

ON THE DISPERSION OF SOUND IN HELIUM

by
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I. Introduction

The propagation of plane sound waves through a gas where the mean free path of the molecules is not completely negligible compared with the wave length of sound was first studied by Primakoff⁽¹⁾ and later again by Tsien and Schamberg⁽²⁾. The starting point of their calculation is the Burnett equations, i.e., the hydrodynamical equations obtained from the velocity distribution function of a non-uniform gas correct up to the second approximation according to the method of development devised by Hilbert and Enskog. In our last report⁽³⁾ we pointed out how their results were in error because of an error in sign of one of the Chapman's coefficients. After correcting this it has been found that to the second order of approximation and for both the elastic spheres and the Maxwellian molecules the change of the velocity is about six or seven times larger than the results obtained from Chapman's original expressions. Furthermore the effect is about twice as large as would be obtained by treating the Stokes-Navier equations as exact. In fact the effect is of such an order of magnitude that experimental verification seems to be possible. Hence it is thought to be worthwhile to carry out the calculation for one real gas using the best known knowledge of the force

(1) H. Primakoff, J. Acous. Soc. Am. 13, 14, 1942.

(2) H. S. Tsien and R. Schamberg, J. Acous. Soc. Am. 18, 334, 1946.

(3) "On the Transport Phenomena in Rarified Gases" by C. S. Wang Chang and G. E. Uhlenbeck. APL/JHU CM-443, Feb. 20, 1948.

law. This task has been made much easier because of the work of Dr. Hirschfelder* and his co-workers who calculated all the collision integrals $\Omega^{(n)}$ necessary for the determination of the coefficients in the pressure tensor and the heat flux vector.

The dispersion formula derived in this way by using the revised Burnett-Chapman pressure tensor and heat flux vector holds true only for a monoatomic gas where there are no internal degrees of freedom. For polyatomic gases, the additional absorption and dispersion of sound due to each internal degrees of freedom are characterized by the relaxation time, τ_i , the time required for the i th internal degree of freedom to attain equilibrium with the translational motion. Only when the τ_i are known does one know whether this correction to the velocity is negligible compared to the contributions due to the viscosity μ and the heat conduction coefficient γ . From the formula for the elastic spheres and the Maxwell molecules it is seen that the latter effect will be appreciable only when $(f/p)^2 \sim 10^5$ where f is the frequency in megacycles per second and p is the pressure in atmospheres; under atmospheric pressure this corresponds to a value of $1/f$ of the order of 10^{-9} seconds. On the other hand experiments seem to indicate that the rotational relaxation time for most gases are of the order 10^{-8} - 10^{-9} seconds. Thus in most cases the relaxation effect will mask the effects due to μ and γ . We therefore choose to study the affect in a monoatomic gas and especially helium because its interatomic force has been most extensively studied.

(4) See Chapman and Cowling, "The Mathematical Theory of Non-uniform Gases".

* We like to take this opportunity to express our gratitude to Dr. Hirschfelder for sending us their reports and permitting us to make free use of them.

We will limit ourselves to classical calculations. To avoid making the necessary quantum corrections we will calculate the effect for the temperature range $T = 200$ to 500°K . This temperature range has the further advantage that the non-ideal behavior of helium can be neglected.

In the next section we will give the general expression for the coefficients ω and Θ in the second order pressure tensor and heat flux vector, and show how they can be written formally as proportional to μ^2 , μv , or v^2 . In section 3 we will give the derivation for the velocity of sound up to the second approximation. The last section will be devoted to the application to helium.

II. The Second Order Pressure Tensor and Heat Flux Vector; the Coefficients

ω and Θ

The expression for the pressure tensor has been given by both Burnett and Chapman, and in his book, Chapman also derived the heat flux vector. In our report⁽³⁾ we have presented them in tensor notation. There the values of the Θ 's and ω 's are given for elastic spheres and for Maxwell molecules.* The aim of this section is mainly to give general expression for Θ and ω and to show how they are connected with the collision cross-sections. In the problem of sound propagation one usually limits oneself to small amplitudes so that only linear terms need be considered. To save writing we will restrict ourselves therefore to the linear terms. The heat flux vector and

*Some of the numbers in Table II on p. 13 of the previous report are in error. The third column should read:

