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[1]E. Hinnov, J. E. Hesser, and L. C. Johnson, Princeton Plasma Physics Laboratory MATT-462, 1967 (unpublished).

[2]These rates need not be accurately known. We have extracted them from calculations of corresponding transitions in Ca^+ by O. Bely, Phys. Letters 23, 442 (1966), and from calculations of Petrini, Ref. 9.

[3]L. C. Johnson, T. K. Chu, H. Hendel, and E. Hinnov, Bull. Am. Phys. Soc. 13, 1557 (1968).

[4]Yu. I. Ostrovskii and N. P. Penkin, Opt. i. Spektroskopiya 10, 8 (1961). [English transl.: Opt. Spectry. (USSR) 10, 3 (1961)]; Opt. i Spektroskopiya 11, 565 (1961) [English transl.: Opt. Spectry. (USSR) 11, 307 (1961)].

[5]A. Gallagher, Phys. Rev. 157, 24 (1967).

[6]R. H. Garstang and S. J. Hill, Publ. Astron. Soc. Pacific 78, 70 (1966).

[7]M. J. Seaton, in Atomic and Molecular Processes, edited by D. R. Bates (Academic Press Inc., New York, 1962); H. van Regemorter, Astrophys. J. 136, 906 (1963).

[8]E. Hinnov *et al.*, Phys. Fluids 6, 1779 (1963); N. Rynn *et al.*, Rev. Sci. Instr. 38, 1378 (1967).

[9]D. Petrini, Compt. Rend. Acad. Sci. (France) 260, 4929 (1965).

[10]F. M. Bacon and J. W. Hooper, Phys. Rev. 178, 182 (1969).

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Convergent Generalization of the Boltzmann Equation for a Hard-Sphere Lorentz Gas

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A generalization of the Boltzmann equation for a classical Lorentz gas with hard-core interaction is presented. The N-body streaming operator is evaluated directly from the dynamics, thereby avoiding the binary collision expansion. A cluster expansion is developed in a form that results in exponential decay of the dynamical correlations and regularizes all divergent diagrams. Virtual collisions, represented by virtual binary kernels, are related to configuration-space restrictions, which in turn are responsible for the collisional damping. A prescription is given for the convergent l-body collision integral.

I. INTRODUCTION

An outstanding problem in the kinetic theory of dense gases is the derivation of higher density corrections to the Boltzmann equation[1-3] and to the transport coefficients.[4-8] A related problem is the generalization of the Boltzmann equation for the Lorentz gas,[9] a system composed of a large number of fixed scattering centers and a small number of noninteracting scattered particles.

The Lorentz gas provides a useful model for the study of the kinetic theory of dense gases. It contains many of the features of dense-gas theory, yet it is more tractable due to the linearity of the multiple collision integrals in the particle distribution function. Hauge and Cohen[10] studied a special case of the Lorentz gas known as the Ehrenfest wind-tree model. They obtained all contributions to first order in the density beyond the Boltzmann term for the self-diffusion coefficient. Van Leeuwen and Weijland[11] evaluated the self-diffusion coefficient for a two- and a three-dimensional hard-sphere Lorentz gas obtaining the first logarithmic density-dependent terms. This paper discusses a divergent-free generalization[12] of the Boltzmann equation for a Lorentz gas with classical hard-core interaction.[13]

The appropriate variable describing the Lorentz gas is the particle distribution function

$$f(\vec{r},\vec{p};t)=\int\cdots\int d\vec{\xi}_1\cdots d\vec{\xi}_N D(1,\ldots,N;t)\,. \quad (1.1)$$

It is the integral over all scattering center positions $\vec{\xi}_1,\ldots,\vec{\xi}_N$, of the joint particle-scattering center distribution function, $D(1,\ldots,N;t)$. We assume that D obeys the Liouville equation and has the initial value

$$D(1,\ldots,N;0)=Z_N^{-1}W(1,\ldots,N)f(\vec{r},\vec{p};0)\,, \quad (1.2)$$

$$Z_N=\int\cdots\int d\vec{\xi}_1\cdots d\vec{\xi}_N W(1,\ldots,N)\,, \quad (1.3)$$

$$W(1,\ldots,N)=e^{-\beta\Phi}\,, \quad (1.4)$$

where Φ is the total potential energy of the scattering centers and the scattered particle.

The initial value of D [Eq. (1.2)] allows a random distribution of the scattering centers consis-

tent with the exclusion of any overlap of the hard cores. The function $W(1,\ldots,N)$, is unity when none of the hard cores are overlapping, and is zero otherwise.

We describe briefly the ϵ method[14] for deriving an equation satisfied by the particle distribution function. Using the initial-value solution of the Liouville equation and the initial-value assumption on D, we may write the Laplace transform of the particle distribution function $f_\epsilon(\vec{r},\vec{p})$ as

$$f_\epsilon(\vec{r},\vec{p}) = \Gamma_\epsilon(\vec{r},\vec{p}) f(\vec{r},\vec{p};0)\,, \tag{1.5}$$

$$\Gamma_\epsilon(\vec{r},\vec{p}) = \int\cdots\int d\vec{\xi}_1\cdots d\vec{\xi}_N S_\epsilon(1,\ldots,N) Z_N^{-1} W(1,\ldots,N), \tag{1.6}$$

where $S_\epsilon(1,\ldots,N)$ is the Laplace transform of the time displacement operator, or the streaming operator.[15] The equation satisfied by the particle distribution function is

$$\epsilon f_\epsilon(\vec{r},\vec{p}) - f(\vec{r},\vec{p};0) + \vec{v}\cdot(\partial/\partial\vec{r}) f_\epsilon(\vec{r},\vec{p}) = K_\epsilon(\vec{r},\vec{p}) f_\epsilon(\vec{r},\vec{p}), \tag{1.7}$$

$$K_\epsilon(\vec{r},\vec{p}) = S_\epsilon^{-1}(\Gamma_\epsilon - S_\epsilon)\Gamma_\epsilon^{-1}\,, \tag{1.8}$$

$$S_\epsilon = [\epsilon + \vec{v}\cdot(\partial/\partial\vec{r})]^{-1}\,. \tag{1.9}$$

