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The relaxation time of bi-particle correlations in a gas (*)

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Résumé. — Le temps de relaxation des corrélations est la durée moyenne de passage dans la partie positive du potentiel. Le rôle de l'environnement est précisé à partir de la notion d'interaction triple dégénérée. Une formule du coefficient de viscosité cinétique en bon accord avec l'expérience est alors obtenue.

Abstract. — The relaxation time of correlations is the average crossing time through the positive part of the potential. The role of the environment is specified by using the idea of a degenerated triple interaction. A formula for the kinetic viscosity coefficient, in good agreement with experiment, is thus obtained.

1. **Introduction.** — The postulate of linear relaxation suggested by J. Frey and J. Salmon is written below, ${}^{12}\Psi$ and ${}^{123}\Psi$ designating the position correlations functions at local equilibrium [1], [2], [3] :

$${}^{12}\chi = {}^{12}f - {}^1f {}^2f {}^{12}\Psi \quad (1)$$

$${}^{123}\chi = {}^{123}f - {}^1f {}^2f {}^3f {}^{123}\Psi \quad (2)$$

$$\int \frac{d^3x d^3w F_\beta^3}{m} \frac{\partial {}^{123}f}{\partial w_\beta} = \frac{{}^{12}\chi}{\tau} \quad (3)$$

The notations used are those of P. Hoffmann [4], [5].

τ designates the average value of the crossing time through the repulsive part of the interaction potential during a binary collision and 1f is the single distribution function. This postulate leads to the Frey-Salmon kinetic equation in which 1n is the particle density, 1v the average velocity vector, T the temperature and k Boltzmann's constant :

$$\frac{\partial {}^1f}{\partial t} + w_\alpha \frac{\partial {}^1f}{\partial x_\alpha} = \frac{{}^1nkT}{2m} \tau B \left[3 {}^1f + (w_\alpha - v_\alpha) \frac{\partial {}^1f}{\partial w_\alpha} + \frac{kT}{m} \frac{\partial^2}{\partial w_\alpha \partial w_\alpha} {}^1f \right] \quad (4)$$

B is the integral :

$$B = - \frac{8\pi}{3kT} \int_0^\infty \frac{d\varphi}{dx} \frac{d{}^{12}\Psi}{dx} (x)^2 dx$$

thus, for a dilute gas, with ${}^{12}\Psi = \exp\left(-\frac{\varphi}{kT}\right)$, φ is the interaction potential

$$B = \frac{8\pi}{3k^2T^2} \int_0^\infty \left(\frac{d\varphi}{dx}\right)^2 \exp\left(-\frac{\varphi}{kT}\right) (x)^2 dx \quad (5)$$

The expression for the kinetic viscosity coefficient $\mu = m(\tau B)^{-1}$ only agrees with experimental results when one introduces a coefficient of scale c common to all the gases in calculating τ . Thus, for a Sutherland potential characterized by a diameter σ and a minimum potential $-kT_i$, the following formula was obtained where

$\mu = \frac{m}{\tau B}$ was calculated with the form limit when $T_m \rightarrow +\infty$ [8], [9] :

(*) La traduction en français de cet article a été soumise pour publication aux Comptes Rendus de l'Académie des Sciences.

$$\mu = \frac{9}{64 c \sigma^2} \left(\frac{mkT}{\pi} \right)^{1/2} \exp\left(-\frac{T_i}{T}\right) \left[2 \int_0^{+\infty} \left(\lambda^2 + \frac{T_i}{T} \right)^{3/2} e^{-\lambda^2} d\lambda - \sqrt{\pi} \left(\frac{T_i}{T} \right)^{3/2} \right]^{-1}, \quad (6)$$

$$\mu = \lim_{T_m \rightarrow \infty} \frac{m}{\tau B},$$

where :

$$\tau = c \frac{m\sigma}{kT_m} \frac{16}{3\sqrt{\pi}} \left(\frac{kT}{m} \right)^{1/2} \int_0^{\infty} \left[\left(\lambda^2 + \frac{T_i}{T} \right)^{3/2} - \left(\frac{T_i}{T} \right)^{3/2} \right] e^{-\lambda^2} d\lambda .$$

$$B = \frac{8\pi\sigma}{3} \left[\frac{T}{T_m} \exp\left(-\frac{T_m - T_i}{T}\right) \left[\exp\left(\frac{T_m}{T}\right) \left(\frac{T_m^2}{T^2} - \frac{2T_m}{T} + 2 \right) - 2 \right] \right] + \frac{8\pi\alpha}{3k^2 T^2} \int_{\sigma}^{\infty} \left(\frac{d\varphi}{dr} \right)^2 \exp\left(-\frac{\varphi}{kT}\right) r^2 dr . \quad (7)$$

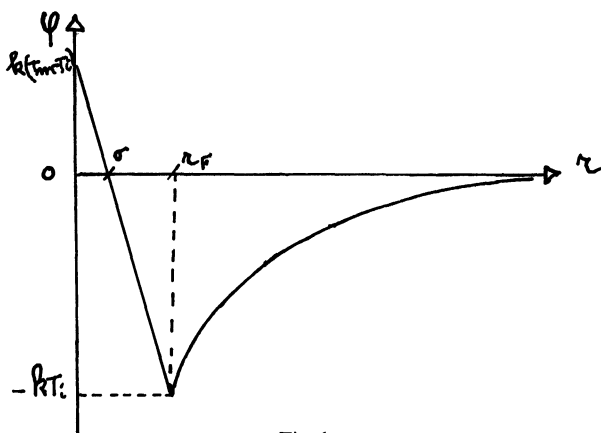


Fig. 1.