θ_3	Multiplied by 1.030	instead of	Multiplied by 1.039
θ_4	Multiplied by 0.806		Multiplied by 0.814
θ_5	$\frac{\mu^2}{PT} \left(\frac{105}{4} \times 0.918 \right.$		$\frac{\mu^2}{PT} \left(\frac{105}{4} \times 0.930 \right.$
	$\left. + 3 \times 0.806 \frac{T}{\mu} \frac{d\mu}{dT} - 0.150 \right)$		$\left. + 3 \times 0.814 \frac{T}{\mu} \frac{d\mu}{dT} - 0.150 \right)$

the pressure tensor can then be written as:

$$p_i^{(2)} = \theta_2 \cdot \frac{2T}{3} \frac{\partial^2 u_\alpha}{\partial x_i \partial x_\alpha} + \theta_4 \frac{\partial}{\partial x_\alpha} (D_{i\alpha} - \frac{D_{\lambda\lambda}}{3} \delta_{i\alpha}) \quad (1)$$

$$p_{ij}^{(2)} = - \frac{\omega_2}{\rho} \left(\frac{\partial^2 p}{\partial x_i \partial x_j} - \frac{1}{3} \frac{\partial^2 p}{\partial x_\alpha \partial x_\alpha} \delta_{ij} \right) + \omega_3 \left(\frac{\partial^2 T}{\partial x_i \partial x_j} - \frac{1}{3} \frac{\partial^2 T}{\partial x_\alpha \partial x_\alpha} \delta_{ij} \right)$$

Thus among the θ 's and the ω 's only θ_2 , θ_4 , ω_2 , and ω_3 will be of interest to us. These are given by:

$$\begin{aligned} \theta_2 &= - \frac{2k^2 T}{3m n^2} \int f^{(0)} A^2 c^2 d\vec{\xi} \\ \theta_4 &= \frac{4k^2 T^2}{15m n^2} \int f^{(0)} A B c^4 d\vec{\xi} \\ \omega_2 &= \frac{2kT}{15n^2} \int f^{(0)} B^2 c^4 d\vec{\xi} \\ \omega_3 &= \frac{4k^2 T}{15m n^2} \int f^{(0)} A B c^4 d\vec{\xi} \end{aligned} \quad (2)$$

A and B are functions of c , the dimensionless molecular velocity,

$$c_i = \sqrt{\frac{m}{2kT}} \xi_i$$

and the solutions of the integral equations:

$$\begin{aligned} nI(Ac_i) &= f^{(0)} \left(c^2 - \frac{5}{2} \right) c_i \\ nI \left[B(c_i c_j - \frac{c^2}{3} \delta_{ij}) \right] &= 2f^{(0)} \left(c_i c_j - \frac{c^2}{3} \delta_{ij} \right) \end{aligned} \quad (3)$$

I is a linear and isotropic operator defined by:

$$\begin{aligned} n^2 I(\phi) &= \int d\vec{\xi}_1 \int d\Omega g I(g, \theta) f^{(0)} f_1^{(0)} (\phi + \phi_1 - \phi' - \phi'_1) \\ &= \int d\vec{\xi}_1 \int d\Omega g I(g, \theta) f^{(0)} f_1^{(0)} \Delta \phi \end{aligned}$$

where g is the relative velocity, $I(g, \theta)$ is the collision cross-section, and ϕ' , ϕ'_1 are the values of ϕ and ϕ_1 after collision. The integral equations (3) can not be solved in closed form. One solves them by expanding A and B in sets of orthogonal functions, the most convenient sets being the Sonine polynomials. One writes: (5)

$$A = \sum_{r=1}^{\infty} a_r S_{\frac{3}{2}}^{(r)}(c^2) \quad (4)$$

$$B = \sum_{r=1}^{\infty} b_r S_{\frac{5}{2}}^{(r-1)}(c^2)$$

Substituting (4) into (3), multiplying by $S_{\frac{3}{2}}^{(m)}(c^2)c_i$ and $S_{\frac{5}{2}}^{(m-1)}(c^2)(c_i c_j - \frac{c^2}{3} \delta_{ij})$ respectively, contracting and integrating over $d\vec{\xi}$, one obtains for both a_1 and b_1 an infinite set of linear equations of the form:

$$\sum_{r=1}^{\infty} a_r a_{rm} = -\frac{15}{4} \delta_{m1} \quad (5)$$

$$\sum_{r=1}^{\infty} b_r b_{rm} = 5 \delta_{m1}$$

where

$$a_{rm} = \int d\vec{\xi} S_{\frac{3}{2}}^{(m)}(c^2) c_\alpha \int d\vec{\xi}_1 d\Omega g I(g, \theta) f^{(0)} f_1^{(0)} \Delta [S_{\frac{3}{2}}^{(r)}(c^2) c_\alpha]$$

$$b_{rm} = \int d\vec{\xi} S_{\frac{5}{2}}^{(m-1)}(c^2) (c_\alpha c_\beta - \frac{c^2}{3} \delta_{\alpha\beta}) \int d\vec{\xi}_1 d\Omega g I(g, \theta) f^{(0)} f_1^{(0)} x$$

$$\Delta [S_{\frac{5}{2}}^{(r-1)}(c^2) (c_\alpha c_\beta - \frac{c^2}{3} \delta_{\alpha\beta})]$$

(5) See Chapman and Cowling, p. 123 for the definitions of the Sonine polynomials $S_m^{(n)}(x)$; we will follow as much as possible the notation of Chapman and Cowling.

