.

It seems evident that for an arbitrary point and at any time there is, in general, *some* partition of the particles into disjoint groups which is such that these groups are statistically and dynamically independent for *all* of the prior trajectory occurring at and before the initial instant (that is, for what might be called the "early" history of the point). Thus, for example, those points $[m]$, for which, say, the groups $[m_\lambda]$ are statistically and dynamically independent in the time interval $[0,-\infty]$ have histories in this interval which can be schematized as indicated (for this interval) in Fig. 2. In this diagram the heavy lines in the early history denote the groups $[m_\lambda]$, and within these groups the history may have any degree of complexity. Clearly, the only points whose early history cannot be so schematized for some choice of sets $[m_\lambda]$ are those for which all m particles are "linked up" prior to the initial instant.

It will become apparent that the statistical and dynamical independence of early histories guarantees the asymptotic nature of the functionals to be derived. Also, it determines how the particles are assigned to

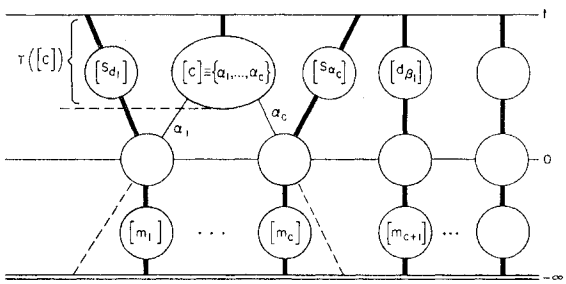

FIG. 2. Schematic of the history of not-more-than-singly-connected points illustrating disconnected sets $[d_\beta]$ and singly connected sets $[s_\alpha]$. Supposing other particles to be absent, the group $[c]$ has a history which is complete.

[17] This is why one must know correlations during the entire interval prior to the initial instant.

[18] The phase $[m_\lambda]_{-t}$ is the projection of $[m]_{-t}$ onto the m_λ-particle phase space.

the argument sets of the Liouville functions which occur as factors in the asymptotic form.

B. Complete Points

It seems appropriate to investigate first the behavior of Liouville functions for points which have the simplest of early histories; namely, all points $[c]$ which are such that, for time t, *every* particle is statistically and dynamically independent before (and at) the initial instant. Furthermore, we shall not make any further qualifications about the point $[c]$, so that the particles may have any sort of collision structure after the initial instant. Thus, the points $[c]$ we have in mind have histories which can be schematized for time t by the case of Fig. 2, which occurs when there are no other particles present except those in the set $[c]$ (i.e., by the projection of the history of all the particles onto the space of the particles, $[c]$) so that the early history of the particles $[c]$ is as indicated by the dashed lines.

Such points will be called "complete" for time t. We hasten to add that this notion does *not* mean that no collisions are in progress at time t, but only refers to the fact that for such points collision events occur only *within* the interval and not at or prior to the initial instant.[19] It is clear (from the meaning of statistical and dynamical independence) that if a point is complete for a time t it remains so for all larger times. Moreover, because we are assuming finite range, repulsive forces, it seems evident that (except possibly for a set of zero measure) all points *eventually* become complete so that *the volume of points which are complete increases indefinitely with the time.*

If $[c]$ is a point which is complete for time t, $\xi([c];t)$ reduces to a product of one-particle Liouville functions evaluated at certain one-particle phase points and at time t. In fact, one can establish by a simple argument that, if $[c]$ is complete for t, then

$$\xi([c];t)=\mathcal{S}([c])\prod_{\alpha\in[c]}\xi_1(\alpha;t)\,, \tag{3.3}$$

where $\mathcal{S}([c])$ is a time-independent substitution operator defined by the statement:

$$\mathcal{S}([c])=\lim_{t\to\infty}S_{-t}([c])\prod_{\alpha\in[c]}S_t(\alpha)\,. \tag{3.4}$$

To determine the effect of the operator $\mathcal{S}([c])$ on the phase of some member, say β, of the set $[c]$, consider the operator the limiting value of which is $\mathcal{S}([c])$. The instruction for this operator is: For the given point $[c]$, compute according to the natural motion of all c particles, the image β_{-t} of particle β at a time t seconds *earlier*, and the compute the phase that β would have had at time t if it had moved *freely* from the point β_{-t}. Now, although neither $S_{-t}([c])$ nor $\prod_{\alpha\in[c]}S_t(\alpha)$ operating separately yields a limiting value for large times (because the positions which result keep changing), their product does have a limit because eventually any point $[c]$ is complete. For, once we have gotten the image point of some particle at a time earlier than the time at which it begins its first collision, for all larger times the projection forward according to free particle motion always gives the same point.[20]

This understanding of the operator $\mathcal{S}([c])$ not only makes Eq. (3.3) a definite algorithm (considering the c-body scattering problem as solved), but it also provides the basis for a simple proof of this equation which is sketched in Appendix B.

Some features of this first property of the Liouville functions are noteworthy. Because of the remark made above that almost all points eventually become complete, one can say that the result Eq. (3.3) holds for almost all phases $[c]$, if the time is long enough; that is, if it is greater than some time, say, the time $T([c])$ indicated in Fig. 2.[21] In this sense, the result holds asymptotically in time. However, since *no* time is larger than $T([c])$ for all possible phases $[c]$, the approach of $\xi([c];t)$ to the form given by Eq. (3.3) is *nonuniform*; for any time t there are always points which are not complete for which this form is not a good approximation. Still, it is true that Eq. (3.3) becomes valid in a region which grows with time and one can say that, because the Liouville functions obey the product condition initially, they have a "molecular chaos" property asymptotically in time.

C. Not-More-Than-Singly-Connected Points

Included in all the points which are not complete for time t, there are some for which the asymptotic form is still simple in the sense that, like that in Eq. (3.3), it is still a time-independent functional of Liouville functions of lower order evaluated at time t. These points will be called "not-more-than-singly-connected" and can be schematized as in Fig. 2, where the groups $[d_\beta]$ will be referred to as "disconnected," while the groups $[s_\alpha]$ will be described as "singly connected." The meaning of these concepts, which is suggested by the "connectivity" of the trajectories as indicated in Fig. 2, is already essentially correct but a brief elaboration will be given. Then the asymptotic form of a Liouville function evaluated at such a point and some related results will be presented.

A suitable characterization of the kind of point to be discussed is as follows: Consider, for a point $[m]$ and at time t, the partitions of the set $[m]$ into groups $[m_\lambda]$, which are statistically and dynamically independent before the initial instant. The point $[m]$ will be not-more-than-singly-connected if there is *some* such parti-

[19] To emphasize the uncorrelated character of the early history of such points, one can think of them as being "chaotic" in an appropriate special sense.

[20] This is illustrated in Fig. 9 where the double-primed points are time-dependent while the single-primed ones are not.

[21] Because of the way increasing time is indicated by the diagrams, this time corresponds to an interval as is indicated.

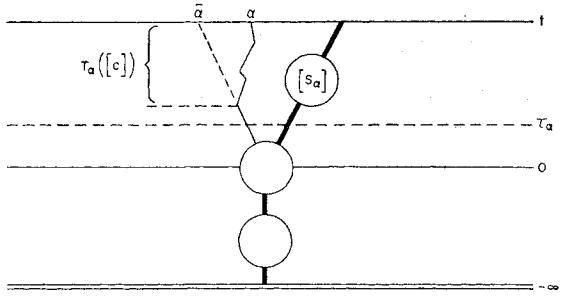

FIG. 3. Subdiagram illustrating relation of a singly connected group to the other particles.

tion for which the history following the initial instant satisfies certain conditions.

The *first* such condition is anticipated by the terminology; namely, *at most one* particle from each group $[m_\lambda]$ collides with members of other groups in the interval $[0,t]$. This condition implies that, in general, some of the $[m_\lambda]$ may be statistically and dynamically independent subsets of $[m]$ (under the natural motion of all m particles) during the *entire* infinite interval $[t,-\infty]$. Each such set will be referred to as being "disconnected." Each of the remaining subsets will contain a *single* "connecting" particle, all of these taken together composing a set, say, $[c]\equiv\{\alpha_1,\cdots,\alpha_c\}$. For convenience, these remaining subsets of $[m]$ will be denoted by $\{\alpha,[s_\alpha]\}$ where $\alpha\in[c]$ and the set $[s_\alpha]$ is the rest of the group which contains the particle α.

With the imposition of this first condition the sets $[s_\alpha]$ are not yet "singly connected." One can only say that the first collision of any particular connecting particle α must occur at some finite time after the initial instant[22]; particle α moves along a straight-line path (its "leg") from its last collision with members of the group $[s_\alpha]$ to its first collision with other members of the group $[c]$. There is still the possibility, however, that, after colliding with other members of $[c]$, particle α may become reinvolved with the group $[s_\alpha]$.

The elimination of this possibility is the role of the *second* condition which is that once the connecting particle (if any) of one of the groups $[m_\lambda]$ has begun a collision with a particle of some other group, this connecting particle is dynamically independent of the remaining members of its group. Thus, in a group $\{\alpha,[s_\alpha]\}$ the particle α is dynamically independent of $[s_\alpha]$ after it begins its involvement with other members of the group $[c]$. When this condition is satisfied, the set $[c]$ of all connecting particles has a phase point (projection of $[m]$ onto the space of the set $[c]$) which is complete. Furthermore, each of the sets $[s_\alpha]$ are dynamically independent subsets of $[m]$ during some portion of the time following the initial instant; in fact, the group $[s_\alpha]$ say, becomes dynamically independent of all other particles after its involvement with particle α is finished.

When these two conditions are satisfied by a point, it can certainly be schematized as in Fig. 2. There is a *third* condition, however, which must be fulfilled if the property of "being singly connected" is to be useful. One must also have that the point $[m]$ be such that for the prior trajectory of *each* $(1+s_\alpha)$-particle point $\mathcal{S}([c])(\alpha,[s_\alpha])$, the particle α is dynamically independent of $[s_\alpha]$ under $(1+s_\alpha)$-particle dynamics in an interval $[\tau_\alpha,t]$, where the time τ_α is between the initial instant and the time $t-T_\alpha([c])$, when the particle α begins its first collision with other members of $[c]$. The character of these times is indicated in Fig. 3 which gives the graphical meaning of this condition; namely, that *each* subgroup $\{\alpha,[s_\alpha]\}$ must have such a diagram where the kinked trajectory of the particle α represents its actual track under the natural motion of all m particles. The possibility being ruled out is a case where there is an "aiming to collide" between some members of $[s_\alpha]$ and the dashed track of particle α.[23]

When all three of these conditions are met, the groups $[s_\alpha]$ will be called singly connected. One can say that a group is singly connected if it collides only with the leg of a single particle and if it does so *while the leg is still "free."*

If a point $[m]$ is not-more-than-singly-connected, one has for $\xi([m];t)$ the following form:

$$\xi([m])=S([c])\prod_{\alpha\in[c]}\xi(\alpha,[s_\alpha])\prod_\beta\xi([d_\beta]),\quad(3.5)$$

where the sets are chosen in accordance with the above discussion; namely, $[c]$ is the set of connecting particles for the singly connected sets $[s_\alpha]$, while the $[d_\beta]$ are the disconnected sets. A heuristic proof of the validity of Eq. (3.5) is presented in Appendix B. It should be understood that Eq. (3.5) like Eq. (3.3) is an asymptotic expression in the sense that, if it is valid for a particular point $[m]$ at time t, it is valid for $[m]$ for all longer times. This, as has been mentioned, is a consequence of the assumed statistical and dynamical independence of the groups $\{\alpha,[s_\alpha]\}$ before the initial instant.[24]

As might be expected, the scattering operators $\mathcal{S}_m$ have a reduction which is analogous to the asymptotic form for Liouville functions. For not-more-than-singly-connected points and, in fact, for the slightly wider class of points for which no qualifications are made about correlations, one has that

$$\mathcal{S}([m])=\mathcal{S}([c])\prod_{\alpha\in[c]}\mathcal{S}(\alpha,[s_\alpha])\prod_\beta\mathcal{S}([d_\beta]),\quad(3.6)$$

[22] Otherwise, $[m_\lambda]$ and, say, $[m_{\lambda'}]$ would not be dynamically independent at the initial instant.

[23] In the sequel, consider the notion of collision history as amended to include this kind of information about the prior trajectory of $[m]$.

[24] Since the features of $[m]$, as exhibited in Figs. 2 and 3, which allow the steps in the derivation of the result, are still present if the $t=0$ line is shifted to earlier times.

where the sets have the same meaning as in Eq. (3.5).[25]

The generality and flexibility of these two results should be noted. For example, one may choose to lump any or all of the $[d_\beta]$ with one or several of the $[s_\alpha]$, and/or to make explicit use of the fact that any of the $[d_\beta]$ may have a structure like that of the point $([c]+\sum_\alpha[s_\alpha])$, and/or to make explicit some disconnected subgroups of $[c]$, and so forth. In any of these cases, obvious formulas specializing Eqs. (3.5) and (3.6) may be written at will. In particular, notice that by recognizing, for a given point, those groups of particles which are disconnected (e.g., in Fig. 2 the group $\{[c]+\sum_\alpha[s_\alpha]\}$ is itself disconnected), one derives for the Liouville function an analog of the product condition which is valid for an *interval* of time rather than for an instant.

