DENSITY EXPANSION OF THE VISCOSITY OF A MODERATELY DENSE GAS

J. V. Sengers

National Bureau of Standards, Washington, D. C. (Received 25 June 1965)

An expansion of the pair distribution function of a moderately dense gas not in equilibrium in powers of the number density n has been derived by various authors.^{1,2} With the aid of this result, also an expansion of the kinetic equation for the one-particle distribution function f_1 has been obtained leading directly to an expansion of the transport coefficients in powers of the density.³ It has been shown that a similar density series for the transport coefficients can be derived from the correlationfunction expressions.⁴⁻⁶

The coefficients of the successive powers of the density contain integrals which can be classified according to dynamical events, involving successively larger numbers of particles, for which the integrand does not vanish. One tries to establish the existence of these terms by showing that for any reasonable f_1 the phase volume, corresponding to the dynamical events that contribute to the integrand, is finite. This has been verified in three dimensions up to the triple collision term.^{2,4,5,7}

However, it was pointed out by Weinstock⁶ and Dorfman and Cohen⁸ that the phase space associated with certain dynamical events involving four or more particles is no longer finite. Goldman and Frieman, and recently also Swenson, arrived at a similar conclusion.⁹

The divergence of the phase volume implies that the corresponding terms in the density series both of the pair distribution function and of the transport coefficients become infinite, unless special cancellations occur, e.g., between contributions from different dynamical events which would lead to an essential reduction of the available phase space, as was pointed out by Dorfman and Cohen.⁸ It is the purpose of this Letter to investigate the latter point in more detail. To that end we have investigated the first density correction to the viscosity of a gas of rigid spheres in two dimensions (rigid disks). A divergence of the phase volume similar to the one corresponding to four particles in three dimensions occurs already in the triple collision term in two dimensions.¹⁰

Collisions involving three particles lead to a first density correction η_{k1} to the viscosity. In addition, the binary collisions lead to a kinetic contribution η_{k2} and a potential contribution η_{φ} to the viscosity linear in the density due to the spatial dependence of the distribution function.¹¹ Thus, up to terms linear in the density the viscosity η can be written as

$$\eta = \eta_0 + \eta_{k1} + \eta_{k2} + \eta_{c0}, \qquad (1)$$

where η_0 is the viscosity in the low-density limit. In order to study the divergence part we only consider the contribution η_{k1} from triple collisions. Limiting ourselves to the first Enskog approximation (i.e., only one Sonine polynomial is taken into account in the solution of the kinetic equation for f_1), η_{k1} is given by

$$\frac{\eta_{k1}}{\eta_0} = \frac{d}{n^2 \sigma} \int d\vec{\mathbf{V}}_1 d\vec{\mathbf{V}}_2 d\vec{\mathbf{V}}_3 \int d\vec{\mathbf{r}}_2 d\vec{\mathbf{r}}_3 \vec{\mathbf{V}}_1^{\ 0} \vec{\mathbf{V}}_1 : \theta_{12} \tau_2(12,3) f_0(V_1) f_0(V_2) f_0(V_3) \{\vec{\mathbf{V}}_1^{\ 0} \vec{\mathbf{V}}_1 + \vec{\mathbf{V}}_2^{\ 0} \vec{\mathbf{V}}_2 + \vec{\mathbf{V}}_3^{\ 0} \vec{\mathbf{V}}_3\},$$
(2)

where σ is the diameter of the disks, \vec{V}_i the peculiar velocity and \vec{r}_i the position vector of particle *i*, and $f_0(V_i)$ the normalized equilibrium Maxwell distribution. The differential operator θ_{12} and the dynamical operator $\tau_2(12,3)$ have been defined elsewhere.¹² The coefficient *a* is related to the solution of the ordinary Boltzmann equation

$$a = \frac{1}{8\sqrt{\pi}} \left(\frac{m}{kT}\right)^{5/2}.$$
 (3)

In order to evaluate the integral in (2) we

transform it into a surface integral by the method of Green.¹² A detailed analysis in both two and three dimensions will be published in the future. Three dynamical events have to be studied; these are shown in Fig. 1. If the motion is followed backwards in time in all three diagrams, the collision between 1 and 2 is followed by a collision between 1 and 3. The diagrams differ in that as a third collision, 1 and 2 can collide again (recollision), 2 and 3 can collide (cyclic collision), or 3 would have col-



FIG. 1. Schematic representation of the dynamical events.

lided with 2 if the collision between 1 and 3 had not caused any change in velocities (hypothetical cyclic collision).¹² In general, one also has to consider the contribution from four successive real collisions.¹³ However, they do not contribute to the divergent part and therefore do not need to be considered here.