Both are eight fold integrals and for centrally symmetric molecules, six of these integrals can be carried out leaving only the collision integrals of the type:

$$\Omega^l(n) = \sqrt{\pi} \left(\frac{m}{4kT} \right)^{n+\frac{3}{2}} \int_0^\infty dq e^{-\frac{mq^2}{4kT}} q^{2n+3} \int_0^\pi d\theta \sin \theta (1 - \cos^l \theta) I(q, \theta)$$

Chapman has calculated a_{rs} and b_{rs} for r and s up to $r = s = 3$. (6):

The linear equations (5) are solved by successive approximations. To the first approximation one stops at the first term in the sum obtaining:

$$a_1 = -\frac{15}{4a_{11}} \quad b_1 = \frac{5}{b_{11}}$$

In general to the m th approximation one stops at $r = s = m$, and

$$a_i = \frac{(-1)^i \frac{15}{4} A_{ii}}{A} \quad b_i = \frac{(-1)^{i-1} 5 B_{ii}}{B}$$

where A is the determinant $|a_{rs}|$, and A_{ii} is the minor of the element a_{ii} . The a and b are functions of the temperature and they depend on the force law.

One can show that to all order of approximations the following relations always hold true:

$$a_1 = -\frac{2m}{5k^2T} \nu = -\frac{3}{2} \frac{\nu'}{kT} \quad (6)$$

$$b_1 = \frac{2\mu}{kT}$$

(6) See p. 161, Chapman and Cowling.

and to simplify later writing we have written:

$$\gamma = \frac{15}{4} \frac{k}{m} \gamma'$$

where γ' has the same dimension as μ . In fact, it is exactly μ for Maxwell molecules. For other spherically symmetric molecular models

$$\gamma' = s\mu.$$

s is a dimensionless temperature dependent quantity. It is different for different force laws but it is never very much different from unity. To the first approximation s is 1 for all these models. Furthermore, one can show that for molecules interacting with the inverse power law r^{-n} all the a 's and b 's have the same temperature dependence, so that their ratios are independent of the temperature. It is therefore convenient to write:

$$\theta_2 = \varepsilon_1 \frac{\gamma^{1/2}}{\rho T} \quad \theta_4 = T\omega_3 = \frac{\varepsilon_2 \mu \gamma'}{\rho} \quad \omega_2 = \varepsilon_3 \frac{\mu^2}{\rho} \quad (7)$$

where the coefficients ε_1 , ε_2 , and ε_3 are dimensionless constants independent of T for the r^{-n} law molecules. This way of writing is only formal. For other molecular models the ε 's are still temperature dependent.

III. The Velocity of Sound.

In this section we will derive the expression for the velocity of sound from the Burnett equations. This will, of course, be mainly a repetition of known work, but to get uniformity in notation it seems not too superfluous to do it briefly again. Making use of (7) and (1), and considering only one dimensional motion one obtains for the total pressure tensor and the total

heat flux vector up to the second order the following expressions:

$$\begin{aligned}
 p_{xx} &= p - \frac{4}{3} \mu \frac{\partial u}{\partial x} - \frac{2}{3} \varepsilon_3 \frac{\mu^2}{\rho \rho} \frac{\partial^2 p}{\partial x^2} + \frac{2}{3} \varepsilon_2 \frac{\mu \gamma'}{\rho T} \frac{\partial^2 T}{\partial x^2} \\
 q_x &= -\frac{15k}{4m} \gamma' \frac{\partial T}{\partial x} + \frac{2}{3} \varepsilon_1 \frac{\gamma'^2}{\rho} \frac{\partial^2 u}{\partial x^2} + \frac{2}{3} \varepsilon_2 \frac{\mu \gamma'}{\rho} \frac{\partial^2 u}{\partial x^2}
 \end{aligned} \tag{8}$$