The requirement that $f_\epsilon(\vec{r},\vec{p})$ obeys Eq. (1.7) for arbitrary initial-particle distribution function, results in the expression for $K_\epsilon(\vec{r},\vec{p})$, Eq. (1.8).[16] The generalized collision operator $K_\epsilon(\vec{r},\vec{p})$ describes the change in $f_\epsilon(\vec{r},\vec{p})$ due to all interactions with the scattering centers.

The remainder of this paper is concerned with the evaluation of the generalized collision operator. In Sec. II, the (Laplace transformed) streaming operator is evaluated directly from the hard-sphere dynamics. The successive interactions of the particle with the fixed scattering centers are represented by products of binary kernels (binary collision operators).[17] In Sec. III, the operator $\Gamma_\epsilon(\vec{r},\vec{p})$, which determines the time evolution of $f(\vec{r},\vec{p};t)$, is expressed as a cluster expansion. We find that the effect of multiple collisions give rise to a collisional damping such that the dynamical correlations decay exponentially for large distances. In Sec. IV, we derive a convergent generalization of the Boltzmann collision integral for a hard-sphere Lorentz gas.

II. EVALUATION OF STREAMING OPERATOR

The streaming operator is evaluated directly from the hard-sphere dynamics. The procedure we discuss may also be applied to dense gases and need not be restricted to hard-core potentials. For a given momentum of the scattered particle, we decompose the configuration space into regions in which a collision with a scattering center may or may not take place. Since the interaction is strictly pair wise, it suffices to decompose the relative position of the particle with a single scattering center, $\vec{r}_j = \vec{r} - \vec{\xi}_j$. The entire space of $\vec{r}_j$, denoted by l_j, consists of the regions

$A_j = \theta(\sigma - b_j)\theta(\tau_j^-)$: the particle aims to collide with scattering center j in the past; (2.1)

$Z_j = \theta(\sigma - b_j)\theta(-\tau_j^+)$: the particle aims to collide with scattering center j in the future; (2.2)

$I_j = \theta(\sigma - b_j)\theta(-\tau_j^-)\theta(\tau_j^+)$: the particle overlaps scattering center j; (2.3)

$N_j = \theta(b_j - \sigma)$: the particle does not aim to collide with scattering center j. (2.4)

The unit-step function is defined by $\theta(x) = 1$ for $x > 0$ and $\theta(x) = 0$ for $x \leq 0$. The collision parameters are the hard-core radius σ, the impact parameter b_j, and the time the particle moves freely from the initial position to the hard-core boundary

$$\tau_j^\pm = [\vec{r}_j\cdot\hat{v} \mp (\sigma^2 - b_j^2)^{1/2}]\, v^{-1}\,. \tag{2.5}$$

The space of $\vec{r}_j$ is the sum of the four regions

$$l_j = A_j + Z_j + I_j + N_j\,,$$

modulo a set of measure zero, since the regions exclude the boundaries $|\vec{r}_j| = \sigma$ and $b_j = \sigma$. The entire configuration space of N scattering centers is expressed as the product

$$\prod_{j=1}^{N} (A_j + Z_j + I_j + N_j)\,.$$

The streaming operator is evaluated for all initial nonoverlapping configurations

$$W(1,\ldots,N) S_\epsilon(1,\ldots,N) = \prod_{j=1}^{N} (A_j + Z_j + N_j) S_\epsilon(1,\ldots,N)\,. \tag{2.6}$$

We may commute $W(1,\ldots,N)$ and $S_\epsilon(1,\ldots,N)$ in the expression for $\Gamma_\epsilon(\vec{r},\vec{p})$ [Eq. (1.6)], since initial nonoverlapping configurations transform into final nonoverlapping configurations and vice versa.[17] We first decompose the configuration space into the region where no interactions take place and the region where at least one interaction can take place

$$W(1,\ldots,N)S_\epsilon(1,\ldots,N) = \prod_{j=1}^{N}(Z_j+N_j)S_\epsilon + \sum_{j=1}^{N}\prod_{k\neq j}[A_k\theta(\tau_k^- - \tau_j^-)+Z_k+N_k] \times A_j S_\epsilon(1,\ldots,N). \quad (2.7)$$

The first term in Eq. (2.7) contains the condition that the particle either aims to collide with a scattering center in the future or does not aim to collide. Since $S_\epsilon(1,\ldots,N)$ streams the particle backwards in time, it reduces to the free streaming operator $S_\epsilon=[\epsilon+\vec{v}\circ(\partial/\partial\vec{r})]^{-1}$. In the second term, the particle aims to collide with scattering center j (in the past) before encountering any of the other scattering centers. For times t less than τ_j^-, the particle does not reach the hard-core boundary and $S_\epsilon(1,\ldots,N)$ generates only free streaming

$$S_\epsilon(1,\ldots,N) \rightarrow \int_0^{\tau_j^-} dt\, e^{-\epsilon t} S(t). \quad (2.8a)$$

For times t greater than τ_j^-, the particle reaches the hard-core boundary, undergoes a real collision, and continues streaming (with interaction) along $-\mathcal{R}_j\vec{v}$

$$S_\epsilon(1,\ldots,N) \rightarrow \int_{\tau_j^-}^{\infty} dt\, e^{-\epsilon t} S(\tau_j^-)\mathcal{R}_j S(1,\ldots,N;t-\tau_j^-). \quad (2.8b)$$