We use as integration variables in the evaluation of (2) the velocities of the particles, the collision parameters of the first (1-2) and the second (1-3) collision, and the time τ between these two collisions as indicated by Green.¹² The latter time is made dimensionless by introducing the variable $\tau^* = \tau/\tau_0$, where τ_0 is the mean free time needed to travel a distance σ . Carrying out the integration over all variables except τ^* , it turns out that the resulting integrand for large values of τ^* behaves as τ^{*-1} . Consequently, if we take into account all events with various τ^* up to a maximum value T^* , the triple-collision contribution to the viscosity diverges as

$$\eta_{k1}^{\prime} / \eta_0^{=} A n \sigma^2 \ln T^* + O(n \sigma^2).$$
 (4)

One may also consider T^* as the maximum time difference between the first and the last collision as the two time intervals are linearly related in the limit $T^* \rightarrow \infty$.

It can be shown that for the model considered, the coefficient A is the sum of the following three contributions corresponding to the three different types of triple collision events:

recollisions:
$$A_{\gamma} = \frac{1}{225} \frac{\sqrt{3}}{\pi} \left(216 - 112 \frac{\pi}{\sqrt{3}} \right);$$

cyclic collisions: $A_{c} = \frac{1}{225} \frac{\sqrt{3}}{\pi} (226 - \frac{1}{2}405 \ln 3);$

hypothetical collisions: $A_h = -\frac{1}{225} \frac{\sqrt{3}}{\pi} \times 32$.

The contributions from the different diagrams do not cancel, and the total coefficient A be-

comes

$$A = \frac{1}{225} \frac{\sqrt{3}}{\pi} \left(410 - 112 \frac{\pi}{\sqrt{3}} - \frac{1}{2} 405 \ln 3 \right)$$
$$\simeq -0.03826.$$
(5)

Of course, the possibility that this coefficient would vanish if all higher Enskog approximations would be taken into account still cannot be ruled out entirely, but seems very unlikely.

If T^* would be chosen proportional to the mean free time between collisions and, consequently, proportional to $1/n\sigma^2$, then -A would represent the coefficient of a contribution proportional to $n\sigma^2 \ln n\sigma^2$.¹⁴

Finally, we remark that the Enskog theory of dense gases gives for the considered contribution to the viscosity of a gas of rigid disks

$$\frac{\eta_{k1}}{\eta_0} = -\left(\frac{4}{3} - \frac{\sqrt{3}}{\pi}\right) \frac{1}{2}\pi n \sigma^2 \simeq -1.228n \sigma^2.$$

As remarked by Dorfman and Cohen,⁸ the latter contribution is obtained if the triple-collision integral is evaluated in a much more limited part of phase space (corresponding to socalled overlapping configurations).

The author is much indebted to M. S. Green for his encouragement and advice, and to E. G. D. Cohen and J. R. Dorfman for stimulating discussions leading to the analysis described. He also is indebted to R. A. Piccirelli and J. M. J. van Leeuwen for valuable suggestions.

¹E. G. D. Cohen, Physica <u>20</u>, 1025 (1962).

²M. S. Green and R. A. Piccirelli, Phys. Rev. <u>132</u>, 1388 (1963).

³S. T. Choh and G. E. Uhlenbeck, "The Kinetic Theory of Phenomena in Dense Gases," University of Michigan Research Report, 1958 (unpublished).

⁴R. Zwanzig, Phys. Rev. <u>129</u>, 486 (1963); K. Kawasaki and I. Oppenheim, Phys. Rev. <u>136</u>, A1519 (1964).