D. General Asymptotic Form

For the points discussed so far, the correlation carried from the initial time is, in fact, just such as to make no contribution to the error. Furthermore, the asymptotic forms for them are essentially contained in papers I and II. We turn now to the most general kind of point some of which do indeed contribute to the error.

It is clear that, although we can always find a partition of $[m]$ into groups $[m_\lambda]$, which are statistically and dynamically independent before the initial instant, one cannot generally expect that the point is not-more-than-singly-connected. For an arbitrary point one must expect to find that *every* such partition has at least one subgroup containing *at least two* members which interact with some other group during the interval $[0,t]$, and such points will be called "more-than-singly-connected." Of course, we can still expect that some of the groups $[m_\lambda]$ will consist of single particles, while others will have a connecting particle α and its complement $[s_\alpha]$, such that $[s_\alpha]$ is singly connected, and still others will be totally disconnected. Thus, an arbitrary point $[m]$ for time t has a history which can be schematized as in Fig. 4, where the sets, $[i]$, $[c]$, and the $[s_\alpha]$ partition $[m]$ into $(c+2)$ disjoint parts and any or all of the $[s_\alpha]$ may be empty. Completely disconnected sets are not indicated but are to be considered as subsumed in the groups $[s_\alpha]$ in any convenient way.[26] The set $[i]$ is the part of $[m]$ which contains no singly connected parts like the $[s_\alpha]$; it may, in fact, split into statistically and dynamically independent sets before the initial instant, but this need not be indicated. We will refer to $[i]$ as the "incomplete more-than-singly-connected group" of the point or more briefly, as the "incomplete group."[27] A simple example of a point

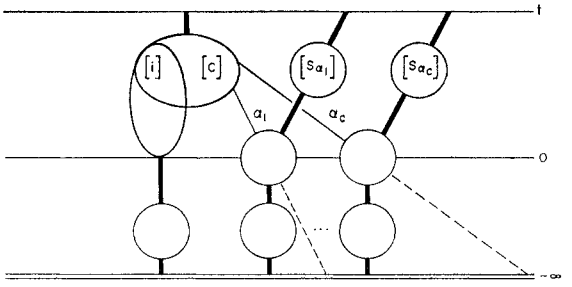

FIG. 4. Schematic of the history of more-than-singly-connected points. The projection of such points onto the space of the group $[i]+[c]$ illustrates a tightly connected point.

which is more-than-singly-connected is diagrammed in Fig. 9, where the set $[i]$ consists of particles 1 and 2 and the set $[c]$ consists of particle 3.

By arguments similar to those used to establish the previous asymptotic forms, it can be shown that, if $[m]$ is any more-than-singly-connected point (as schematized in Fig. 4), then one has for $\xi([m];t)$ that

$$\xi([m];t)=\mathcal{S}([i]|[c])\prod_{\alpha\in[c]}\xi(\alpha,[s_\alpha];t), \qquad (3.7)$$

where $[i]$ is the incomplete group of the point, $[c]$ is the complete group, the $[s_\alpha]$ are the singly connected groups, and all these sets form a partition of $[m]$ into disjoint parts. The operator $\mathcal{S}([i]|[c])$ is defined by

$$\mathcal{S}_{i,c}([i]|[c])\equiv\xi([i]_{-t};0)\mathcal{S}([i]+[c]); \qquad (3.8)$$

that is, it is the operator in c-particle space which substitutes $\mathcal{S}([i]+[c])[c]$ for the phase point $[c]$, and multiplies the result by the number $\xi([i]_{-t};0)$, which depends on both the time and the initial correlations of the set $[i]$. In particular, we will adopt the conventions $\mathcal{S}_{0,c}=\mathcal{S}_c$ and $\mathcal{S}_{i,0}=\xi_i$.

A heuristic proof of the result given by Eq. (3.7) is indicated in Appendix B; the argument parallels that used to establish Eq. (3.5) and begins by proving the result for the case when there are no sets which are not-more-than-singly-connected. It should be emphasized that this expression for $\xi([m];t)$ is valid for *any* point $[m]$, because the sets $[i]$, $[c]$, and the $[s_\alpha]$ can always be chosen as described.

One perhaps anticipates that the generalized scattering operator $\mathcal{S}([m_1]|[m_2])$ has properties consistent with its "mixed" nature (i.e., a function of time as well as an operator). With respect to the set $[m_2]$, its behavior is similar to that of $\mathcal{S}([m_1]+[m_2])$, while, with respect to the set $[m_1]$, it has properties like those of $\xi([m_1]+[m_2];t)$. In fact, if $[m]$ is a more-than-singly-connected point and if $[m_1]$ and $[m_2]$ partition $[m]$ into two disjoint groups, then one has that

$$\begin{aligned}\mathcal{S}([m_1]|[m_2])&=\mathcal{S}([i_1]|[i_2]+[c_1]+[c_2])\prod_{\alpha\in[c_1]}\mathcal{S}(\alpha,[s_{1\alpha}]|[s_{2\alpha}])\\&\times\prod_{\alpha\in[c_2]}\mathcal{S}([s_{1\alpha}]|\alpha,[s_{2\alpha}])\prod_\beta\mathcal{S}([d_{1\beta}]|[d_{2\beta}]),\end{aligned} \qquad (3.9)$$

[25] To see this apply the argument of Appendix B to $S_{-t}([m])\times\Pi_{\alpha\in[m]}S_t(\alpha)$ and then take the limit.

[26] The extension of the result to be given to the case where some of the $[m_\lambda]$ are entirely disconnected will be obvious.

[27] Notice that if, for a given point at time t the group $[i]$ is empty, the point is not-more-than-singly-connected. Alternatively, for a given point, when the time becomes so large that the particles in $[i]$ become statistically and dynamically independent, the point becomes not-more-than-singly-connected.

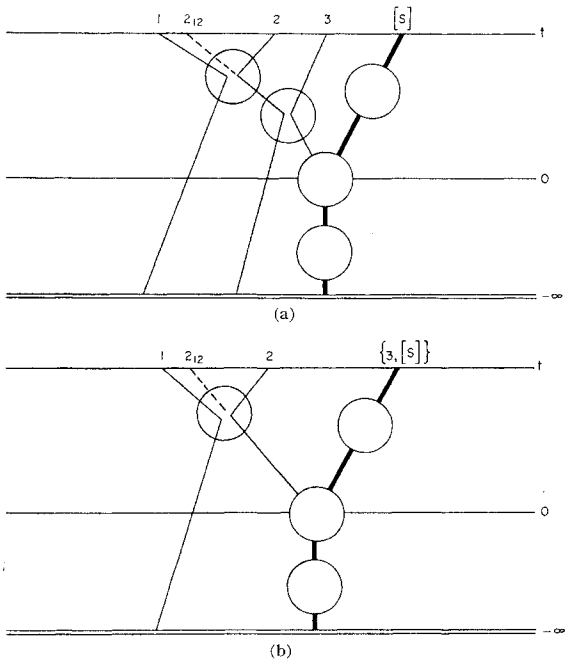

FIG. 5. Diagram (a) illustrates a history which is a case of those schematized in (b) and for which a further reduction in the asymptotic form is possible.

where the subscripts 1 and 2 on the various sets indicate whether the set is contained in $[m_1]$ or $[m_2]$ while the "name" of the set has its previous meaning; for example, $[i_1]$ is the part of the incomplete group of the point which is contained in $[m_1]$ while $[i_2]$ is the part in $[m_2]$. Although a proof of this general result will not be given, a result which is essentially a special case of Eq. (3.9) is established in Appendix B and the argument used for this purpose is easily generalized.

E. Additional Reductions

In the preceding discussion various aspects of the histories of phase points have been characterized by introducing concepts of completeness and connectivity. The results which have been established in terms of these concepts are the asymptotic forms for Liouville functions embodied in Eqs. (3.3), (3.5), and (3.7) and the corresponding equations for the scattering operators, $\mathcal{S}_m$ and $\mathcal{S}_{m_1m_2}$. Indeed, the form given by Eq. (3.7) includes all the others as special cases. These asymptotic forms corresponding to schematics of histories are, however, only the *general ones*, which apply to a whole class of histories. For any particular member of such a class, further reductions, the form of which depends on the point, are usually possible. Since character of such reductions should be understood in analyzing the error, some elaboration will be given.

For example, consider a history which can be diagrammed as in Fig. 5(a). This is one of those having a schematic as in Fig. 5(b). According to Eq. (3.5), the asymptotic form for points the schematic of which is as in Fig. 5(b) is

$$\xi(123[s]) = \mathcal{S}(12)\xi(23[s]). \tag{3.10a}$$

According to Fig. 5(a), the same form applies to, $(2_{12}3[s])$ where $2_{12} = \mathcal{S}(12)2$; that is,

$$\xi(2_{12}3[s]) = \mathcal{S}(2_{12}3)\xi(3[s]). \tag{3.11}$$

Hence, for points having a history as in Fig. 5(a),

$$\xi(123[s]) = \mathcal{S}(12)\mathcal{S}(23)\xi(3[s]). \tag{3.10b}$$

Clearly, the point, $\mathcal{S}(12)\mathcal{S}(23)(3,[s])$ may also have singly connected parts and then further reductions of the same kind could be made by repeated application of Eq. (3.5). It should be noted that the product of the $\mathcal{S}_m$ which has appeared in Eq. (3.10b) is *ordered* according to the order of the collision events for the point.

The general result suggested by these remarks is indeed correct. It is that, for a point with a particular not-more-than-singly-connected history, the asymptotic form can be written as a "corresponding" *not-more-than-singly-connected, ordered product* of the operators $\mathcal{S}_m$, operating on the appropriate product of ξ_m's, where some of the ξ_m are "hooked on" to the operators and some are entirely disjoint. Here, a not-more-than-singly-connected product of the $\mathcal{S}_m$ is one in which any factor has an argument containing no more than one member in common with the argument of any predecessor.

Similarly, consideration of examples of particular histories of more-than-singly-connected points shows that Eq. (3.7) can also be successively applied to "subdiagrams" of a schematic such as the one in Fig. 4; i.e., each of the sets $[s_\alpha]$ may have a schematic of this kind, and so on. The result suggested is again the correct one; namely, that any more-than-singly-connected point with a particular history has an asymptotic form which is a corresponding *not-more-than-singly-connected, ordered product* of the $\mathcal{S}_{m_1m_2}$ operating on an appropriate product of the ξ_m, where again some of them are hooked on and some are not. A not-more-than-singly-connected product of the $\mathcal{S}([i]|[c])$ is one in which any factor has an argument, say, $[i_\sigma]+[c_\sigma]$, containing no more than one member in common with the *complete part*, say, $[c_{\sigma'}]$, of the argument of any predecessor; that is, if the arguments of two factors do have a common member it is contained in the complete part of the factor which occurs first.[28]

To summarize this entire discussion of the properties of the Liouville functions and the scattering operators naturally associated with them, one can say that, for initial values which satisfy the product condition, a Liouville function can be expressed by a product of Liouville functions of lower order for those groups of particles which are not more than singly connected,

[28] For example, one has $\mathcal{S}(12|3)\mathcal{S}(45|3)$ and $\mathcal{S}(12|3)\mathcal{S}(34|5)$ but not, say, $\mathcal{S}(12|3)\mathcal{S}(14|5)$.

wherein each factor of the product the single particle through which the others are connected is taken at a point determined by the solution of the scattering problem for all those particles which are more-than-singly-connected, and where the whole product is multiplied by a factor determined by the full dynamics of these particles and the initial correlations among those which are incomplete. This is the result expressed by Eq. (3.7). In addition, the generalized scattering operators $\mathcal{S}([m_1]|[m_2])$ have a similar reduction determined by the history of the point $([m_1]+[m_2])$. Several other results which were given can be considered as special cases of these two.[29] We terminate our discussion by remarking that the same expressions which are valid asymptotically can also be valid for a *given* time t under weaker conditions than those stated; namely, when only those parts of the given conditions which are relevant to the structure *during* the interval, $[0,t]$, are satisfied.[30] Then, however, the forms do not necessarily apply for longer times.

IV. THE TERMS IN THE ASYMPTOTIC FUNCTIONAL

The asymptotic functional $f([n]|f_1)$ has already been assumed to have the structure of a "power series" in $f_1(t)$. To determine the coefficient operator $\tau_l^{(n)}$ of this series a principle suggested by the long-time behavior of Liouville functions will be used. The principle states that there should be no contribution to the error from certain points. Imposing such a requirement yields a recursion relation for the $\tau_l^{(n)}$ which defines them to be a sum of not-more-than-singly-connected products of the scattering operators $\mathcal{S}_m$. With the $\tau_l^{(n)}$ so defined, $f([n]|f_1)$ is completely determined and the question of the finiteness of the integrals arises. It is shown that the $\tau_l^{(n)}$ vanish identically for certain points whose contributions could lead to divergence of the integral.