The free-streaming operator is denoted by $S(t) = \exp(-t\vec{v}\cdot\partial/\partial\vec{r})$, and the momentum rotation operator is denoted by

$$\mathcal{R}_j\vec{v} = \vec{v} - 2\vec{v}\cdot\hat{\sigma}\hat{\sigma}, \quad (2.9)$$

$$\vec{\sigma} = \vec{r}_j - \vec{r}_j\cdot\hat{v}\hat{v} + (\sigma^2 - b_j^2)^{1/2}\hat{v}. \quad (2.10)$$

The free streaming in expression (2.8a) over the time interval $0<t<\tau_j^-$ can be represented as free streaming for an infinite time minus free streaming for time $t>\tau_j^-$

$$\int_0^{\tau_j^-} dt\, e^{-\epsilon t} S(t) = S_\epsilon - \int_{\tau_j^-}^{\infty} dt\, e^{-\epsilon t} S(\tau_j^-)S(t-\tau_j^-). \quad (2.11)$$

We introduce the binary kernel representation. We have shown that the hard-sphere dynamics can be decomposed into three elements: uninterrupted free streaming, represented by S_ϵ; free streaming to the hard-core boundary followed by a rotation of the particle momentum, represented by $S(\tau_j^-)\mathcal{R}_j$; and free streaming to the hard-core boundary with no rotation, represented by $S(\tau_j^-)(-)$. The latter two dynamical situations can be represented with binary kernels

$$S_\epsilon B^r(j) = A_j e^{-\epsilon\tau_j^-} S(\tau_j^-)\mathcal{R}_j S_\epsilon, \quad (2.12)$$

$$S_\epsilon B^v(j) = A_j e^{-\epsilon\tau_j^-} S(\tau_j^-)(-)S_\epsilon. \quad (2.13)$$

In the real collision kernel $S_\epsilon B^r(j)$, A_j ensures that the particle aims to collide in the past, $S(\tau_j^-)$ generates free streaming to the hard-core boundary, $\mathcal{R}_j$ rotates the particle momentum, and S_ϵ generates further free streaming. In the virtual kernel $S_\epsilon B^v(j)$, the rotation operator is replaced by (-1). The virtual (or noninteracting) binary kernel is introduced to simplify the representation of those regions of configuration space where no collision occurs. In the example of Eq. (2.11), which may be written

$$\int_0^{\tau_j^-} dt\, e^{-\epsilon t} S(t) = S_\epsilon + S_\epsilon B^v(j), \quad (2.14)$$

the virtual kernel is employed when the particle aims to collide with a scattering center but does not stream freely for a time long enough to reach it. A second use of the virtual binary kernel is to represent the dynamical situation where a collection of scattering centers is prevented from interrupting the particle's free streaming to the hard-core boundary of scattering center j.

We demonstrate the latter use of the virtual binary kernel by transforming the configuration space restrictions in Eq. (2.7) into a sum of products of virtual collision operators. These conditions (of not interrupting the particle motion) may be rewritten

$$\prod_{k\neq j}[A_k\theta(\tau_k^- - \tau_j^-)+Z_k+N_k]A_j = W(1,\ldots,N)\prod_{k\neq j}[1-A_k\theta(\tau_j^- - \tau_k^-)]A_j. \quad (2.15)$$

The right-hand side of Eq. (2.15) is the product of the factors: One minus the condition that a scattering center interrupts the particle motion. The function $W(1,\ldots,N)$ ensures the exclusion of hard-core overlap. The conditions of interrupting the particle motion can be represented with the virtual binary kernel as in Eq. (2.14). Let the sum of products of virtual kernels be denoted by

$$\sigma(1,\ldots,j-1,j+1,\ldots,N)=\Big(1-\sum_{k\neq j}B^{v}(k)\Big)^{-1}. \tag{2.16}$$

The configuration space restrictions in Eq. (2.15) can be written

$$W(1,\ldots,N)\prod_{k\neq j}\Big(1-A_k\theta(\tau_j^- -\tau_k^-)\Big)\times A_j e^{-\epsilon\tau_j^-}S(\tau_j^-)=W(1,\ldots,N)\times S_\epsilon\,\sigma(1,\ldots,j-1,j+1,\ldots,N)A_j e^{-\epsilon\tau_j^-}S(\tau_j^-). \tag{2.17}$$

Equation (2.17) is verified by expanding the sum of products in Eq. (2.16) and applying the streaming operator in $B^v(k)$ to all $\vec{r}$-dependent terms to its right-hand side: $S(\tau)F(\vec{r})=F(\vec{r}-\vec{v}\,\tau)$. Equations (2.14), (2.15), and (2.17) demonstrate that all configuration space restrictions in the streaming operator, except the restriction of no overlap, can be represented with virtual binary kernels.

As a result of the above manipulations, the N-scatterer streaming operator can be written

$$W(1,\ldots,N)S_\epsilon(1,\ldots,N)=W(1,\ldots,N)\times\Big(S_\epsilon+S_\epsilon\sum_{j=1}^{N}\sigma(1,\ldots,j-1,j+1,\ldots,N)\times[B^v(j)+B^r(j)S_\epsilon{}^{-1}W(1,\ldots,N)S_\epsilon(1,\ldots,N)]\Big). \tag{2.18}$$

If we solve this recursion relation for $W(1,\ldots,N)\times S_\epsilon(1,\ldots,N)$ and make use of the nilpotent property of the binary kernels,[18] then we obtain the representation

$$W(1,\ldots,N)S_\epsilon(1,\ldots,N)=W(1,\ldots,N)S_\epsilon\,\sigma\Big(1-\sum_{j=1}^{N}B^r(j)\sigma\Big)^{-1}. \tag{2.19}$$

The operator σ without labels denotes $\sigma(1,\ldots,N)$. The function $W(1,\ldots,N)$ is redundant when placed to the right of the operator $B^r(j)S_\epsilon{}^{-1}$, since the conditions on its left-hand side exclude hard-core overlap. The streaming operator [Eq. (2.19)] generates sequences of real collisions, as represented by the real binary kernels. Each real collision is preceded and followed by restrictions on the scattering centers which prevent interruption of the particle motion, as represented by the sum of products of virtual binary kernels in the σ operators.