⁵M. H. Ernst, J. R. Dorfman, and E. G. D. Cohen, Physica <u>31</u>, 493 (1965); E. G. D. Cohen, in Proceedings of the IUPAP Conference on Statistical Mechanics, Aachen, 1964 (unpublished), p. 140.

⁶J. Weinstock, Phys. Rev. <u>132</u>, 454 (1963); J. Weinstock, to be published.

⁷S. Ono and T. Shizume, J. Phys. Soc. Japan <u>18</u>, 29 (1963); J. Weinstock, Phys. Rev. <u>132</u>, 470 (1963).

⁸J. R. Dorfman and E. G. D. Cohen, Phys. Letters <u>16</u>, 124 (1965).

⁹R. Goldman and E. A. Frieman, Bull. Am. Phys. Soc. <u>11</u>, 531 (1965); R. J. Swenson, private communication.

¹⁰J. R. Dorfman, private communication.

 ${}^{11}\eta_{k2} + \eta_{\varphi} = \eta_0 \pi n \sigma^2/2$ for rigid disks. ${}^{12}M$. S. Green, Phys. Rev. <u>136</u>, A905 (1964). ${}^{13}G$. Sandri, R. D. Sullivan, and P. Norem, Phys. Rev. Letters <u>13</u>, 743 (1964); J. Foch and E. G. D. Cohen,

private communication.

¹⁴Recently, Kawasaki and Oppenheim informed the author that they actually found a term logarithmic in the density by resumming the formal density series.

ANOMALOUS BEHAVIOR OF SUPERCONDUCTING MICROGEOMETRIES IN THE PRESENCE OF HIGH CURRENT DENSITIES*

R. P. Groff, J. M. Mochel, and R. D. Parkst

University of Rochester, Rochester, New York (Received 6 August 1965)

In this Letter we report two striking effects we have observed in the behavior of superconducting microgeometries in magnetic fields and in the presence of high current densities.

The first of the anomalous effects occurs in the sample geometry shown in Fig. 1. This geometry is prepared by machining the design with a sharp tungsten point on a 1500Å-thick Sn film which is on a glass substrate. The microbridge at point A is a few microns long and a few microns wide. A small copper coil at point B (not shown) together with the superconducting circuit forms a transformer, and therefore a constant-voltage ac source, which supplies current to the microbridge when an



FIG. 1. x-y recorder traces of voltage versus magnetic field for a 2.7μ -wide Sn microbridge. The arrows indicate whether the sweep was obtained by increasing or decreasing the field. In each trace the field was increased from zero to some terminal point and then decreased to zero. The different traces correspond to different values of the terminal sweep point. The base lines correspond to zero resistance. The resistance corresponding to the voltage peak is approximately that of the normal resistance of the microbridge.

ac current is fed to the copper coil. A small copper Helmholtz pair provides at point A a magnetic field which is perpendicular to the plane of the film. Points A and B are far apart so that the fringing field of the driving coil at B is negligibly small at the microbridge, and the fringing field of the Helmholtz coil at point A is negligibly small at point B. A lock-on amplifier, which has as its reference source the signal to the copper coil at B, is used to measure the voltage across the microbridge.

In Fig. 1 is shown an x-y recorder tracing of the voltage across a 2.7μ -wide Sn microbridge as a function of the magnetic field when 150kc/sec current is flowing through the bridge. An abrupt voltage spike is observed at a particular value of the magnetic field as the field is increased and again at a higher value of the field as the field is decreased. The location of the second peak depends upon the magnitude of the excursion of the magnetic field before the field is decreased. We note the following observations from a study of five microbridges with widths ranging from 2.7 to 10 μ .

(1) The voltage spikes are observed only at frequencies above 10 kc/sec.

(2) Sharp voltage spikes are obtained at a given temperature only for current densities which lie in a very narrow range. If the current is too small, there are no voltage spikes. If the current is too large, the spikes broaden into wide rectangular steps.

(3) If the field sweep is stopped at the peak of the voltage spike, the voltage persists at the peak level.

(4) The magnetic field at which the first spike occurs (H_1) depends only weakly upon the width of the microbridge. An increase in the width of the microbridge from 2.7 to 10 μ results in a decrease of H_1 of approximately 30%.

(5) H_1 is temperature dependent, increasing