The linear one dimensional Burnett equations are:

$$\begin{aligned}
 \frac{1}{\rho} \frac{\partial p}{\partial t} + \frac{\partial u}{\partial x} &= 0 \\
 \rho \frac{\partial u}{\partial t} + \frac{\partial p}{\partial x} - \frac{4\mu}{3} \frac{\partial^2 u}{\partial x^2} - \frac{2\varepsilon_3}{3} \frac{\mu^2}{\rho \rho} \frac{\partial^3 p}{\partial x^3} + \frac{2\varepsilon_2}{3} \frac{\mu \gamma'}{\rho T} \frac{\partial^3 T}{\partial x^3} &= 0 \\
 \frac{3}{2T} \frac{\partial T}{\partial t} + \frac{\partial u}{\partial x} - \frac{15}{4} \frac{\gamma'}{\rho T} \frac{\partial T}{\partial x} + \frac{2\varepsilon_1}{3} \frac{\gamma'^2}{\rho \rho} \frac{\partial^3 u}{\partial x^3} + \frac{2\varepsilon_2}{3} \frac{\mu \gamma'}{\rho \rho} \frac{\partial^3 u}{\partial x^3} &= 0
 \end{aligned} \tag{9}$$

Eliminating u from these equations and expression T in terms of p and ρ by the equation of state and finally introducing

$$\begin{aligned}
 \frac{p-p_0}{p_0} &= \pi_0 e^{i(\sigma x - \omega t)} \\
 \frac{\rho-\rho_0}{\rho_0} &= r_0 e^{i(\sigma x - \omega t)}
 \end{aligned} \quad \pi_0, r_0 \ll 1$$

one obtains two homogeneous equations for π_0 and r_0 . Equating the determinant to zero leads then to the dispersion equation:

$$\begin{aligned}
 V_0^2 \sigma^2 = \omega^2 + \frac{4i}{3} \frac{\mu}{\rho_0} \omega \sigma^2 + \frac{5i}{2} \frac{\gamma'}{\rho_0} \omega \sigma^2 - \frac{3i}{2} \frac{\gamma'}{\rho_0} V_0^2 \frac{\sigma^4}{\omega} \\
 - \frac{10}{9} \frac{\varepsilon_3 \mu^2}{\rho_0^2} \sigma^4 + \frac{\mu \gamma'}{\rho_0^2} \sigma^4 \left(\frac{8}{9} \varepsilon_2 - \frac{10}{3} \right) + \frac{4\varepsilon_1}{9} \frac{\gamma'^2}{\rho_0^2} \sigma^4
 \end{aligned} \tag{10}$$

where V_0 is the velocity at zero frequency. Equation (10) relates the angular frequency ω to the σ , which is in general complex

$$\sigma = \sigma_1 + i\sigma_2$$

The real part σ_1 is related to the velocity of sound while the imaginary part σ_2 gives the absorption coefficient. Equation (10) can be solved for σ by successive approximations by treating " μ " and " γ' " as small quantities. This is obviously consistent with the Hilbert-Enskog development.

1) Zeroth order: Euler

$$V = V_0$$

$$\sigma_2 = 0$$

2) First order: Stokes-Navier

$$V = V_0$$

$$\sigma_2 = \frac{2}{3} \frac{\mu}{\rho_0} \frac{\omega^2}{V_0^3} + \frac{1}{2} \frac{\gamma'}{\rho_0} \frac{\omega^2}{V_0^3}$$

3) Second order: Burnett

$$V = V_0 \left\{ 1 + \left(\frac{2}{3} + \frac{5}{9} \varepsilon_3 \right) \frac{\mu^2}{\rho_0^2} \frac{\omega^2}{V_0^4} + \left(\frac{5}{3} - \frac{4}{9} \varepsilon_2 \right) \frac{\mu \gamma'}{\rho_0^2} \frac{\omega^2}{V_0^4} \right. \\ \left. - \frac{\gamma'^2}{\rho_0^2} \frac{\omega^2}{V_0^4} \left(\frac{3}{8} + \frac{2}{9} \varepsilon_1 \right) \right\} \quad (11)$$

$$\sigma_2 = \frac{2}{3} \frac{\mu}{\rho_0} \frac{\omega^2}{V_0^3} + \frac{1}{2} \frac{\gamma'}{\rho_0} \frac{\omega^2}{V_0^3}$$

Formally one can go on and on, the equation (10) can even be solved exactly. But for a consistent theory one should stop at the second approximation because we have not taken the terms of the third order in μ and γ' into account in the pressure tensor and the heat flux vector. Thus the influence

of the Burnett terms on the absorption coefficient will not be seen till one includes in the equations $p^{(3)}$ and $q^{(3)}$. In the present paper we will limit ourselves to the second approximation and therefore will calculate only the velocity of sound. The second order effect due to the Stokes-Navier terms alone can be obtained from (11) by putting the ε 's equal to zero.