The equivalence of the above representation of $W(1,\ldots,N)S_\epsilon(1,\ldots,N)$ with the binary collision expansion[19] is readily demonstrated:

$$W(1,\ldots,N)S_\epsilon(1,\ldots,N)=W(1,\ldots,N)S_\epsilon\Big(\sigma^{-1}-\sum_{j=1}^{N}B^r(j)\Big)^{-1}=W(1,\ldots,N)S_\epsilon\Big(1-\sum_{j=1}^{N}[B^r(j)+B^v(j)]\Big)^{-1}. \tag{2.20}$$

Using the nilpotent property of the binary kernels, the expansion of Eq. (2.20) in products of binary collision operators $B(j)=B^r(j)+B^v(j)$ reduces to the sum of all products with distinct adjacent labels, i.e., the binary collision expansion. The streaming operator can also be written in pseudopotential form[17]

$$W(1,\ldots,N)S_\epsilon(1,\ldots,N)=W(1,\ldots,N)\Big(\epsilon+\vec{v}\cdot\frac{\partial}{\partial\vec{r}}-\sum_{j=1}^{N}B(j)S_\epsilon{}^{-1}\Big)^{-1}, \tag{2.21}$$

$$B(j)S_\epsilon{}^{-1}=\theta(\sigma-b_j)\delta(\tau_j^-)\{\mathcal{R}_j-1\}, \tag{2.22}$$

where δ is the Dirac δ function.

The method presented here of evaluating the N-scatterer streaming operator (beginning with the hard-core dynamics) leads to the binary collision expansion and circumvents the problem of evaluating the binary collision operator for all configurations, overlapping and nonoverlapping. In the paper of Ernst *et al.*,[17] the binary collision expansion is used as a starting point, and the various binary collision operators appearing in the literature are discussed as to whether they correctly represent the hard-core dynamics for nonoverlapping configurations. It is of interest to have these two reciprocal developments relating the hard-core dynamics and the binary collision expansion. A rigorous evaluation of the binary collision operator for hard-core interaction has not been published,[20] however, it can be bypassed using the approach of this paper.

III. CLUSTER DECOMPOSITION

An expansion of the N-scatterer dynamics, contained in the particle distribution function [Eq. (1.5)] in clusters of l-scatterer dynamics is usually derived from the Ursell-like expansion of the streaming operator[21]:

$$S_\epsilon(1,\ldots,N)=\sum_{B\subset(1,\ldots,N)} U_\epsilon(B)\,. \qquad (3.1)$$

The sum is over all subsets of the set $(1,\ldots,N)$. As a result of this cluster expansion, we obtain an expansion of the operator $\Gamma_\epsilon(\vec{r},\vec{p})$:

$$\Gamma_\epsilon(\vec{r},\vec{p})=S_\epsilon+\sum_{l=1}^{N}\frac{1}{l!}\int\cdots\int d\vec{\xi}_1\cdots d\vec{\xi}_l \times n(1,\ldots,l)\,U_\epsilon(1,\ldots,l)\,. \qquad (3.2)$$

The reduced distribution $n(1,\ldots,l)$ is given by

$$n(1,\ldots,l)=[N!/(N-l)!]\times\int\cdots\int d\vec{\xi}_{l+1}\cdots d\vec{\xi}_N\, Z_N^{-1}W(1,\ldots,N)\,. \qquad (3.3)$$

The cluster streaming operator $U(1,\ldots,l)$ may be written succinctly

$$U_\epsilon(1,\ldots,l)=S_\epsilon B(1,\ldots,l)\,, \qquad (3.4)$$

where $B(1,\ldots,l)=$ only those products in

the expansion of $\{1-\sum_1^l[B^\gamma(j)+B^v(j)]\}^{-1}$ that contain all l labels. (3.5)

It is well known that this cluster expansion [Eq. (3.2)] leads to divergent collision integrals.[22] Kawasaki and Oppenheim[23] first demonstrated how to sum the most divergent contributions using a sum over ring diagrams. The technique has successfully been applied to both the moderately dense gas[24] and the Lorentz gas.[11,13]

We demonstrate a modified cluster expansion that is equivalent to a summation of all divergent diagrams. The long-time divergence of all collision integrals is thereby regularized. The presentation is simplified with the following approximation: We allow overlap of the hard cores

$$Z_N^{-1}W(1,\ldots,N)S_\epsilon(1,\ldots,N)\to V^{-N}S_\epsilon(1,\ldots,N), \qquad (3.6)$$

$$S_\epsilon(1,\ldots,N)=S_\epsilon\sigma\left[1-\sum_1^N B^\gamma(j)\sigma\right]^{-1} \qquad (3.7a)$$

$$=S_\epsilon\left(1-\sum_1^N[B^\gamma(j)+B^v(j)]\right)^{-1} \qquad (3.7b)$$

and let $N!/(N-l)!\,V^l\to n^l$ in the limit $N\to\infty$, where $n=N/V$ is the density of scattering centers.