A. Determination of the $\tau_l^{(n)}$

It was shown in Sec. IB that the error made by supposing f_n to be a time-independent functional of f_1 can be expressed [see Eq. (2.4)] as a series of integrals whose integrands are sums of products of Liouville functions, some of which are operated upon by, say, $\tau_q^{(n)}$. The integrand of the lth term in $\mathcal{E}_n$ is parameterized by the phase $[n]$ and is a function of the one-particle phases of the set $[l]$, which are the variables of integration. Referring to Eqs. (2.4) and (2.5), we see that the Liouville functions which appear depend on these variables in two ways: either directly as arguments or indirectly in that, because of the operation of one of the substitution operators in, say, $\tau_q^{(n)}$, for some of the ξ_m one of the arguments is itself a function of the phase of some of the particles in the set, $[n]+[l]$. Therefore, any ξ_m which appears has an argument which is a definite point in m-space and this point is completely determined as soon as the point $([n]+[l])$ is fixed.

We have said in Sec. IIIB that almost every point becomes complete so that for, fixed $[m]$, $\xi([m];t)$ eventually attains the form given by Eq. (3.3). Therefore, we can say that, for any fixed point in $(n+l)$-particle space (since then the arguments of all the ξ_m are fixed), there is always a time long enough so that every ξ_m in the integrand of the lth term of the error can be replaced by the form given in Eq. (3.3). The resulting expression for the integrand for such points, which will be called "complete in the wider sense," is that

$$\sum_{[h]+[k]=[l]} \mathfrak{D}^{(n)}([n];[h];t)\vartheta_0([k];t) = \sum_{[h]+[k]=[l]} D^{(n)}([n];[h])I_0([k]) \times \prod_{\alpha\in[n]+[l]} \xi_1(\alpha;t), \quad (4.1)$$

where $D^{(n)}([n];[h])$ and $I_0([k])$ are defined by the expressions which result when, in the defining equations for $\mathfrak{D}^{(n)}([n];[h])$ and $\vartheta_0([k])$ [i.e., Eqs. (2.5) and (2.7)], one simply replaces each factor $\xi([m])$ by a factor $\mathcal{S}([m])$ so that, for example, I_0 is defined in terms of $\mathcal{S}_m$ in the same way as ϑ_0 is defined in terms of ξ_m. Because of the nature of points which are complete (w.s.) if Eq. (4.1) is valid for a given $(n+l)$-particle point at time t, it remains valid for all longer times.[31] More importantly, this form is valid in a region of $(n+l)$-particle space which grows with time and eventually becomes essentially the whole space.

Now, if the asymptotic functional $f([n]|f_1)$ is to become a better approximation as the time increases, the error must decrease in time and we may suppose that it does so term by term. But, because the region where Eq. (4.1) applies grows in time, unless the integrand of the lth term vanishes for this region, we would expect a contribution to this term which would either approach a constant value or diverge with time. We will therefore choose the $\tau_q^{(n)}$ so that the integrand of the lth term vanishes for all points which are complete in the wider sense. Since for this it is necessary and sufficient that the difference $D^{(n)}$, which appears as a factor in each term of Eq. (4.1), vanishes for such points, this means that we require the $\tau_l^{(n)}$ to be defined by the equation:

$$\mathcal{S}([n]+[l]) = \sum_{\substack{[v]+[w]=[n]+[l] \\ [n]\subset[v]}} \tau^{(n)}([v])A([v];[w]), \quad (4.2)$$

[29] It is noteworthy that the $\psi_l^{(n)}$ and the corresponding "Ursell" operator (see Sec. IVB) have analogous properties which are implied by these properties of the ξ_m.

[30] For example, for the point diagrammed in Fig. 6 particle 4 may be disconnected during the interval t and one can write the appropriate case of Eq. (3.6) *until* the time $t+\Delta t$.

[31] For brevity, the phrase "in the wider sense" will often be denoted by "(w.s.)."

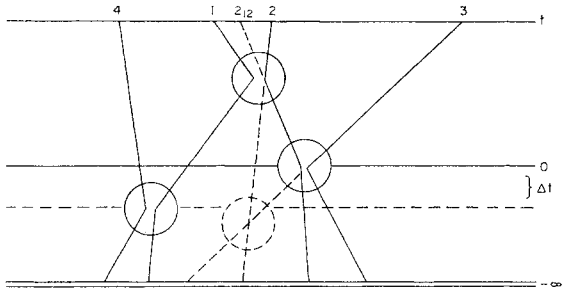

FIG. 6. An example of a not-more-than-singly-connected point. If only the aiming to collide between particles 2 and 3 were present, particle 3 would still be singly connected in a certain sense.

which is simply a transcription of the statement that $D^{(n)}$ vanishes.[32] The quantity $A([u];[v])$ is the sum of products of the $\mathcal{S}_m$ which results when one substitutes $\mathcal{S}_m$ for ξ_m in the expression for $\mathcal{C}([u];[v])$ given by Eq. (2.6).

Equation (4.2) is a recursion relation for the $\tau_l^{(n)}$. It can be completely solved and yields the result that $\tau_l^{(n)}$ is equal to the sum over all *distinct*, not-more-than-singly-connected, *ordered* products of operators S_m, where $m \geqslant 2$, with the following conditions: (a) The set of indices (arguments) involved in any product includes the set $[n]$ and is included in the set $[n]+[l]$; and (b) the first $\mathcal{S}$ operator in any product contains at least all of $[n]$.[33]

Since the explicit expressions for $\tau_l^{(n)}$ are not needed in the sequel, a proof of this statement will not be given. For completeness, however, we include in Appendix C a statement of the detailed form of the coefficient of each term of the sum which defines $\tau_l^{(n)}$ and also the explicit expressions for the first few sums $\tau_l^{(2)}$, up to $l=4$.

With the $\tau_l^{(n)}$ explicitly given in terms of the scattering operators $\mathcal{S}_m$, *all* the terms in the functional series for $f([n]|f_1)$ are completely determined for any value of n.[34] Thus, the question of the detailed form of the functional series is answered. Since the argument just given was only heuristic, it remains to be shown that f_n does indeed approach this particular functional.

B. Convergence of the Integrals

Before analyzing the error which is now determined by this definition of the $\tau_l^{(n)}$, we wish to show that certain points, the contributions from which *could* cause the terms in $f([n]|f_1)$ to diverge, in fact, make no contribution to the integrals because the $\tau_l^{(n)}$ vanish for such points.

The points in question are those containing groups which are essentially of the kind we have called singly connected. That such groups lead to the *growth* or *divergence* of the kind of cluster integral we encounter is suggested by the following analysis for the modified Ursell functions $\psi_l^{(n)}$ and the analogous operator $U_l^{(n)}$, which is defined in terms of the $\mathcal{S}_m$ in the same way as the $\psi_l^{(n)}$ are defined in terms of the ξ_m.

Considering first the functions $\psi_l^{(n)}$, which are the integrands of the terms in the series for f_n, it has already been pointed out in papers I and II that the regions where they are nonvanishing have significant features which were called "growing legs," so named because their length increased with the time. In fact, each growing leg corresponds to a subgroup of particles which is singly connected in a certain sense. The growth in time of the integral over such a region is a consequence of fact that the integrand is independent of the length of these growing legs.

The existence of growing legs and the lack of dependence on their length is illustrated by the following example. Suppose that the 2-particle point (1,2) is complete for time t and consider the integral of $\psi^{(2)}(12;3,t)$ over those phases of particle 3, such that the 3-particle phase point (1,2,3) has the infinite history represented in Fig. 6 (i.e., project out particle, 4, from this diagram). For the history of the point (1,2,3), represented by this diagram, it is intended that the $(2_{12}-3)$ collision occurs anywhere before the $(1-2)$ collision and that there are no additional "aimings to collide" between particle 3 and any extension to earlier times of any particle (e.g., the indicated aiming to collide between particle 3 and the extension of particle 2 does *not* occur). In the sense of including this last qualification, particle 3 is singly connected to the others. Thus, for fixed momentum of particle 3 and fixed impact parameter of the $(2_{12}-3)$ collision, the integration is over the location of this collision and corresponds to "sliding" particle 3 along the leg.

Now, for fixed time t $\psi^{(2)}(12;3,t)$ vanishes if this collision occurs *before* the initial time and does not vanish if it occurs after.[35] Hence, in the space of the variable $(\mathbf{r}_{2_{12}}-\mathbf{r}_3)$, this function is nonvanishing in a cylinder whose length is proportional to the time, and this cylinder is the "growing leg." Furthermore, for the points being considered, the value of this function is independent of the *location* of the collision (i.e., the length of the collision cylinder).[35] Thus, the integration over the sliding variable can be performed making the contribution of such points proportional to the time.

Similar circumstances cause, say, the lth term to have contributions proportional to the time. Furthermore,

[32] For convenience, the notation of Eq. (2.5) has been simplified by suppressing the distinction between the two sets in $\tau_l^{(n)}$.

[33] Not-more-than-singly-connected products of scattering operators have been defined in Sec. IIIE.

[34] It may be remarked that Bogoliubov's asymptotic expressions for the probability densities, while also given as a density series, *seem* to have coefficients of different form. It has been pointed out that, at least for the first few terms of f_2, the expressions are equivalent. E. G. D. Cohen, however, has given expressions for the coefficients τ_l^n, which have the same form as those given here and which are identical with them for $n=2$ up to four particles.

[35] To see this notice that regardless of when it occurs, for the point being considered $\xi(123)$ is equal to $\mathcal{S}(12)\xi(23)\xi(1)$ and $\xi(12)$ is equal to $\mathcal{S}(12)\xi(1)\xi(2)$ so that the value of $\psi^{(2)}(12;3)$ is $\mathcal{S}(12)[\xi(23)-\xi(2)\xi(3)]\xi(1)$. If the collision occurs before the initial instant $\xi(2_{12}3)$ is equal to $\xi(2_{12})\xi(3)$ and only then does this expression vanish. This means that regions for which particle 3 is far from the others contribute significantly to the integral.

one can verify that for any point of the kind schematized by Fig. 2, where the groups $[s_\alpha]$ are singly connected in an appropriate sense, $\psi_l^{(n)}$ has a value which is independent of the locations of the connecting collisions and which vanishes if any of them occur prior to the initial instant. Since the integration over such points can be arranged to involve sliding the location of the connecting collisions, one has that, in general, groups which are singly connected in an appropriate sense lead to growth of the integrals of $\psi_l^{(n)}$.[36]

Whereas groups which are singly connected in a certain sense lead to growth of integrals of $\psi_l^{(n)}$, they lead to divergence of integrals whose integrands involve time-independent operators of the cluster type. Such integrals arise, for example, if one notices that there is a time so large that one can use Eq. (3.3) to express every ξ_m occurring in, say, the integrand $\psi_l^{(n)}$. Also, for very large times, the volume in phase space where this happens becomes very large. Hence, one might try to approximate the integral over $\psi_l^{(n)}$ by the integral over the asymptotic value of the integrand; that is, by

$$\int d([l])U^{(n)}([u];[l]) \coprod_{\alpha\in[n]+[l]} \xi_1(\alpha;t),$$

where $U_l^{(n)}$ are the modified Ursell operators already defined. But, by reasoning parallel to that used for the original integrals of $\psi_l^{(n)}$ one can see that there are *infinitely long* regions in configuration space where the integrands $U_l^{(n)}$ are nonvanishing and have a value independent of the variable locating position *along* the cylinder. We call these regions "infinite legs"; they again correspond to groups which are singly connected (in the same sense as is relevant to the discussion of $\psi_l^{(n)}$) and, clearly, they cause divergence of the integrals (so that the attempted approximation fails).

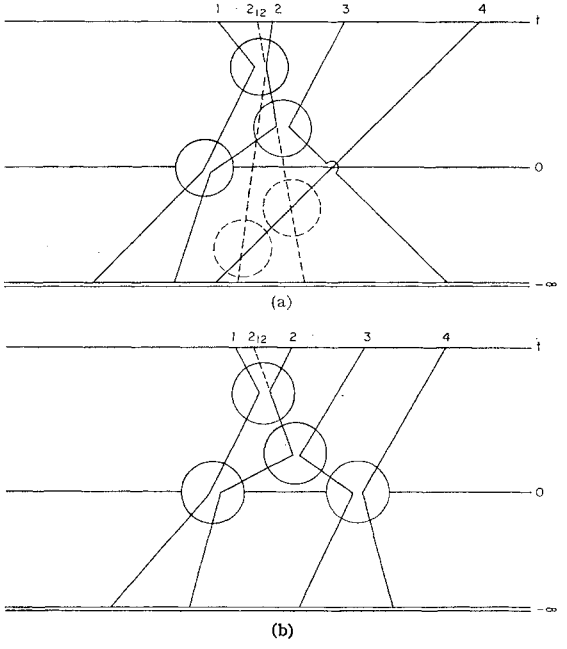

FIG. 7. An example of a more-than-singly-connected point; particle 4 is singly connected (w.s.) in (b) or if only one of the aimings in (a) occurs.

[36] In particular, the lth term has a contribution proportional to t^l arising from the case when *each* of the l particles is singly connected.

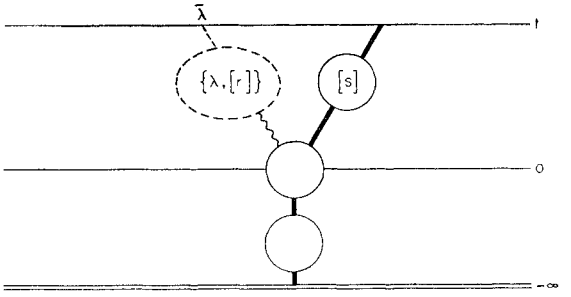

FIG. 8. An illustration of point which is arbitrary except that the group $[s]$ is not-more-than-singly-connected (w.s.) to the others through *some* leg of the connecting particle λ.