2. **Dissipation zone.** — We suggest a new definition of the characteristic time of linear relaxation starting from the ideas of a dissipation zone and of a degenerate triple interaction.

The intermolecular potential φ exhibits a repulsive part which is particularly intense for φ positive — i.e. for r smaller than σ . We consider this bounded sphere as a dissipation zone and associate with it τ_2 which is the average crossing time through this zone of the reduced particle of mass $m/2$ and of velocity g relative to infinity (Fig. 2).

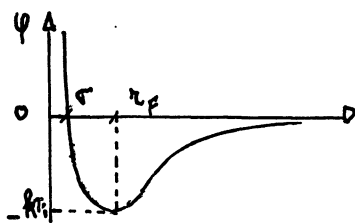


Fig. 2.

On designating the impact parameter by b and the distance of least approximation by r_m , the expression for τ_2 is written :

$$\tau_2 \simeq 2 \int_0^{\sigma} \int_0^{\infty} \left[\int_{r_m}^{\sigma} \left(g^2 \left(1 - \frac{b^2}{r^2} \right) - \frac{4}{m} \varphi(r) \right)^{-1/2} dr \right] \frac{2\pi b}{\pi\sigma^2} \left(\frac{m}{4\pi kT} \right)^{3/2} \exp\left(-\frac{mg^2}{4kT}\right) 4\pi g^2 db dg . \quad (8)$$

3. **Linear relaxation and the degenerated triple interaction.** — The fine distribution functions of the B.B.G.K.Y. hierarchy are probabilistic and non-statistical away from equilibrium [6]. This is why a temporal average over a suitable time leads to semifine distribution functions with which one introduces hypotheses of irreversibility and predicts measurable quantities.

For these conditions, let us write the second B.B.G.K.Y. equation :

$$\frac{\partial}{\partial t} + w_\beta \frac{\partial}{\partial x_\beta} \frac{F_\beta^{12}}{m} \frac{\partial}{\partial w_\beta} {}^{12}f + \int d^3x d^3w \frac{F_\beta^3}{m} \frac{\partial^{123}f}{\partial w_\beta} = 0. \tag{9}$$

Using the relation (2), the last term is separated into three parts

$$\int d^3x d^3w \frac{F_\beta^3}{m} \frac{\partial}{\partial w_\beta} {}^{123}f = \int d^3x d^3w {}^3f {}^{123}\Psi \left[\frac{F_A^3}{m} {}^2f \frac{\partial^1f}{\partial w_A} + \frac{F_B^3}{m} {}^1f \frac{\partial^2f}{\partial w_B} \right] + \int d^3x d^3w \frac{F_A^3}{m} \frac{\partial^{123}\chi}{\partial w_A} + \int d^3x d^3w \frac{F_B^3}{m} \frac{\partial^{123}\chi}{\partial w_B}. \tag{10}$$

The number 1 and 2 particles evolve according to a two-particle Liouville equation and consequently conserve their velocity correlation as long as neither collides with a third particle.

When one of these events takes place, one of the last two terms of (10) is involved.

Henceforth, the hypothesis of linear relaxation is reduced to an irreversibility quotient from of ${}^{12}\chi$ by τ_2 , thus :

$$\int d^3x d^3w \frac{F_A^3}{m} \frac{\partial^{123}\chi}{\partial w_A} = \frac{{}^{12}\chi}{\tau_2}, \tag{11}$$

and an identical term relative to the number 2 particle.

The whole expresses the degeneration of the triple interaction for the velocity correlations and the fundamental expression (3) is reobtained with :

$$\tau = \frac{\tau_2}{2}. \tag{12}$$

Thus, the relaxation time intervening in the kinetic equation becomes equal to half of the average crossing time through the repulsive part of the interaction potential.

With a Sutherland potential [7], the formula of the viscosity coefficient becomes :

$$\mu = \frac{9}{32} \left(\frac{mkT}{\pi} \right)^{1/2} \frac{1}{\sigma^2} \exp\left(-\frac{T_i}{T}\right). \tag{13}$$

In the case of argon, the relative difference between theory and experiment [10] is less than 1 % from 300 to 2 200 K (Table I).

4. Conclusion. — The introduction of the idea of a dissipation zone, defined as the repulsive and positive

Table I. — Viscosity coefficient of argon.

T K	10 ⁵ μ _{Th} Pa.s	10 ⁵ μ _{exp} [9] Pa.s	Δμ/μ
—	—	—	—
250	1.926	1.949	0.012
300	2.272	2.272	0.000
400	2.877	2.852	0.009
600	3.865	3.830	0.009
700	4.280	4.250	0.008
1 000	5.372	5.350	0.004
1 300	6.283	6.280	0.000
1 600	7.083	7.100	0.002
1 900	7.803	7.840	0.005
2 200	8.463	8.510	0.006

$$\sigma = 2.829 \times 10^{-10} \text{ m}; \quad T_i = 110.8 \text{ K}; \\ m = 6.633 \times 10^{-26} \text{ kg}.$$

part of the potential where the forces are very intense, and the fine analysis of the triple interaction term in BY2, allow us to improve the first theory deduced from the hypothesis of linear relaxation. By means of a simple formula we obtain a strict agreement with experiment.

It is advisable to note that for T_i equal to zero, this formula becomes that of the viscosity of a dense sphere gas. The numerical coefficient 9/32 is then to be compared with those deduced from Boltzmann's equation and from the mean free path theory which give respectively 10/32 and 2/3 π which encompasses the value 9/32.

Also, the disappearance of the scale c coefficient of formula (6) seems to be an argument in favour of the ideas introduced in this note.

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