The divergence encountered in the usual cluster expansion [Eqs. (3.1)–(3.5)] is a consequence of allowing the particle to interact with l-scattering centers in the entire configuration space of the N-scattering centers. The possibility that other scattering centers can interrupt (or interfere with) the particle motion decreases the probability of long free streaming between successive real collisions. We can expect the virtual collisions to play a role in the collisional damping (reduction of the probability of long free streaming), since the virtual binary kernels have been shown to represent the configuration space restrictions of noninterruption of the particle motion.

We discuss a modified cluster expansion that explicitly demonstrates the connection between the products of virtual binary kernels in the σ operators and a collisional damping. We cluster expand the streaming operator of Eq. (3.7a) so that all virtual binary kernels are retained in each term

$$S_\epsilon(1,\ldots,N)=\sum_{B\subset(1,\ldots,N)} U_\epsilon(1,\ldots,N|B)\,, \qquad (3.8)$$

$U_\epsilon(1,\ldots,N|B)=$ only those products in the expansion of Eq. (3.7a) that contain all labels of the set B. (3.9)

The modified cluster expansion [Eqs. (3.8) and (3.9)] results in the expansion of Γ_ϵ

$$\Gamma_\epsilon(\vec{r},\vec{p})=\sum_{l=0}^{N}\Gamma_\epsilon^{(l)}(\vec{r},\vec{p})\,, \qquad (3.10)$$

$$\Gamma_\epsilon^{(l)}(\vec{r},\vec{p})=\binom{N}{l}\int\cdots\int d\vec{\xi}_1\cdots d\vec{\xi}_N V^{-N}U_\epsilon(1,\ldots,N|1,\ldots,l), \qquad (3.11)$$

where the superscript l denotes the number of real collisions [$U(1,\ldots,N|1,\ldots,l)$ contains the full dynamics for scattering with l scattering centers].

The first term in Eq. (3.10) represents streaming without interaction

$$\Gamma_\epsilon^{(0)}=V^{-N}\int\cdots\int d\vec{\xi}_1\cdots d\vec{\xi}_N S_\epsilon\sigma(1,\ldots,N)=S_{\epsilon+\nu}=\int_0^\infty dt\,e^{-(\epsilon+\nu)t}S(t)\,. \qquad (3.12)$$

The collision frequency ν is given by

$$\nu=-n\int d\vec{\xi}_1 B^v(1)S_\epsilon^{-1}=nv\int d\vec{b}_1, \qquad (3.13)$$

where $\vec{b}_1 = \vec{r}_1 \times \hat{v}$. Thus, the sum of products of virtual binary kernels in $\sigma(1, \cdots, N)$ gives rise to an exponential damping of the time of particle free streaming.

We verify the evaluation of $\Gamma_\epsilon^{(0)}$. The configuration space restrictions may be written

$$S_\epsilon \sigma(1, \ldots, N) = \int_0^\infty dt\, e^{-\epsilon t} \times \Big(1 - \sum_1^N A_j \theta(t - \tau_j^-) + \sum_{i \neq j} A_i A_j \theta(\tau_j^- - \tau_i^-)\theta(t - \tau_j^-) - \sum_{i \neq j \neq k \neq i} A_i A_j A_k \theta(\tau_j^- - \tau_i^-) \times \theta(\tau_k^- - \tau_j^-)\theta(t - \tau_k^-) + \cdots\Big) S(t) . \quad (3.14)$$

Equation (3.14) results from expanding $\sigma(1, \ldots, N)$ in a sum of products of virtual binary kernels and explicitly writing the Laplace transform integral. The restriction can be written in the more transparent form

$$S_\epsilon \sigma(1, \ldots, N) = \int_0^\infty dt\, e^{-\epsilon t} \prod_1^N [1 - A_j \theta(t - \tau_j^-)] S(t). \quad (3.15)$$

Thus, $S_\epsilon \sigma(1, \ldots, N)$ allows all configurations of the scattering centers that do not interrupt the free streaming for a time t. The exponential damping factor is obtained from the integral over the configuration space specified by Eq. (3.15), in the limit as $N \to \infty$.

A typical term in $\Gamma^{(1)}$, representing a single real collision is

$$S_\epsilon \sigma B^\gamma(1)\sigma = \prod_{j \neq 1} [1 - A_j \theta(\tau_1^- - \tau_j^-)] A_1 \times e^{-\epsilon \tau_1^-} S(\tau_1^-) \mathcal{R}_1 \times \int_0^\infty dt\, e^{-\epsilon t} \prod_{k \neq 1} [1 - A_k \theta(t - \tau_k^-)] S(t). \quad (3.16)$$

The scattering centers $\vec{\xi}_2, \ldots, \vec{\xi}_N$ are prevented from interrupting the particle motion before and after the real collision. They can occupy the entire volume V except for the region in a collision cylinder of volume V_c. Scattering center $\vec{\xi}_j$ is in V_c whenever the hard-core boundary (the hemisphere whose outward normal vector $\hat{n}$ satisfies $\hat{n} \cdot \vec{v} > 0$) intersects the particle trajectory. If τ is the duration of the trajectory, then an effective collision frequency ν^1 may be defined as

$$\nu^1 = n V_c / \tau . \quad (3.17)$$

The effective collision frequency depends on the geometry of the particle trajectory (in this case on the angle between $\vec{v}$ and $\mathcal{R}_1 \vec{v}$ and the times τ_1^- and t). It satisfies the inequality $\nu^1 \leq \nu$ with the equality holding only when the trajectory is rectilinear. The collision frequency ν^1 is less than ν [Eq. (3.13)], since the volume in which a scattering center can be located while its hard-core boundary intersects the trajectory, is less than the volume swept out by the hard-core cross section in a time τ, which is $v\tau \int d\vec{b}$.