Now, the terms in the functional series given by Eq. (1.2) with the $\tau_l^{(n)}$ defined by the Eq. (4.2) are similar to the above integrals involving the $U_l^{(n)}$. Again, infinite legs corresponding to groups which are singly connected in some wider sense would provide an obvious source of divergence. Thus, to see that the functional series has more than purely formal character, we want to show that this source of divergence is ineffective.

The reason for again qualifying the meaning of singly connected particles is this: To discuss the behavior of the $\psi_l^{(n)}$ or $U_l^{(n)}$, one has to know whether a supposed singly connected set has aimings to collide with "free-particle extensions" of the final phases of any particles other than some one connecting particle. In fact, such aimings alone (without any actual collisions) can serve to singly connect one or several particles.[37] In considering the integrals in $f([n]|f_1)$ one must expect that aimings to collide with free-particle extensions of "derived points," which are produced by the operation of scattering operators, become relevant (simply because to evaluate $\tau_l^{(n)}$ one must evaluate scattering operators at such points). For example, for the integration of the 4-particle term in the asymptotic functional $f(12|f_1)$ over the position of particle 4 (with all other variables fixed), the point with a history as in Fig. 7(a) where one or the other (not both) aimings occurs corresponds to an infinite leg just as does the point diagrammed in Fig. 7(b). Indeed, for the terms in $f([n]|f_1)$, infinite legs correspond to points for which there are groups which are not-more-than-singly-connected *in the wider sense*.

A sufficient characterization of such groups is the

[37] For example, a discussion similar to that just given applies for points indicated by the case of Fig. 6 for which the (2–3) aiming does occur while the (2_{12}—3) collision does not. Thus, in discussing integrals of $\psi_l^{(n)}$ or $U_l^{(n)}$, particle 3 should be considered as singly connected.

following: A group of particles will be said to be not-more-than-singly-connected in the wider sense if (a) it interacts with all other particles being considered *at most* through a single leg, *virtual* or *real*, of a single connecting particle, and (b) if this interaction occurs (if at all) while the leg of the connecting particle is still "free." This means that the relation of a group which is singly connected (w.s.) to others can be schematized as in Fig. 8 where the group $[s]$ has *no other* aimings to collide except the one indicated by the wavy line.

Consider the integration of the lth term in $f([n]|f_1)$ over points for which the set $[l]$ contains a group $[s]$ which (as in Fig. 8) is not-more-than-singly connected (w.s.). The integration can be arranged to involve "sliding" the location of the collision of the set $[s]$ with particle λ to arbitrary early times. Thus, the integral will diverge because of an infinite leg *unless* the coefficient operator, $\tau_l^{(n)}$ vanishes for such regions. But, in fact, we have proved that: *If a point* $([n]+[l])$ *is such that* $[l]$ *contains a group* $[s]$ *which is not-more-than-singly-connected (w.s.) then* $\tau^{(n)}([n];[l])$ *vanishes identically.* This result is a special case of a more general one discussed in the next section and a description of the proof of it is given in Appendix E. There is nothing in this statement to preclude that the set $[l]$ constituting the integration variables, in fact, contains *many* groups which are not-more-than-singly-connected (w.s.) (i.e., the region of integration may be the wider sense analog of Fig. 2), so that we have proved that none of the many possible infinite legs cause divergence of the terms in the asymptotic functional.

With this result which is, of course, not sufficient to prove convergence of the terms in the asymptotic functional,[38] we leave the discussion of the character of the asymptotic form and begin a discussion of the error.

V. ANALYSIS OF THE ERROR

Recalling the discussion leading to the definition of the coefficient operators $\tau_l^{(n)}$, one can also take the converse position and say that if the $\tau_l^{(n)}$ are defined by Eq. (4.2), then the factors $\mathfrak{D}_l^{(n)}$ in the error integrand and hence the error integrands themselves vanish at least for points which are complete in the wider sense. In fact, it can be shown that this conclusion is also true for any point which is not-more-than-singly connected in the wider sense (i.e., which can be diagrammed by the wider sense analog of Fig. 2). Nevertheless, the error does not, of course, vanish for all points and, in particular, it does not vanish for any point for which there is an incomplete group $[i]$ (as illustrated in Fig. 4). To establish the asymptotic theorem one must understand the contributions such points make to the error after very long times.

For the expression of the error derived in Sec. II, however, the contributions from points having groups of particles which are singly connected in the wider sense cause the terms to grow with time. The reason for this is that the integrands of the terms are analogous to the $\psi_l^{(n)}$; the regions where they are nonvanishing possess growing legs, each corresponding to a group of particles which is singly connected in the wider sense. Moreover, the value of the error integrand is independent of the location of the connecting collisions (i.e., of the length of the legs) so that the integration over sliding variables can be performed yielding a result which grows with increasing time. These remarks are illustrated, for example, by the contribution to the 4-particle term of the error $\mathcal{E}(12;t)$, from the point diagrammed in Fig. 7(b); one finds that the integration over particle 4 yields a result which is proportional to the time.[39]

This growth with time of the terms of the expression for the error given by Eq. (2.4) does *not* mean that the proposed asymptotic form is incorrect. Rather, it only implies that, like the original series for f_n, this particular expression for the error is suitable for short times but not for long times. Or, one can say loosely that in this form one has expanded too far, representing decreasing functions of the time by power series in the time, so that one must somehow partially sum up again.

Formal analogy with the structure and properties of the asymptotic functional suggests an expression for the error as a functional series in powers of $f_1(t)$; it is the products of $f_1(t)$ in this series which were previously "expanded." The coefficient operators which appear will be determined so that this series is indeed formally identical to the original one. Moreover, it will be shown the terms in the new series do not grow in time because of growing legs of the integrands. Finally, a partial evaluation of the error which is valid for large times will be derived.

A. The Error as a Functional of $f_1(t)$

The form of the terms in the error expressed as a power series in $f_1(t)$ is suggested by the terms in the asymptotic functional. There is an obvious correspondence between any of these terms and the schematic of a complete point (none of which has any singly connected sets hooked on to its legs); namely, for each such point $[c]$, where, say, $[n]\subset[c]$, there is a term with the integrand $\tau^{(n)}([c])\prod_{\alpha\in[c]}f_1(\alpha;t)$. As we have already remarked, the difference between f_n and this series of terms (i.e., the error, $\mathcal{E}_n$) has no contributions

[38] See Sec. VIB for further remarks.

[39] The integrand to which we refer involves the four cases of $\mathfrak{D}^{(2)}(12;[l])$ which occur for the four subsets of $\{3,4\}$. Because the $(1-2)$ collision is complete, $\mathfrak{D}^{(2)}(12;0)$ vanishes. The fact that particle 4 is singly connected (w.s.) via the leg 3_{123} implies, for example, that $\mathcal{S}(12)\xi(14)$ is equal to $\mathcal{S}(12)\xi(1)\xi(4)$ and $\mathcal{S}(12)\mathcal{S}(23)\xi(14)$ is equal to $\mathcal{S}(12)\mathcal{S}(23)\xi(1)\xi(4)$; in fact, it implies [by using the appropriate case of Eqs. (3.5) or (3.6)] that $\mathfrak{D}^{(2)}(12;4)$ vanishes. In conjunction with the appropriate form of Eq. (3.7) for $\xi(1234)$ and $\xi(123)$, it also implies that the integrand has the value $[\mathcal{S}(12|3)-\mathcal{S}(123)\xi(1)\xi(2)](\xi(34)-\xi(3)\xi(4))$ [the operator multiplying $\xi(3)$ is $\mathfrak{D}^{(2)}(12;3)$ and the other term is $\mathfrak{D}^{(2)}(12;34)$]. Clearly, this expression vanishes if the $(3_{123}-4)$ collision occurs before the initial time and this yields the growing leg.

from points which are complete (w.s.), and *in addition*, none from any point which is not-more-than-singly-connected (w.s.).

Now, let a more-than-singly-connected point which has no singly connected groups hooked on be called a "tightly connected point."[40] Noticing the similarity of these points to complete points, consider a representation of f_n by a generalized series of terms each of which corresponds to the schematic of a tightly connected point; namely, for each such point, $([i]+[c])$, where, say, $[n]\subset[i]+[c]$, there is a term with the integrand $\tau^{(n)}([i]|[c])\prod_{\alpha\in[c]}f_1(\alpha;t)$. It is natural to expect by formal analogy that the difference between f_n and such a series has no contributions either from points which are tightly connected (w.s.) or from *any* general more-than-singly-connected point (w.s.), which does have singly connected groups hooked on as in Fig. 4.[41] Since *any* point is *some* more-than-singly-connected point, the suggestion is, therefore, that the coefficient operators $\tau^{(n)}([i]|[c])$ can be determined so that this generalized series is a formal *identity*. Moreover, because the terms in this generalized series for f_n which correspond to points having no incomplete part $[i]$ should be just the terms of the asymptotic functional, the suggested identity for the error can be formalized as

$$\mathcal{E}([u];t)=\sum_{l\geqslant 0}\frac{1}{l!}\int d([l])\sum_{\substack{[i]+[c]=[n]+[l]\\ i\geqslant 1}}\tau^{(n)}([i]|[c])\times\prod_{\alpha[\in c]}f_1(\alpha;t), \quad (5.1)$$

where the terms for $i=1$, which are included here for simplicity, are, in fact, found to be necessary.

Thus far, the analogy with the asymptotic functional suggests only a form of the terms in the error, but by pursuing it further, one can also understand the nature of the generalized coefficient operators $\tau^{(n)}([i]|[c])$. Notice that if one expresses the factors $f_1(t)$ in terms of Liouville functions by using the series for f_1 in terms of them [Eq. (2.1)], the asymptotic functional is thereby expressed as a sum over all terms of the form

$$\tau^{(n)}([c])\prod_{\alpha\in[c]}\xi(\alpha,[q_\alpha])\prod_{\beta=1}^{p}\xi([r_\beta]),$$

where $[c]$, the $[q_\alpha]$, and the $[r_\beta]$ form a partition of, say, $[n]+[l]$ into disjoint parts such that $[n]\subset[c]$ and one or more of the sets $[q_\alpha]$ may be empty.[42] Since, according to Sec. IVA, $\tau^{(n)}([c])$ is a sum of all not-more-than-singly-connected products of the $\mathcal{S}_m$, this means that a typical term of the asymptotic functional is a not-more-than-singly-connected product of $\mathcal{S}_m$ operating on a product of the ξ_m, some of which are hooked on and some of which are not and, moreover, that it is the sum of all such terms. According to the discussion in Sec. IIIE, such terms are the possible asymptotic forms for not-more-than-singly-connected points. Therefore, we can describe the asymptotic functional as the sum of *all the possible asymptotic forms for not-more-than-singly-connected points*. To pursue our analogy further, it seems natural to insist that the coefficient operators $\tau^{[n]}([i]|[c])$ be *some* linear combination of all not-more-than-singly-connected products of the generalized scattering operators $\mathcal{S}([m_1]|[m_2])$ where $[m_1]\subset[i]$. For, then the proposed generalized series for f_n, consisting of the sum of the asymptotic functional and the expression for the error given by Eq. (5.1), can be described as the sum of *all possible asymptotic forms*.

B. Determination of $\tau^{(n)}([m_1]|[m_2])$

To determine *which* linear combination of not-more-than-singly-connected products of the $\mathcal{S}_{m_1m_2}$ defines $\tau^{(n)}([i]|[c])$, we will use a method analogous to the one used to determine the operator coefficients of the asymptotic functional.

Since the expression for $\mathcal{E}_n$ given by Eq. (5.1) is supposed to be an identity, the difference between f_n and the sum of this expression and the asymptotic functional should vanish identically. An expression for this difference in terms of Liouville functions can be derived in parallel with the derivation of Eq. (2.4) for the error; this argument is also indicated in Appendix A. The result has the structure of Eq. (2.4) and differs only in that the sum in the factor $\mathfrak{D}_l^{(n)}$ which appears in the terms of the integrands is replaced by a sum over the terms $\tau^{(n)}([u]|[v])\mathfrak{A}([v];[w])$. In order that the terms of the difference between f_n and its representation as a functional series vanish identically, it is necessary and sufficient that these analogs of the factors $\mathfrak{D}_l^{(n)}$ vanish. Moreover, the vanishing of these factors is transcribed by the equation

$$\xi([u]+[l])=\sum_{\substack{[u]+[v]+[w]=[n]+[l]\\ [n]\subset[u]+[v]}}\tau^{(n)}([u]|[v])\times\mathfrak{A}([v];[w]), \quad (5.2).$$

where for convenience we have put

$$\tau^{(n)}([v])\equiv\tau^{(n)}(0|[v]). \quad (5.3)$$

Thus, expressing the error as a functional of f_1 is equivalent to expressing a Liouville function by a decomposition into the sum of all its possible asymptotic forms; namely, by Eq. (5.2).