The term representing l real collisions is given by

$$\Gamma_\epsilon^{(l)}(\vec{r}, \vec{p}) = (n^l / l!) \times \int \cdots \int d\vec{\xi}_1 \cdots d\vec{\xi}_l\, S_{\epsilon + \nu'} B^1(1, \ldots, l), \quad (3.18)$$

$B^1(1, \ldots, l)$ = only those products in

$$\Big(1 - \sum_1^l B'^{\gamma}(j)\Big)^{-1}$$

that contain all l labels, (3.19)

$$B'^{\gamma}(j) = \theta(\sigma - b_j)\delta(\tau_j^-)\mathcal{R}_j S_{\epsilon + \nu'} . \quad (3.20)$$

Equation (3.18) results from integrating the N-l scatterer positions $\vec{\xi}_{l+1} \cdots \vec{\xi}_N$ over the allowed volume in configuration space as determined by the σ operators.

Each collision event in $\Gamma^{(l)}$ (comprised of a sequence of real collisions with l scattering centers) contains exponential damping of the particle free streaming. The variable collision frequency ν^1 depends on the set of independent collision parameters specifying the particle trajectory. Virtual binary kernels or virtual collisions do not appear explicitly in the cluster operator $B^1(1, \ldots, l)$ rather they are contained implicitly in the exponential damping factor. It is precisely the nonconstancy of the collision frequency that renders the modified cluster expansion [Eq. (3.18)] unsuitable for deriving a generalized collision integral. Whatever advantage there may be in a representation containing only sequences of real collisions, is outweighed by the complicated dependence of ν^1 on the collision parameters. Thus, we find it necessary to develop a modified cluster expansion that contains some virtual binary kernels explicitly, while retaining the essential collisional damping.

Therefore, we write the streaming operator in the following form:

$$S_\epsilon(1, \ldots, N) = S_\epsilon \Big(1 - \sum_1^N B(j)\eta\Big)^{-1} , \quad (3.21)$$

$$B(j)=B^{\gamma}(j),\quad \text{if } j \text{ is a label contained in an irreducible sequence which is not repeated in the sequence,}$$

$$=B^{\gamma}(j)+B^{v}(j),\quad \text{otherwise,} \tag{3.22}$$

$$\eta=1,\quad \text{if } \eta \text{ does not appear between labels in the same irreducible, sequence,}$$

$$=\sigma^{1},\quad \text{otherwise,} \tag{3.23}$$

$$\sigma^{1}=\sigma(1,\ldots,N),\quad \text{such that the labels of the } B^{v}(j) \text{ kernels do not appear elsewhere in the product.} \tag{3.24}$$

An irreducible sequence is a sequence of labels that cannot be factored into disjoint products. Our rearrangement of the streaming operator places all virtual binary kernels that will contribute to the exponential damping factor in the σ^{1} operators. The remaining virtual binary kernels do not contribute to the damping and therefore will appear explicitly in the cluster expansion.

We perform a cluster expansion of $S_{\epsilon}(1,\ldots,N)$ [Eq. (3.21)] in the same manner as Eqs. (3.8) and (3.9) by collecting together all products of $B(j)$ operators having the same set of labels. The integral over the scattering center positions of the virtual binary kernels in the σ^{1} operators results in an exponential damping factor with a constant collision frequency ν. In the previous collisional damping [Eqs. (3.18)–(3.20)], all times of particle free streaming were damped with a variable collision frequency ν^{1}; in the present scheme, only those times of free streaming between collisions (real or virtual) in the same irreducible sequence are damped with a constant collision frequency ν.

The modified cluster expansion of $S_{\epsilon}(1,\ldots,N)$ [Eq. (3.21)] results in the l-scatterer operator,

$$\Gamma_{\epsilon}^{(l)}(\vec{r},\vec{p})=(n^{l}/l!)\times\int\cdots\int d\vec{\xi}_{1}\cdots d\vec{\xi}_{l}\,U_{\epsilon}^{M}(1,\ldots,l), \tag{3.25}$$

where

$$U_{\epsilon}^{M}(1,\ldots,l)=S_{\epsilon}\times \text{ all products in } [1-\textstyle\sum B^{M}(j)]^{-1} \text{ containing all } l \text{ labels,} \tag{3.26}$$

$$B^{M}(j)=B^{\gamma}(j)\gamma,\quad \text{if } j \text{ is a label in an irreducible sequence which appears only once in the sequence,}$$

$$=(B^{\gamma}(j)+B^{v}(j))\gamma,\quad \text{otherwise,} \tag{3.27}$$

$$\gamma=S_{\epsilon}^{-1}S_{\epsilon+\nu},\quad \text{if } \gamma \text{ occurs between two labels in the same irreducible sequence,}$$

$$=1,\quad \text{otherwise.} \tag{3.28}$$

The modified cluster expansion [Eqs. (3.21)–(3.28)] may also be obtained from a resummation procedure similar to the ring-diagram sum of Kawasaki and Oppenheim.[23] One would sum over all virtual binary kernels (contained in an irreducible sequence) whose labels do not appear elsewhere in the product. The ring-diagram sum for the Lorentz gas sums only over those irreducible sequences classified as rings, i.e., those sequences containing only two repeated labels. The present scheme regularizes all irreducible collision events.

Since we are considering the Lorentz gas with hard-core interaction, the collision frequency

$$\nu=-n\int d\vec{\xi}_{1}B^{v}(1)S_{\epsilon}^{-1},$$

is finite. In order to extend the regularization procedure to more general interactions, where ν diverges, it is necessary to sum over both real and virtual binary kernels to obtain a finite collision frequency

$$-n\int d\vec{\xi}_{1}[B^{\gamma}(1)+B^{v}(1)]\,S_{\epsilon}^{-1}.$$

Such a modification is accomplished with the substitutions: $B^{v}(j)\to B^{\gamma}(j)+B^{v}(j)$ in Eq. (3.24) and $B^{\gamma}(j)\to 1$ in Eq. (3.22), when j is a label in an irreducible sequence which is not repeated in the sequence.

IV. GENERALIZED COLLISION INTEGRAL

The generalized collision integral K_{ϵ} is obtained by substituting the cluster expansion of Γ_{ϵ} [Eqs. (3.10) and (3.25)] into the inversion formula, [Eq. (1.8)]. The transition $\Gamma_{\epsilon}\to K_{\epsilon}$, can also be considered as following from the irreducible factorization of Γ_{ϵ},[13] or from its expansion in connected diagrams.[25] The irreducible factorization of the operator Γ_{ϵ} has the form

$$\Gamma_{\epsilon}=S_{\epsilon}\Big(1-\sum_{l=1}^{\infty}K^{(l)}\Big)^{-1}. \tag{4.1}$$

$$K^{(l)}=(n^{l}/l!)$$

$$\times \int\cdots\int d\vec{\xi}_1\cdots d\vec{\xi}_l\, S_\epsilon^{-1}[\,U^M(1,\ldots,l)]_{\rm ir}\,, \tag{4.2}$$

where $[\cdots]_{\rm ir}$ restricts the sum of products in $U^M(1,\ldots,l)$ to irreducible sequences. The generalized collision integral is the sum over the irreducible products in Γ_ϵ

$$K_\epsilon = \sum_{l=1}^{\infty} K^{(l)} S_\epsilon^{-1}\,. \tag{4.3}$$

We demonstrate that the single-scatterer collision integral correctly yields the Boltzmann collision integral for the Lorentz gas. The first term in Eq. (4.3) is given by

$$K^{(1)} S_\epsilon^{-1} = n\int d\vec{\xi}_1\,[B^r(1)+B^v(1)]\,S_\epsilon^{-1}\,. \tag{4.4}$$

Decomposing $d\vec{\xi}_1$ into components along and perpendicular to the particle velocity $\vec{v}$, $d\vec{\xi}_1 = d\vec{b}_1 \times v d\tau_1^-$ ($d\vec{b}_1 = b_1 db_1 d\phi$, b_1 = impact parameter, ϕ_1 = azimuthal angle) we obtain the Boltzmann collision integral

$$K^{(1)} S_\epsilon^{-1} = nv\int d\vec{b}_1\,\{\mathfrak{R}_1 - 1\}\,. \tag{4.5}$$

We now discuss the generalization of the binary collision integral, or the l-scatterer collision integral. We choose as integration variables the set $(\vec{b}_j, \tau_j^-)$, where $\vec{b}_j$ is the impact vector of and τ_j^- the time of free streaming to the first encounter with the jth distinct scatterer [we relabel the products in Eq. (4.2) so that the jth distinct label, ordering from left to right, is j]. The jth distinct binary kernel contains the factor A_j, which determines the domain of $\vec{b}_j$ and τ_j^-. The θ functions in all the other binary kernels place further constraints on the set $(\vec{b}_j, \tau_j^-)$ which satisfy the requirements of the geometry of the particle trajectory. Thus, we may write the l-scatterer collision integral

$$K^{(l)} S_\epsilon^{-1} f_\epsilon(\vec{r},\vec{p}) = (nv)^l \int\cdots\int_{\substack{\text{collision}\\ \text{domain}}} d\vec{b}_1\cdots d\vec{b}_l\, d\tau_2^-\cdots d\tau_l^- \times\sum \exp[-(\epsilon+\nu)\tau_{\rm coll}]\,(-)^\mu f_\epsilon(\vec{r}',\vec{p}')\,. \tag{4.6}$$

The integral over the independent collision parameters has the domain which satisfies the requirements of the collision geometry (the product of θ functions determining the domain are written symbolically as the "collision domain"). The time, $\tau_{\rm coll}$, is the duration of the l-scatterer collision event, i.e., it is the time of free streaming from the hard-core boundary of the first encounter to the hard-core boundary of the last encounter. The sum $\sum$ is over all irreducible sequences (specifying real or virtual collision) formed from the l labels such that the labels first occur in the order 1, 2, 3, ..., and excluding virtual collisions with a scatterer whose label is not repeated. The factor, $(-)^\mu$, is the phase due to μ virtual collisions. The variables $\vec{r}'$ and $\vec{p}'$ are, respectively, the position and momentum of the particle just after the last collision. The presence of the damping factor $e^{-\nu\tau_{\rm coll}}$ ensures the existence of the integral over the collision times in the long time limit $\epsilon \to 0$.

The l-scatterer collision integral in the time domain has the form

$$(nv)^l \int\cdots\int d\vec{b}_1\cdots d\vec{b}_l\, d\tau_2^-\cdots d\tau_l^- \times\sum e^{-\nu\tau_{\rm coll}} \times\theta(t-\tau_{\rm coll})\,(-)^\mu f(\vec{r}',\vec{p}';t-\tau_{\rm coll})\,. \tag{4.7}$$

The generalized collision integral is the sum of formula (4.7) over the number of scattering centers.

V. DISCUSSION

We have presented (using the ϵ method) a generalization of the Boltzmann equation for a hard-sphere Lorentz gas, which regularizes all divergent terms. The role of the virtual binary kernels in representing the configuration space restrictions that prevent the scattering centers (not participating in a real collision sequence) from interrupting the particle motion has been demonstrated. The collisional damping factor has been shown to stem from these configuration space restrictions. In order to obtain a constant collision frequency, we have found it necessary to retain some virtual binary kernels (or their equivalent configuration space restrictions) explicitly in the generalized collision integral. We have given a prescription for the l-scatterer collision integral in the form of a convergent integral over a set of independent collision parameters. The collision events contained in the l-scatterer collision integral comprise all (l label) irreducible sequences of real and virtual binary kernels (excluding virtual binary kernels whose labels occur only once). The specification of these events is a rigorous consequence of the Liouville equation and cannot be obtained *a priori*.