In order to derive the recursion relation which will determine the $\tau^{(n)}$ such that the decomposition, Eq. (5.2), is an identity, notice that since Eq. (5.2) is to hold for any $(n+l)$-particle point, it must certainly hold for any particular point we choose. Thus, just as we determined the $\tau_l^{(n)}$ by requiring that the factors $\mathfrak{D}_l^{(n)}$ vanish for points which are complete (w.s.), we

[40] Such points are more-than-singly-connected in the proper sense of being *only* more-than-singly-connected.

[41] At this point the qualification "in the wider sense" is used by analogy; the precise sense of it is given in Sec. VB.

[42] This remark is validated in Appendix A.

will require that Eq. (5.2) be an identity for points which are tightly connected (w.s.).

A point which is tightly connected (w.s.) has the history indicated by the wider sense analog of the case of Fig. 4 for which there are no particles $[s_\alpha]$, so that the early history of the particles $[c]$ is given by the dashed lines. The qualification "in the wider sense" means essentially that the particles $[c]$ have no aimings to collide. In other words, a group, say, $[i]+[c]$, has a phase point which is tightly connected *in the wider sense* if the implied phase and the "derived" phases[43] of every subgroup, say, $[m]$, are themselves tightly connected in such a way that the complete group for any one of these points consists of those of its particles which are in the complete group $[c]$ of all of the particles. Thus, for such a point, if $[m]$ is any projected point or one of its associated derived points, one has according to Eq. (3.7) that

$$\xi([m];t)=\mathcal{S}([m_1]|[m_2])\prod_{\alpha\in[m_2]}\xi_1(\alpha;t), \quad (5.4)$$

where $[m_1]$ is the part of $[m]$ in the incomplete group $[i]$ of the point while $[m_2]$ is the part in the complete group $[c]$.

Using the reduction given by Eq. (5.4), it is clear that for points which are tightly connected (w.s.), the terms of the decomposition, Eq. (5.2) can be evaluated and that the result is a relation between $\tau^{(n)}$ and the $\mathcal{S}_{m_1m_2}$. This result, however, does not determine the $\tau^{(n)}$. To find a recursion relation which does determine them, it will be sufficient to demand that the $\tau^{(n)}$ have the following property: Supposing that $[m_1]$ and $[m_2]$ partition the set $[n]+[l]$, *if any particles of the set* $[m_1]$ *have free legs* (w.s.), *then* $\tau^{(n)}([m_1]|[m_2])$ vanishes.[44] The notion of "having a free leg (w.s.)" is to be understood in the following sense: A particle of a complete group which has no singly connected group hooked on will be said to "have a free leg."[45] Moreover, if a particle remains a member of the complete group for any projected point and its associated derived points, then it has essentially *all* its legs free and it will be referred to as "having free legs in the wider sense."[46]

To arrive at the desired recursion relation using this property notice that for points which are tightly connected (w.s.) *every* particle of the complete group has a free leg (w.s.) so that, say, $\tau^{(n)}([m_1]|[m_2])$ vanishes unless its incomplete part $[m_1]$ is entirely contained in the incomplete group of the point. Thus, in the relation between $\tau^{(n)}$ and $\mathcal{S}_{m_1m_2}$ resulting from the application of Eq. (5.4) many terms simply vanish and one arrives at the following conclusion: Assuming that $\tau^{(n)}$ vanishes when any particles of its incomplete part have any free legs (w.s.), then for points which are tightly connected (w.s.) the validity of the decomposition, Eq. (5.2), implies the recursion relation

$$\mathcal{S}([i]|[c])=\sum\tau^{(n)}([u]|[v_1]+[v_2])\times A([v_1];[w_1]|[v_2];[w_2]), \quad (5.5)$$

where the summation is such that: $[u]+[v_1]+[w_1]=[i]$, $[v_2]+[w_2]=[c]$, $[n]\subset[u]+[v_1]+[v_2]$, and where $[i]$ and $[c]$ partition, say, $[n]+[l]$. The "generalized A sum," $A([v_1];[w_1]|[v_2];[w_2])$, is the mixed quantity analogous to the A sum appearing in the defining equation, Eq. (4.2), for the $\tau_l^{(n)}$; indeed, it is obtained from $A([v_1]+[v_2];[w_1]+[w_2])$ by replacing each factor $\mathcal{S}([m])$ by $\mathcal{S}([m_1]|[m_2])$ where $[m_1]$ is the part of $[m]$ contained in $[v_1]+[w_1]$. An explicit expression for the generalized A sum is given in Appendix D along with a slightly more explicit statement of the derivation of Eq. (5.5).

As might be expected by their similarity to Eqs. (4.2), Eqs. (5.5) have a solution of the desired form; the generalized $\tau^{(n)}$ is a sum of all not-more-than-singly-connected products of the $\mathcal{S}_{m_1m_2}$. Notice that for the case when the incomplete part $[i]$ is empty, Eqs. (5.5) reduce to Eqs. (4.2).

It might seem that we have only achieved a definition of the $\tau^{(n)}$ which makes the decomposition Eq. (5.2) an identity only for a particular kind of point. In fact, however, the recursion relations define $\tau^{(n)}$ which yield the desired identity for *any* point $([n]+[l])$. This is a trivial consequence of the fact that the "last" recursion relations, namely, Eq. (5.5) for the case when $[c]$ is empty, define the quantities $\tau^{(n)}([n]+[l]|0)$ in just such a way as to yield the identity regardless of how the others are defined.

Before continuing it should be verified that the $\tau^{(n)}$ defined by Eq. (5.5) have the property assumed in their derivation; namely, that any of them vanishes if its

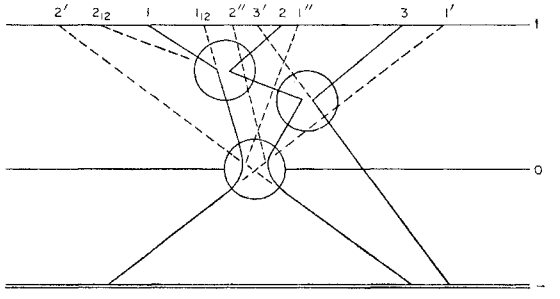

FIG. 9. A tightly connected point for which the incomplete group is {1,2}; points generated by various operators are illustrated.

[43] A "derived" phase of, say, $[m]$ is one for which one of the particles is displaced to a point produced by operating with a not-more-than-singly-connected product of $\mathcal{S}$ operators.

[44] This property may be considered as being a *particular way* of securing that, in evaluating Eq. (5.2) for some more-than-singly-connected point, terms which are not the appropriate asymptotic form for the point do not contribute. The appearance of objects like $\tau^{(n)}(\lambda|[v])$ in Eq. (5.2) is related to the possibility of determining $\tau^{(n)}$'s with this property.

[45] In Sec. IV, the leg of a particle which belonged to a complete group was "free" prior to its involvement with other particles; "having a free leg" means the trajectory is free during the *entire* prior time.

[46] For example, in Fig. 9 particle 3 has a free leg (w.s.), while in Fig. 10 it does not.

incomplete part contains particles having free legs (w.s.). A proof of this property can be given which parallels that of a related property to be discussed presently. Instead of displaying the formal proof, however, we will limit ourselves to an illustration of the property in a particular case.[47]

For this purpose, we consider the point which has a history given by Fig. 9 and show that $\tau^{(2)}(13|2)$ vanishes because particle 3 has a free leg. The solution of the recursion relation Eq. (5.5) for $\tau^{(2)}(13|2)$ in terms of the $\mathcal{S}_{m_1m_2}$ is given by

$$\begin{aligned}\tau^{(2)}(13|2)=&\mathcal{S}(13|2)-\mathcal{S}(1|23)\xi(3)\\&-\mathcal{S}(3|12)\xi(1)+\mathcal{S}(123)\xi(1)\xi(3)\\&-\mathcal{S}(12)[\xi(13)-\mathcal{S}(1|3)\xi(3)\\&\quad-\mathcal{S}(3|1)\xi(1)+\mathcal{S}(13)\xi(1)\xi(3)],\end{aligned}\tag{5.6}$$

where we have replaced $\mathcal{S}(0|[m])$ and $\mathcal{S}([m]|0)$ by their equivalents; that is, by $\mathcal{S}([m])$ and $\xi([m])$, respectively. Now, according to Fig. 9, the points 1 and 3 are uncorrelated at the initial time as are 1_{12} and 3. Hence, we have that

$$\begin{aligned}\mathcal{S}(13|2)=&S_{-t}(123)S_t(1)\dot{S}_t(3)\xi(1)\xi(3)\\&=\xi(1'')\xi(3''),\end{aligned}\tag{5.7a}$$

and that

$$\begin{aligned}\mathcal{S}(12)\xi(13)=&S_{-t}(1_{12}3)S_t(1_{12})S_t(3)\xi(1_{12})\xi(3)\\&=\xi(1_{12})\xi(3),\end{aligned}\tag{5.7b}$$

where the various points are indicated in Fig. 9. The remaining terms can be evaluated by inspection to yield, term by term, that

$$\begin{aligned}\tau^{(2)}(13|2)=&\xi(1'')\xi(3'')-\xi(1'')\xi(3')-\xi(1')\xi(3'')\\&+\xi(1')\xi(3')-\xi(1_{12})\xi(3)+\xi(1_{12})\xi(3)\\&+\xi(1_{12})\xi(3)-\xi(1_{12})\xi(3)=0,\end{aligned}\tag{5.8}$$

since $3'$ and $3''$ are the same point.

C. Behavior of Terms in the Error

Having determined the $\tau^{(n)}$ so that the decomposition Eq. (5.2) is an identity, we have ensured that the expression for the error in terms of f_1, given by Eq. (5.1), is also at least a formal identity and a definite algorithm. Whether this expression is useful for understanding the error for large times depends on the character of its terms. It will be shown that there are no contributions to these terms from either growing or infinite legs so that the only contributions are from points which are tightly connected (w.s.). An expression for the error which embodies this result and is valid for large times is derived.

Since the coefficient operators $\tau^{(n)}$ in Eq. (5.1) are mixed quantities, being partly functions and partly operators, there is the possibility that the regions where they are nonvanishing exhibit both the growing and infinite legs discussed in Sec. IVB. For any term, the growing legs might occur in the $[i]$ subspace and the infinite legs in the $[c]$ subspace.

For example, suppose that the 2-particle phase point (1.2) is complete for t and consider the integral of $\tau^{(2)}(14|23)f_1(2)f_1(3)$ over those phases of particles 3 and 4 such that (1,2,3,4) has a history which can be diagrammed by the case of Fig. 6 for which the $(2-3)$ aiming does not occur. In parallel to the argument in Sec. IVB concerning integrals over the $\psi_l^{(n)}$, if $\tau^{(2)}(14|23)$ vanishes when the $(1_{12}-4)$ collision occurs before the initial time, it will have a growing leg unless it also vanishes when this collision occurs after the initial time. The $(2_{12}-3)$ collision leads to an infinite leg (in the same way as these arose for the $U_l^{(n)}$), unless the integrand vanishes for all locations of this collision. Note that showing that the integrand vanishes for all locations of both collisions would establish that these particular possible growing and infinite legs do not occur.

This example suggests that not-more-than-singly-connected groups could lead to both growing and infinite legs for the integrands of the error. Furthermore, the discussion of the integrals involving the $\tau_l^{(n)}$ suggests that it is points having groups which are singly connected *in the wider sense* which lead to this behavior. Since the presence of growing legs would cause the error to increase in time while the presence of infinite legs would cause the terms to diverge, Eq. (5.1) would be useless if this actually occurred.

Therefore, one wants to prove a generalization of the previous result for the $\tau_l^{(n)}$; namely, that supposing that $[h]$ and $[k]$ partition $[n]+[l]$ *if the point* $([n]+[l])$ *is such that* $[l]$ *contains a group* $[s]$ *which is not-more-than-singly-connected* (w.s.), *then* $\tau^{(n)}([h][k])$ *vanishes*. The type of point contemplated here is again as in Fig. 8 where the point $(\lambda,[r])$ is unspecified; for example, it may be complete in whole or part and there may be subsets of $[r]$ which are themselves not-more-than-singly-connected (w.s.). We have found an inductive proof of this property which is described in Appendix E.

According to this property, $\tau^{(n)}$ certainly vanishes if there is more than one set which is not-more-than-singly-connected (w.s.); that is, it vanishes for all points which can be diagrammed by the wider sense analog of Fig. 4 for which $[n]\subset[i]+[c]$. Since such points are all the points which lead to either growing legs or infinite legs or both, we have proved that the terms in the expression for the error given by Eq. (5.1) neither increase with time nor diverge *because of such legs*.[48]

This property of the coefficient operators of the error together with the one previously given leads to an expression for the error for long times which makes the nature of the contributions explicit.

In considering, say, the n-particle error at a point $[n]$, a time is "long" if it is larger than the time $T([n])$

[47] A general proof is described in Appendix E.

[48] The possibility of secular behavior or divergence for other reasons still remains.

for which $[n]$ becomes complete. Such a criterion is suggested by the fact that the first term of the asymptotic functional is approximated by $\mathcal{S}([n]\prod_{\alpha\in[n]}\xi_1(\alpha;t)$. Since, in turn, this expression approximates ξ_n only for times larger than the time for the point $[n]$ to become complete, one can expect the first term of the asymptotic functional to approximate f_n in the same way. This criterion is also suggested by the behavior of the unintegrated terms (the $l=0$ case) in the error.[49] These terms contribute an essentially fixed error until the time the point $[n]$ becomes complete and then they vanish for this and all longer times. The fact that $\tau^{(n)}([n_1]|[n_2])$, where $[n_1]$ and $[n_2]$ partition the set $[n]$ (i.e., those in the unintegrated terms), vanishes if $[n]$ is complete can be established by an inductive proof.[50]

Consider, then, the n-particle error at a point $[n]$ which is complete for the time t. In addition to the vanishing of the unintegrated terms, one also has that the *only* nonvanishing contributions to the remaining terms are from points which are tightly connected (w.s.). This is a direct consequence of the second property of the $\tau^{(n)}$ since points which are tightly connected (w.s.) are just all those which have no groups which are not-more-than-singly connected (w.s.). This result can be formalized by the statement that if the point $[n]$ is complete for time t,

$$\mathcal{E}([n];t)=\sum_{\substack{l\geq 1;\,i\geq 2\\ [i]+[c]=[n]+[l]}}\frac{1}{l!}\int_{V([i]|[c])}d([l])\times\sum_{\substack{[h]+[k]=[n]+[l]\\ 1\leq[h]\subset[i]}}\tau^{(n)}([h]|[k])\prod_{\alpha\in[k]}f_1(\alpha;t),\quad(5.9)$$

where the symbol $V([i]|[c])$ denotes the volume in l-particle phase space corresponding to all $(n+l)$-particle points which are tightly connected (w.s.) and have the group $[i]$ as their incomplete group. The restriction of the terms of the integrand to those for which the incomplete part of the integrand $[h]$ is contained in $[i]$, the incomplete group of the point, is justified by the first property of the $\tau^{(n)}$; if $[h]$ is larger than $[i]$ it must contain members of $[c]$ all of which have free legs (w.s.) for points which are tightly connected (w.s.).

Like Eq. (5.1) the expression for the error given by Eq. (5.9) is formally exact but only under the condition that, for given $[n]$, the time is so large that $[n]$ is complete, or alternatively that, for given t, one only considers points which are complete. It should be noted that the validity of this expression depends on the asymptotic forms of Liouville functions since these are assumed in deriving the properties of the $\tau^{(n)}$.

[49] This remark is equivalent to the one just made.

[50] In fact, $\tau^{(n)}([n_1]|[n_2])$ vanishes because it is equal to a sum of differences each of which vanishes.

VI. CONCLUDING REMARKS

It has been established that for $n\geq 2$, f_n can be exactly expressed as a functional power series in the one-particle probability density; indeed, according to Eqs. (1.1), (1.2), and (5.1) one has that

$$f([n];t)=\sum_{l\geq 0}\frac{1}{l!}\int d([l])\sum_{[h]+[k]=[n]+[l]}\tau^{(n)}([h]|[k])\times\prod_{\alpha\in[k]}f_1(\alpha;t),\quad(6.1)$$

where the coefficient operators $\tau^{(n)}$ are defined by the recursion relation, Eq. (5.5), as a sum of not-more-than-singly-connected products of the generalized scattering operators $\mathcal{S}_{m_1m_2}$. The group of terms for which $[h]$, the incomplete part of the integrands, is empty constitutes the asymptotic functional $f([n]|f_1)$, while the remaining terms express the error $\mathcal{E}([n];t)$. The terms of $f([n]|f_1)$ are analogs of the irreducible cluster integrals of equilibrium theory and their coefficient operators are sums of time-independent substitution operators. The coefficient operators in the error terms depend explicitly on the time and on the initial correlations [i.e., on $\xi_m(t=0)$].

This series is a formal identity since it follows with no approximation by using the decomposition of a Liouville function given by Eq. (5.2) to reexpress the original series for f_n given by Eq. (2.1).[51] Since, as has been pointed out elsewhere, the Liouville functions ξ_m are analogous to the activity while f_1 plays the role of the number density, this transformation may be considered to be the nonequilibrium analog of the transformation from activity to density. It should be mentioned one of us (R.P.) has shown that when f_1 is Maxwellian, $f([n]|f_1)$ is, exactly to all orders, the usual density series for the equilibrium f_n.

It has been established that points having groups which are not-more-than-singly-connected (w.s.) (i.e., as indicated by Fig. 8) make no contribution to either the terms of the asymptotic functional or the terms of the error. This means that the terms of the asymptotic functional contain no divergence due to infinite legs while the terms of the error neither increase with time because of growing legs nor diverge because of infinite legs. Because all more-than-singly-connected points except those which are tightly connected (w.s.) have groups which are not-more-than-singly-connected (w.s.), this result also implies that the *only* contributions to the error come from points which are not only *incomplete* but, in particular, are *tightly connected* (w.s.). This feature of the error is embodied in Eq. (5.9) which expresses $\mathcal{E}([n];t)$ for long times; that is, for times longer than the time for the point $[n]$ to become complete. We remark that the validity of these results which depend on various properties of the $\tau^{(n)}$ presupposes the validity of the reductions of Liouville func-

[51] This remark is validated in Appendix A.

tions and scattering operators and therefore they presuppose the assumptions stated in the Introduction.

It is significant that the only nonvanishing contributions to the error come from points which are incomplete and tightly connected (w.s.). We interpret this to mean that the asymptotic functional *correctly accounts for all other dynamical events*; that is, it not only accounts for all events which are uncorrelated initially (i.e., complete points) which are an obvious analog of Boltzmann's "chaotic" events but it also accounts for all events where the initial correlations only cause particles to divide into singly connected groups [i.e., points having groups which are not-more-than-singly-connected (w.s.)]. Thus, some types of initial correlation *are* taken into account by the time-dependent functional.

A. Properties of the Integrals

Having achieved these results it must still be established that the integrals in Eq. (6.1) converge at arbitrary times and that the error decreases with increasing time. Clearly, unless both of these features are present either the series is purely formal or $f([n]|f_1)$ is not the asymptotic form or both. We intend to discuss these points in detail in another place where we will give explicit estimates of the error terms. For the moment, we confine ourselves to a few remarks.

Preliminary estimates support the view that all the integrals do converge and that the contributions to the individual error integrals are proportional to inverse powers of the time for large times. It appears that both kinds of integrals have the same possible source of divergence and furthermore that the error integrals decrease with increasing time for the same reason that they converge. Thus, essentially the same investigation will establish both features of our results.

For example, we have considered the contribution to the terms, $\int d(3)\,\tau^{(2)}(0|123)f_1(1)f_1(2)f_1(3)$ and $\int d(3)\,\tau^{(2)}(13|2)f_1(2)$, due to all points which can be diagrammed as in Fig. 10 where it is understood that the $(1_{12}-3)$ aiming-to-collide occurs at the same time as or earlier than the $(2_{12}-3)$ collision. For all the points for which the $(1_{12}-3)$ aiming-to-collide occurs after the initial instant the first of these two integrals is certainly finite while the second one, a term in the two-particle error, simply vanishes. Thus, the convergence of both integrals (and, indeed, the whole value of the second one) is decided by the contributions from all points for which the $(1_{12}-3)$ aiming-to-collide occurs at or earlier than the initial instant. Included in these are those points for which the $(2_{12}-3)$ collision occurs arbitrarily early in time (compared to the initial instant) and such points might cause divergence of the integrals. Since the integrands of both integrals have similar behavior for such points, in this case convergence is indeed decided by essentially the same investigation for integrals of both kinds. We have verified that no divergence occurs for any reasonable behavior of f_1 in momentum space. In addition, we find that for large times the above error integral is proportional to $(t)^{-1}$; that is, $(t)^{-1}$ is a factor of the result which remains a functional of $f_1(t)$. The decrease with time of the error integral and the convergence of both of them are direct consequences of the manner in which the volume of the contributing points depends on the time of the $(2_{12}-3)$ collision. Qualitatively, for a fixed ratio of the time of the $(2_{12}-3)$ collision to the time of the $(1_{12}-3)$ aiming-to-collide, the volume decreases as the $(2_{12}-3)$ collision is shifted to earlier times because then more restrictive requirements are imposed on the momentum of particle 3 in order that the $(1_{12}-3)$ aiming-to-collide continues to occur.

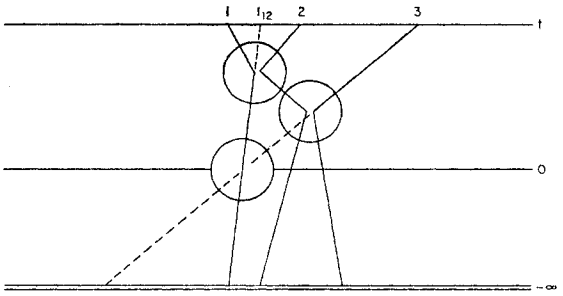

FIG. 10. A simple example of a tightly connected point; the incomplete group is {1,3}. Because of the $(1_{12}-3)$ aiming to collide, particle 3 is not singly connected in the wider sense.

There is no apparent reason to doubt that the features of this example are general. In particular, it is natural to expect that the additional restrictions (e.g., for aimings-to-collide and/or recollisions) on the phase of a particle (or group of them) which are required to make a point tightly connected (w.s.) will result in a decrease with time of the volume corresponding to such a point.

B. Consequences for the Boltzmann Equation

The expressions for f_n which have been derived are relevant to the question of the existence and form of a "generalized Boltzmann equation": that is, an equation for f_1 in which the time rate of change of f_1 due to collisions is approximated to higher orders in the density by time-independent functionals of f_1 only. A possible basis for such a study is provided by using the identity for f_2 obtained from Eq. (6.1) to express the integral term in the exact equation for f_1 (see Paper I). The resulting identity states that the total time derivative of f_1 is expressed as a series of "collision integrals" which are time-independent functionals of f_1 plus a sum of error integrals which depend on initial conditions. The binary collision integral is (except for gradient terms) the same as Boltzmann's while the ternary collision integral is simply

$$\int d(2)d(3)\ \mathbf{F}_{12}\cdot\nabla_{p_1}[S(123)-S(12)S(13)-S(12)S(23)+S(12)]f(1)f(2)f(3).$$

Presuming the validity of the remarks in Sec. VIA concerning the decrease with time of the error, it is reasonable to conjecture that the equation which results from this identity for f_1 by dropping all the error terms is the desired generalized Boltzmann equation and furthermore that the *exact* $f_1(t)$ approaches *some* solution of this approximate equation after long enough times. In view of the slow decrease with time of the error terms, however, the exact nature of this approach and, more generally, the whole relationship of the solutions of the approximate equation to the exact $f_1(t)$ for arbitrary initial conditions may be quite complex. It is perhaps useful to emphasize that the f_1 appearing in Eqs. (6.1) is the exact solution of the infinite hierarchy and *not* the solution of this proposed generalized Boltzmann equation.

A second conjecture is on firmer ground; namely, that the solutions of this generalized Boltzmann would provide an exact description of a stationary state simply because for such states all memory of the initial conditions (i.e., the error terms) must have vanished. Furthermore, this should also be true of the error terms in the expressions for the n-particle probability densities for $n\geqslant 2$. Thus, for a stationary state, f_n will be exactly equal to $f_n([n]\,|\,\tilde{f}_1)$ where $\tilde{f}_1$ denotes the stationary solution of the generalized Boltzmann equation. Thus, the generalized Boltzmann equation is completely adequate for the computation of transport coefficients.[52]

APPENDIX A: EXPRESSION FOR E_n AND RELATED RESULTS

The purpose here is to derive the expression for the error given by Eq. (2.4) and indicate the parallel argument for the difference between f_n and its functional series as given by Eq. (6.1).[53] It is shown that the decomposition of Liouville functions given by Eq. (5.2) implies Eq. (6.1). The expression for $\psi_l^{(n)}$ in terms of Liouville functions is also given.

To derive Eq. (2.4), first form the product of $f_1(\alpha)$, for $\alpha\in[n]+[l]$, by using the formal series, Eq. (2.1), for the case $n=1$. The result is that

$$\prod_{\alpha\in[n]+[l]} f_1(\alpha)=\sum_{k\geq 0}\frac{1}{k!}\int d([k]) \times\sum_{\Sigma_\alpha[k_\alpha]=[k]}\prod_{\alpha\in[n]+[l]}\psi^{(1)}(\alpha,[k_\alpha]), \quad \text{(A1)}$$

where the summation is over all partitions of $[k]$ into $(n+l)$ disjoint parts $[k_\alpha]$, some of which may be empty. Equation (A1) is derived from the direct product expression by first grouping terms with the same number of variables of integration and then making use of the dummy character of these variables.

If one uses this result to express the product of f_1's in the asymptotic functional, again collecting terms with the same number of integration variables and using the dummy character of these variables, one derives that

$$f([n]\,|\,f_1)=\sum_{l\geq 0}\frac{1}{l!}\int d([l])\sum_{[h]+\Sigma_\alpha[k_\alpha]=[l]}\tau^{(n)}([n];[h]) \times\prod_{\alpha\in[n]+[h]}\psi^{(1)}(\alpha,[k_\alpha]), \quad \text{(A2)}$$

where the summation is over all partitions of $[l]$ into $(n+h+1)$ disjoint parts $[h]$ and the $[k_\alpha]$, some of which may be empty.