The compact formula [Eq. (4.6)] for the l-scatterer collision integral may be replaced (when making explicit calculations) by the form in which the collision domain, rotation operators, free-

streaming operators, phase, etc., are written explicitly as products of binary kernels. Therefore, we rewrite the collision integral as

$$K^{(l)} S_{\epsilon}^{-1} = (n^l/l!) \times \int\cdots\int d\vec{\xi}_1\cdots d\vec{\xi}_l \, C(1,\cdots,l) S_{\epsilon+\nu}^{-1}, \quad (5.1)$$

$C(1,\ldots,l)$ = sum of irreducible products (containing l labels) of binary kernels $B'^{r}(j)$ and $B'^{v}(j)$, excluding virtual kernels whose labels are not repeated (5.2)

$$B'^{r}(j) = \theta(\sigma - b_j)\delta(\tau_j^-)\mathcal{R}_j S_{\epsilon+\nu}, \quad (5.3)$$

$$B'^{v}(j) = \theta(\sigma - b_j)\delta(\tau_j^-)(-) S_{\epsilon+\nu}. \quad (5.4)$$

The exponential damping appears in the modified free streaming operator

$$S_{\epsilon+\nu} = (\epsilon + \nu + \vec{v}\cdot\partial/\partial\vec{r})^{-1}.$$

Note that to write the collision integral in undamped form, we would remove the restriction on the virtual kernels that they have repeated labels, and would let $S_{\epsilon+\nu} \to S_{\epsilon}$. With this connection to the undamped collision integral, one can demonstrate the result of van Leeuwen and Weijland[11] that the coefficient of the logarithmic divergence is identical with the coefficient of the logarithmic density term.

An analogous regularization technique has been applied to the calculation of the self-diffusion coefficient of a moderately dense gas,[26] and the equivalence of the diagrams leading to the first logarithmically divergent term and the first logarithmic density-dependent term can be demonstrated in two and three dimensions.

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[1]N. N. Bogolyubov, in Studies in Statistical Mechanics, edited by J. de Boer and G. E. Uhlenbeck (North-Holland Publishing Co., Amsterdam, 1962), Vol. I.

[2]M. S. Green, J. Chem. Phys. 25, 836 (1956).

[3]E. G. D. Cohen, Physica 28, 1025 (1962).

[4]S. T. Choh and G. E. Uhlenbeck, University of Michigan, Navy Theoretical Physics Contract No. Nonr-1224 (15), 1958 (unpublished).

[5]M. H. Ernst, Physica 32, 209 (1966); in Lectures in Theoretical Physics, edited by W. E. Brittin (Gordon and Breach, Science Publishers, Inc., New York, 1967), Vol. 9C.

[6]J. V. Sengers, in Lectures in Theoretical Physics, edited by W. E. Brittin (Gordon and Breach, Science Publishers, Inc., New York, 1967), Vol. 9C.

[7]R. Zwanzig, Phys. Rev. 129, 486 (1963).

[8]K. Kawasaki and I. Oppenheim, Phys. Rev. 136, A1519 (1964).

[9]H. A. Lorentz, Collected Papers (Martinus Nijhoff, The Hague, 1935), Vol. VIII, p. 214.

[10]E. H. Hauge and E. G. D. Cohen, J. Math. Phys. 10, 397 (1969).

[11]J. M. J. van Leeuwen and A. Weijland, Physica 36, 457 (1967); 38, 35 (1968).

[12]Divergence in kinetic theory is discussed in a paper by J. R. Dorfman and E. G. D. Cohen, J. Math. Phys. 8, 282 (1967).

[13]The present paper is based on the author's dissertation, The University of Michigan, ORA Project 07497, 1967 (unpublished).

[14]M. H. Ernst, L. K. Haines, and J. R. Dorfman, Rev. Mod. Phys. 41, 296 (1969).

[15]The Laplace transform of the streaming operator is known as the resolvent operator. For a discussion of the streaming operator see Refs. 1, 4, and 8.

[16]The formalism expressing the collision operator in terms of the evolution operator [Eq. (1.6)] is known as the Zwanzig inversion scheme, see Ref. 7.

[17]M. H. Ernst, J. R. Dorfman, W. R. Hoegy, and J. M. J. van Leeuwen (to be published).

[18]The product of two binary kernels having the same label is zero, see Ref. 17.

[19]A. J. F. Seigert and E. Teramoto, Phys. Rev. 110, 1212 (1958); C. N. Yang and T. D. Lee, *ibid.* 113, 1165 (1959).

[20]J. R. Dorfman (private communication).

[21]J. De Boer, Rept. Progr. Phys. 12, 305 (1949).

[22]J. R. Dorfman and E. G. D. Cohen, Phys. Letters 16, 124 (1965); J. V. Sengers, Phys. Rev. Letters 15, 515 (1965); Phys. Fluids, 9, 1685 (1966); J. Weinstock, Phys. Rev. 140, A460 (1965); R. Goldman and E. A. Frieman, Bull. Am. Phys. Soc. 11, 531 (1965).

[23]K. Kawasaki and I. Oppenheim, Phys. Rev. 139, A1763 (1965).

[24]E. G. D. Cohen, in Fundamental Problems in Statistical Mechanics, edited by E. G. D. Cohen (North-Holland Publishing Co., Amsterdam, 1968).

[25]S. Fujita, Phys. Letters 10, 175 (1964).

[26]W. R. Hoegy and J. V. Sengers (to be published).