Using Eq. (A2) for $f([n]\,|\,f_1)$ and Eq. (2.1) for f_n one has an expression for the error (referred to in the text) in terms of $\psi_l^{(n)}$ and $\psi_k^{(1)}$.

To derive Eq. (2.4) we need to express $\psi_l^{(n)}$ and $\psi_k^{(1)}$ in terms of Liouville functions. The solution of the defining recursion, Eq. (2.2) is, in fact, given by

$$\psi^{(n)}([n];[l])=\sum_{[q]+[r]=[l]}\xi([n]+[q])\vartheta_0([r]), \quad \text{(A3)}$$

where $\vartheta_0([r])$ is defined by Eq. (2.7). This expression is the obvious analog of the equilibrium statement; that is, one sums all distinct products of the ξ_m, the arguments of which partition $[n]+[l]$ into disjoint parts, one of which contains all of $[n]$.

Using the result, Eq. (A3), for the case $n=1$ to express the factors appearing in Eq. (A2), one has after some rearrangement of summations that

$$f([n]\,|\,f_1)=\sum_{l\geq 0}\frac{1}{l!}\int d([l])\sum_{\substack{[v]+[w]=[n]+[l]\\ [n]\subset[v]}}\tau^{(n)}([v]) \times\sum_{\Sigma_\alpha[q_\alpha]+[r]=[w]}\prod_{\alpha\in[v]}\xi(\alpha,[q_\alpha]) \times\sum_{\Sigma_\alpha[r_\alpha]=[r]}\prod_{\alpha\in[v]}\vartheta_0([r_\alpha]), \quad \text{(A4)}$$

where we have dropped the distinction between $[n]$ and the rest of the particles in writing $\tau^{(n)}$.

Now, it can be shown that

$$\sum_{\Sigma_\alpha[r_\alpha]=[r]}\prod_{\alpha\in[v]}\vartheta_0([r_\alpha])=\vartheta_{v-1}([r]) =\sum_{[r_1]+[r_2]=[r]}\vartheta_{v-2}([r_1])\vartheta_0([r_2]), \quad \text{(A5)}$$

where $\vartheta_\sigma([r])$ is defined by Eq. (2.7). Before proving these equalities let us derive their implications. Clearly, if we use the first equality in Eq. (A4) we verify that

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[52] The procedure would be a suitable generalization of the Chapman-Enskog one (see, for example, Refs. 1 and 2). A parallel approach to the transport coefficients proceeds to their expressions in terms of autocorrelation functions via master equations. Investigations in this line are exemplified in the work of L. Van Hove [Physica **23**, 441 (1957)], I. Prigogine [Physica **27**, 629 (1961)], R. Zwanzig [Phys. Rev. **124**, 983 (1961)], and J. Weinstock [Phys. Rev. **132**, 454 (1963)]. The detailed relationship between the two methods has not been given. It has been shown, however, that they yield the same results to lowest order in the density [see M. S. Green, J. Chem. Phys. **22**, 398 (1954)].

[53] These are equations in the text; equations in these Appendices are numbered separately.

the description of the terms in $f([n]|f_1)$ given in Sec. V.A is accurate. Then, using the second equality to evaluate Eq. (A4) for $f([n]|f_1)$, one finds after some further rearranging of the order of summations that

$$f([n]|f_1)=\sum_{l\geq 0}\frac{1}{l!}\int d([l])\sum_{[h]+[k]=[l]}\mathcal{g}_0([k])\times\{\sum_{\substack{[v]+[w]=[n]+[h]\\ [n]\subset[v]}}\tau^{(n)}([v])\sum_{[q_0]+\Sigma_\alpha[q_\alpha]=[w]}\prod_{\alpha\in[v]}\xi(\alpha,[q_\alpha])\mathcal{g}_{v-2}([q_0])\}. \quad \text{(A6)}$$

The expression for f_n in terms of Liouville functions is trivially obtained by substituting the expression for $\psi_l^{(n)}$ given by Eq. (A3) into Eq. (2.1); the result is that

$$f([n];l)=\sum_{l\geq 0}\frac{1}{l!}\int d([l])\times\sum_{[h]+[k]=[l]}\xi([n]+[h])\mathcal{g}_0([k]). \quad \text{(A7)}$$

It follows immediately from Eqs. (A6) and (A7) that the error can be expressed as in Eq. (2.4).

As a third result, notice that the steps leading from the original form of the asymptotic functional to Eq. (A6) can be repeated without any modification for the series of terms for f_n given by Eq. (6.1); the result, which corresponds to Eq. (A6), is that this series is equal to

$$\sum_{l\geq 0}\frac{1}{l!}\int d([l])\sum_{[h]+[k]=[l]}\mathcal{g}_0([k])\times\sum_{\substack{[u]+[v]+[w]=[n]+[h]\\ [n]\subset[u]+[v]}}\tau^{(n)}([u]|[v])\times\mathfrak{A}([v];[w]), \quad \text{(A8)}$$

where we have used the definition of $\mathfrak{A}([v];[w])$ given by Eq. (2.6). This result justifies our remarks in the text about the analogs of the factors $\mathfrak{D}_l^{(n)}$.

Finally, since substituting the decompositions of $\xi([n]+[h])$ given by Eq. (5.2) into Eq. (A7) yields that f_n is equal to the expression (A8) and since this latter expression was just shown to be equal to the functional series for f_n, we have proved the equivalence of the decomposition of Liouville functions and the functional series.

Returning to the proof of the equalities (A5) consider the terms of the left-hand side of the first equality. By definition of $\mathcal{g}_0$, these terms arise by partitioning each set $[r_\alpha]$ into, say, p_α nonempty parts $[r_{\alpha j}]$. Thus, the arguments of the ξ_m in each term form a partition of $[r]$ into, say, p *nonempty* parts $[\omega_i]$, so that this first sum may be re-expressed as the sum over all such partitions with a coefficient, say, c_{vp}. To compute the coefficient, note that for a fixed set of p_α's, there are $p!/\prod_\alpha p_\alpha!$ terms in the original sum which are identical to a term with a given partition of $[r]$ into p parts $[\omega_i]$. Furthermore, each of these terms has a coefficient $(-1)^{\Sigma_\alpha p_\alpha}\times\prod_\alpha p_\alpha!$. Therefore, c_{vp} is given by

$$c_{vp}=\sum_{\Sigma_\alpha p_\alpha=p}\frac{p!}{\prod_\alpha p_\alpha!}[(-1)^{\Sigma_\alpha p_\alpha}\prod_\alpha p_\alpha!], \quad \text{(A9)}$$

and this sum is easily evaluated by using a generating function. In fact, we find that c_{vp} is indeed equal to the coefficient appearing in the definition of $\mathcal{g}_{v-1}([r])$.

That the second equality also holds follows if $\mathcal{g}_\sigma$ obeys the recursion relation:

$$\mathcal{g}_{v-2}([r])=\sum_{[h]+[k]=[r]}\mathcal{g}_{v-1}([h])\xi([k]). \quad \text{(A10)}$$

For, using Eq. (A10) to eliminate $\mathcal{g}_{v-2}([r_1])$, one finds, after some rearrangement of summation, that the right-hand side of the second equality of Eqs. (A5) can be expressed by

$$\sum_{[h]+[k]=[r]}\mathcal{g}_{v-1}([h])\sum_{[k_1]+[k_2]=[k]}\xi([k_1])\mathcal{g}_0([k_2]).$$

This expression is, however, equal to $\mathcal{g}_{v-1}([r])$ because the sum which appears as a factor vanishes unless $[k_2]$ is empty. To see this, note that the arguments of the ξ_m of each term in the sum form a partition of $[k]$ into, say, p nonempty parts. But, a given such term arises in exactly one way with coefficient $(-1)^p p!$ from the terms where $[k_1]$ is empty and in p ways with coefficient $(-1)^{p-1}(p-1)!$ from terms where $[k_1]$ is one of the given sets. Hence, the coefficient of any such term is $(-1)^p p!+p(-1)^{p-1}(p-1)!$, which vanishes identically.[54] Hence, the second equality is established if Eq. (A10) is valid.

To establish this recursion relation note that again each term of the right-hand side is a product of the ξ_m, the arguments of which partition $[r]$ into, say, p nonempty sets, and that these are the terms in $\mathcal{g}_{v-2}([r])$. Furthermore, a given such term arises in exactly one way with coefficient $(-1)^p(v-1+p)!/(v-1)!$ and in p ways with the coefficient $(-1)^{p-1}(v-1+p-1)!/(v-1)!$. The total coefficient is, therefore, $(-1)^p(v-1+p)!/(v-1)!+p(-1)^{p-1}(v-2+p)!/(v-1)!$ which is just the coefficient appearing in $\mathcal{g}_{v-2}([r])$.

APPENDIX B: PROPERTIES OF LIOUVILLE FUNCTIONS AND SCATTERING OPERATORS

Because of the assumed finiteness of the range of force and the correlation length, strong heuristic proofs

[54] This result also established Eq. (A3).

of the reductions of Liouville functions and scattering operators can be given.

To understand the asymptotic form of, say, $\xi([m];t)$, for a complete point $[m]$, notice that when one expresses this function in terms of its initial value according to Eq. (2.3), the product condition allows the factorization of the initial value into one-particle functions. These can then be transformed to time t by using the inverse of Eq. (2.3) with the result that

$$\xi([m];t)=S_{-t}([m])\prod_{\alpha\in[m]}S_t(\alpha)\prod_{\alpha\in[m]}\xi_1(\alpha;t). \quad \text{(B1)}$$

Since the point $[m]$ is assumed to be complete so that the particles are dynamically independent prior to the initial instant, this expression is equivalent to that given in Eq. (3.3) where the operator has been replaced by its limiting value.

An argument for the validity of Eq. (3.5) for not-more-than-singly-connected points proceeds in similar fashion. By assumption the groups $\{\alpha,[s_\alpha]\}$ and $[d_\beta]$ are statistically and dynamically independent at the initial instant so that one can again express the function in terms of its initial value and apply the product condition in the obvious way. When each of the factors $\xi([m_\lambda];0)$ which appears is transformed back to time t by use of $S_t([m_\lambda])$, one sees that to establish Eq. (3.5) one must show that the operator

$$S_{-t}([c]+\sum_\alpha[s_\alpha]+\sum_\beta[d_\beta])\prod_{\alpha\in[c]}S_t(\alpha,[s_\alpha])\prod_\beta S_t([d_\beta])$$

is equivalent to $S([c])$. Since the groups $[d_\beta]$ are statistically and dynamically independent for the entire interval, application of Eq. (3.1) yields that the operator is equivalent to

$$S_{-t}([c]+\sum[s_\alpha])\prod_{\alpha\in[c]}S_t(\alpha,[s_\alpha]).$$

The final step follows by observing that, because of the third condition which is necessary in order that a point be not-more-than-singly-connected (see text), the image point of the particles, α and $[s_\alpha]$, t seconds earlier is the same whether it is computed according to the $(1+s_\alpha)$-particle history of the point $(\bar{\alpha},[s_\alpha])$, or as the projection onto the space of $\{\alpha,[s_\alpha]\}$ of the $(c+\sum s_\alpha)$-particle history of the point $([c]+\sum[s_\alpha])$; that is, in operating on members of the group $\{\alpha,[s_\alpha]\}$, $\mathcal{S}([c])\times S_{-t}(\alpha,[s_\alpha])$ is equivalent to $S_{-t}([c]+\sum[s_\alpha])$.

The derivation of the reduction for a tightly connected point leads in a natural way to the definition of the generalized scattering operator; having accomplished this, the argument just given for including the hooked on sets $[s_\alpha]$ can be used to derive the general result Eq. (3.7). For a tightly connected point, say, $[i]+[c]$, the complete group of which is $[c]$, one can again apply the product condition in the obvious way with the result that

$$\xi([i]+[c];t)=\xi([i]_{-t};0)S_{-t}([i]+[c])\times\prod_{\alpha\in[c]}S_t(\alpha)\prod_{\alpha\in[c]}\xi_1(\alpha;t), \quad \text{(B2)}$$

where the point $[i]_{-t}$ is defined to be equivalent to $S_{-t}([i]+[c])[i]$. But, since for a tightly connected point every member of $[c]$ has a free leg, when operating on members of $[c]$, the time-dependent operator for the second factor in Eq. (B2) is equivalent to $\mathcal{S}([i]+[c])$ as defined by Eq. (3.4). Thus, for tightly connected points one arrives at the case of Eq. (3.7) for which the sets $[s_\alpha]$ are empty.

Because it is needed to prove the properties of the $\tau^{(n)}$ and because it illustrates the argument which proves the general result Eq. (3.9), we want to derive the reduction of $\mathcal{S}([m_1]|[m_2]$ for the point the history of which is diagrammed in Fig. 8. Since $[m_1]$ and $[m_2]$ partition $[r]+\lambda+[s]$, $[m_j]$, say, contains $[r_j]$, the part of it in $[r]$, and $[s_j]$, the part of it in $[s]$. Assuming first that the particle λ is a member of $[m_1]$, and applying the product condition to $\xi([r_1]+\lambda+s_1]_{-t};0)$, one has that

$$S([m_1]|[m_2])=[S_{-t}([r]+\lambda+[s])\xi([r_1];0)]\times[S_{-t}([r]+\lambda+[s])\xi(\lambda,[s_1];0)]\times S([r]+\lambda+[s]). \quad \text{(B3)}$$

The group $[s]$, however, plays no role in determining $[r_1]_{-t}$, so that this point can be computed using $S_{-t}([r]+\lambda)$. Furthermore, because $[s]$ is singly connected, when operating on the group, $\{\lambda,[s_1]\}$, $S_{-t}([r]+\lambda+[s])$ is equivalent to $\mathcal{S}([r]+\lambda)S_{-t}(\lambda,[s])$, and also $\mathcal{S}([r]+\lambda+[s])$ is equivalent to $\mathcal{S}([r]+\lambda)\times\mathcal{S}(\lambda[s])$. Using these three results to re-express Eq. (B3), one finds directly that

$$\mathcal{S}([m_1]|[m_2])=\mathcal{S}([r_1]|[r_2]+\lambda)S(\lambda,[s_1]|[s_2]). \quad \text{(B4a)}$$

In the other case when particle λ is in the group $[m_2]$, one finds in a similar way that

$$\mathcal{S}([m_1]|[m_2])=\mathcal{S}([r_1]|[r_2]+\lambda)\mathcal{S}([s_1]|\lambda,[s_2]). \quad \text{(B4b)}$$

APPENDIX C: COEFFICIENT OPERATORS OF $f([n]|f_l)$

Here we give the coefficient of the terms in the solution of Eq. (4.2) which defines $\tau_l^{(n)}$. Explicit expressions for the first few of the $\tau_l^{(2)}$ are also given.

In general, any term in $\tau^{(n)}$ can be divided into a number of disjoint parts which are singly connected within themselves. Suppose a term has σ such parts and a total of, say, μ, $\mathcal{S}$ operators. Also, let the total number of particles which appear *explicitly* be $(n+l_1)$ and, finally, let the total number of particles appearing in that singly connected part which contains the first $\mathcal{S}$ operator be $(n+l_0)$. The coefficient of this term is $(n+l_0-1)(n+l-2)!(-1)^{\mu-\sigma}/(n+l_1-\sigma)!$.

Denoting the operator $\mathcal{S}([m])$ simply by the symbol

($[m]$), the first few of the $\tau_l^{(2)}$ in terms of the $\mathcal{S}_m$ are given by[55]

$$\tau_0^{(2)}(12)=(12), \tag{C1}$$

$$\tau_1^{(2)}(12;3)=(123)-(12)(13)-(12)(23)+(12), \tag{C2}$$

and

$$\begin{aligned}\tau_2^{(2)}(12;34)&=(1234)-(12)(234)-(12)(134)+(12)(13)(14)\\&\quad+(12)(13)(34)+(12)(14)(13)+(12)(14)(34)\\&\quad+(12)(23)(14)+(12)(23)(24)+(12)(23)(34)\\&\quad+(12)(24)(13)+(12)(24)(23)+(12)(24)(34)\\&\quad-2(12)(13)-2(12)(14)-2(12)(23)-2(12)(24)\\&\quad-(12)(34)-(123)(14)-(123)(24)-(123)(34)\\&\quad-(124)(13)-(124)(23)-(124)(34)+2(123)\\&\quad+2(124)+2(12).\end{aligned} \tag{C3}$$

APPENDIX D: RECURSION RELATION FOR $\tau^{(n)}$

We will establish that for a tightly connected point ($[i]+[c]$), the decomposition, Eq. (5.2), reduces to the recursion relation, Eq. (5.5), if one assumes that $\tau^{(n)}$ vanishes when any particles in its incomplete part have any free legs (w.s.). The result of assuming this property of the $\tau^{(n)}$ and using the reduction, Eq. (5.4), for the point, ($[i]+[c]$), itself is expressed by stating that

$$\begin{aligned}S([i]|[c])\prod_{\alpha\in[c]}\xi_1(\alpha;t)&=\sum\tau^{(n)}([u]|[v_1]+[v_2])\\&\quad\times\mathfrak{A}([v_1]+[v_2];[w_1]+[w_2]),\end{aligned} \tag{D1}$$

where the summation is such that: $[u]+[v_1]+[w_1]=[i]$, $[v_2]+[w_2]=[c]$, $[u]\subset[u]+[v_1]+[v_2]$, and where in the nonvanishing terms of Eq. (5.2) we have split $[v]$ into disjoint parts, $[v_j]$, $[v_1]$ being the part in $[i]$ and similarly for $[w]$. Note that since $[n]\subset[u]+[v_1]+[v_2]$, the groups $[w_j]$ partition some subset of $[l]$. Recalling the definition of $\mathfrak{A}([v];[w])$, the general term in this sum has the form

$$\tau^{(n)}([u]|[v_1]+[v_2])\prod_{\alpha\in[v_1]+[v_2]}\xi(\alpha,[q_\alpha])\prod_\beta\xi([r_\beta]),$$

where the $[q_\alpha]$ and $[r_\beta]$ together form a partition of $[w_1]+[w_2]$. For a point which is tightly connected (w.s.), however, the argument points of every Liouville function of such a term reduces according to Eq. (5.4) so that it is equivalent to

$$\tau^{(n)}([u]|[v_1]+[v_2])\prod_{\alpha\in[v_1]}\mathcal{S}(\alpha,[q_{1\alpha}]|[q_{2\alpha}])\prod_{\alpha\in[v_2]}\mathcal{S}([q_{1\alpha}]|\alpha,[q_{2\alpha}])\prod_\beta\mathcal{S}([r_{1\beta}]/[r_{2\beta}])\prod_{\lambda\in[c]}\xi_1(\alpha;t).$$

[55] We note again that operators with indices in common do not commute.

The group $[q_{j\alpha}]$ is the part of $[q_\alpha]$ contained in $[w_j]$ and similarly for $[r_{j\beta}]$. The product of ξ_1's runs over the group $[c]$ because

$$[v_2]+\sum_{\alpha\in[v_1]+[v_2]}[q_{2\alpha}]+\sum_\beta[r_{2\beta}]=[v_2]+[w_2]=[c].$$

Since this reduction is valid for every term of Eq. (D1), Eq. (5.5) follows with the A sum defined by

$$\begin{aligned}A([v_1];[w_1]|[v_2];[w_2])&=\sum(-1)^p\frac{(v_1+v_2-2+p)!}{(v_1+v_2-2)!}\prod_{\alpha\in[v_1]}\mathcal{S}(\alpha,[q_{1\alpha}]|[q_{2\alpha}])\\&\quad\times\prod_{\alpha\in[v_2]}\mathcal{S}([q_{1\alpha}]|\alpha,[q_{2\alpha}])\prod_\beta\mathcal{S}([r_{1\beta}]|[r_{2\beta}]).\end{aligned} \tag{D2}$$

The sum here is over all disjoint sets for which $[w_j]$ is partitioned into (v_1+v_2) sets, $[q_{j\alpha}]$, some of which may be empty, and p nonempty sets, $[r_{j\beta}]$. In summing on the $[r_{j\beta}]$, permutations are not counted as distinct terms so that the sum over sets with a given index j is the same as in $A([v_1]+[v_2];[w_j]$.

APPENDIX E: PROPERTIES OF THE $\tau^{(n)}$

We have found inductive proofs of the properties of the $\tau^{(n)}$ stated in Secs. IV and V. Actually, we prove equivalent statements; namely, that

(a) If the point $(\lambda,[r])$ is arbitrary except that particle λ has a free leg (w.s.), then $\tau^{(n)}(\lambda[r_1]|[r_2])$ vanishes for any sets $[r_j]$ which partition $[r]$;

(b) If the point $(\lambda,[r],[s])$, where $[n]\subset\{\lambda,[r]\}$, is arbitrary except that the group $[s]$ is not-more-than-singly-connected (w.s.) to $[r]$ through particle λ, then $\tau^{(n)}([r_1]+\lambda_1+[s_1]|[r_2]+\lambda_2+[s_2])$ vanishes for any sets $[r_j]$ which partition $[r]$, any sets $[s_j]$ which partition $[s]$, and for either the case for which $\lambda_1=0$ and $\lambda_2=\lambda$ or the opposite one. (See Fig. 8.) It is our intention to publish the details of the proofs of these statements in another place[56] but the following description may be helpful.

Consider proving the second property. It is easy to verify that it holds for the case of $\tau^{(n)}(0|[n]+l)$ for which there is only one particle l in the group of additional particles $[l]$. Moreover, the recursion relation, Eq. (5.5), for $\mathcal{S}([c_1]|[c_2])$ can be written as an expression for $\tau^{(n)}([c_1]|[c_2])$ by using the fact that $A(0;0|0;v_2)\equiv1$. In this equation one notices that the index pairs (u,v), which denote the "size" of the set pairs $([u]|[v])$, which appear as the arguments of the $\tau_{u,v}^{(n)}$, are all such that $n\leqslant u+v<c_1+c_2$ with $u\leqslant c_1$ and either $u\neq c_1$ or $v\neq c_2$ (but not both); one can say that the index pairs which appear are all "less than" the pair (c_1,c_2). These remarks suggest that one can make a proof by proving that: If the property holds for all the $\tau_{u,v}^{(n)}$ for which (u,v) is less than (c_1,c_2), then it

[56] That is, in the J. Res. Natl. Bur. Std.

holds for $\tau_{c_1c_2}{}^{(n)}$. Recalling that the property is vacuous if there are no additional particles $[l]$ (so that then one can define the property to hold), it is easy to verify explicitly that this implication does yield an induction.

To prove this implication for the case where $[c_j]=[r_j]+\lambda_j+[s_j]$, first evaluate the expression for $\tau^{(n)}([r_1]+\lambda_1+[s_1]|[r_2]+\lambda_2+[s_2])$ [from Eq. (5.5)] assuming the first property (to be proven independently) and the hypothesis of the implication; the result is that

$$\begin{aligned}\tau^{(n)}&([r_1]+\lambda_1+[s_1]|[r_2]+\lambda_2+[s_2])\\ &=\mathcal{S}([r_1]+\lambda_1+[s_1]|[r_2]+\lambda_2+[s_2])\\ &\quad-\sum\tau^{(n)}([u']|[v_1']+[v_1'']+[v_2])\\ &\quad\times A([v_1']+[v_1''];[w_1']+[w_1'']\\ &\qquad+[s_1]|[v_2];[w_2]+[s_2]),\end{aligned}\tag{E1}$$

where the summation is such that: $[u']+[v_1']+[w_1']=[r_1]$, $[v_1'']+[w_1'']=\lambda_1$, $[v_2]+[w_2]=[r_2]+\lambda_2$, $[n]\subset[u']+[v_1']+[v_1'']+[v_2]$. The fact that no $\tau^{(n)}\times([u]|[v])$ appears such that $\lambda_1\subset[u]$ is a consequence of the first property [λ_1 should be treated as having a free leg (w.s.)], while the nonoccurrence of cases where members of $[s]$ are in $[u]$ or $[v]$ is a consequence of the second.[57] Further evaluation of the right-hand side of this equation is made by using the fact that the group $[s]$ is not-more-than-singly-connected (w.s.) so that the result for $\mathcal{S}([m_1]|[m_2])$ given in Appendix B applies to the first term and also to every factor of the A sum of every other term; we have shown that this implies (this is the nontrivial step) that, for every term in Eq. (E1),

$$\begin{aligned}\tau^{(n)}([u']|[v_1']+[v_1'']+[v_2])A([v_1']+[v_1''];[w_1']+[w_1'']+[s_1]|[v_2];[w_2]+[s_2])\\ =\tau^{(n)}([u']|[v_1']+[v_1'']+[v_2])A([v_1'];[w_1']|[v_2]+[v_1''];[w_2]+[w_1''])\mathcal{S}(\lambda_1[s_1]|\lambda_2[s_2]).\end{aligned}\tag{E2}$$

If these results are used to evaluate the terms of Eq. (E1) one finds that, indeed, $\tau^{(n)}([r_1]+\lambda_1+[s_1]|[r_2]+\lambda_2+[s_2])$ vanishes. To prove Eq. (E2) one first verifies that any term of the left-hand side does reduce to one of those appearing on the right-hand side; then, a combinatorial argument (similar to the ones in Appendix A) establishes the coefficient.

The first property is proved by essentially the same argument. By direct verification it holds for $\tau^{(n)}(\lambda|[r])$ where $\{\lambda,[r]\}=[n]$ and the same implication would establish it for $\tau^{(n)}(\lambda+[r_1]|[r_2])$. Assuming the hypothesis of the implication yields an expression for $\tau^{(n)}(\lambda+[r_1]|(r_2])$ which is, in fact, the case of Eq. (E1) for which λ_1 is particle λ and the sets $[s_j]$ are empty. Furthermore, because particle λ is a free leg (w.s.), $\mathcal{S}(\lambda[r_1]|[r_2])$ and all the factors $\mathcal{S}_{m_1m_2}$ in this result have reductions and this implies the validity of the analog of Eq. (E2). The desired result then follows by algebra.

[57] In Eq. (E1) we have assumed that particle λ is not a member of $[n]$ (then $[w_1'']$ could not be equal to λ_1). If $\lambda c[n]$, take $[w_1'']$ to be empty to obtain the correct expression.