IV. Application to Helium.

For the interaction between helium atoms we take the Lennard-Jones potential:

$$V(r) = 4\varepsilon \left[\left(\frac{r_0}{r} \right)^{12} - \left(\frac{r_0}{r} \right)^6 \right]$$

where ε is the minimum for $V(r)$ and r_0 is the value of the interatomic distance for which $V = 0$. Dr. Hirschfelder and his co-workers have made numerical evaluations of the collision integrals $\Omega^{\ell}(n)$ that are necessary for the calculation of a_{rs} and b_{rs} up to $r = s = 3$ for different values of kT/ε .

Using de Boer and Michels' ⁽⁷⁾ values for ε and r_0

$$\varepsilon = 1.402 \times 10^{-15} \text{ ergs}$$

$$r_0 = 2.56 \text{ \AA}$$

we have computed up to the third approximation the a 's and b 's for $kT/\varepsilon = 20, 30, 40, 50$, corresponding to temperatures 203.4, 305.1, 406.8, and 508.5°K. It is found that for these temperatures and within the accuracy of the available values of $\Omega^{\ell}(n)$ the ratios a_i/a_1 and b_i/b_1 are independent of T . The result is shown in Table I. For comparison we give in Table I also the values for Maxwell molecules and the elastic spheres.

(7) J. de Boer and Michels, *Physic* 5, 945, 1938; 6, 409, 1939.

Table I

	a_2/a_1	a_3/a_1	b_2/b_1	b_3/b_1
Maxwell	0	0	0	0
Lennard-Jones	0.0672	0.0127	0.0435	0.0061
Elastic spheres	0.0944	0.0194	0.0613	0.0094

This shows that within this temperature range the helium gas behaves as simple repulsive centers of the power law $5 < n < \infty$.

In Table II are the values of the ϵ 's and γ'/μ for the different models. To exhibit the convergence we give in Table III the values of ϵ in the successive approximations.

Table II

	Maxwell	de Boer and Michels	Elastic spheres
ϵ_1	-45/8	$-(45/8) \times 1.008$	$-(45/8) \times 1.017$
ϵ_2	3	3×0.855	$3 \times 0.800^*$
ϵ_3	2	2×1.007	2×1.014
γ'/μ	1.000	1.004	1.009

Table III

	First Approx.	Second Approx.	Third Approx.
ϵ_1	-45/8	$-(45/8) \times 1.007$	$-(45/8) \times 1.008$
ϵ_2	3	3×0.862	3×0.855
ϵ_3	2	2×1.006	2×1.007

Putting these values for ϵ into equation (11) we find for the velocity of sound

* The difference between this value and the value given on p. 12 of (3) is because of the ratio γ'/μ .

$$V = V_0 \left[1 + \frac{230.9}{72} \frac{\mu^2 \omega^2}{p_0^2 V_0^4} \right]$$

or

$$\frac{\Delta V}{V_0} = \frac{230.9}{72} \frac{\mu^2 \omega^2}{p_0^2 V_0^4} = 44.4 \mu^2 \left(\frac{f}{p} \right)^2 \quad (12)$$

where μ is in c.g.s. units, f in megacycles per second and p is in atmospheres. Treating Stokes-Navier as exact would give:

$$\frac{\Delta V}{V_0} = 27.2 \mu^2 \left(\frac{f}{p} \right)^2 \quad (13)$$

In Table IV are the computed $\Delta V/V_0$ from equations (12) and (13). The values of μ in the second column are the theoretical values. In this temperature range they agree to within 1% of the values calculated from the empirical formula

$$\mu = 5.023 T^{0.647} \times 10^{-6} \text{ c.g.s.}$$

given by Keesom⁽⁸⁾.

(8) W. H. Keesom: Helium p. 107. A detailed discussion on the comparison of the theoretical and experimental results will be published in a paper by Dr. Hirschfelder in the Journal of Chemical Physics.

Table IV

T °K	μ -theory c.g.s.	V/V_0 in % Stokes-Navier	V/V_0 in % Burnett
203.4	1.58×10^{-4}	$0.67 \times 10^{-4} (f/p)^2$	$1.10 \times 10^{-4} (f/p)^2$
305.1	2.05 "	1.14 "	1.86 "
406.8	2.47 "	1.66 "	2.71 "
508.5	2.85 "	2.21 "	3.61 "

Thus we see that the effect is really not too small. For a value of $f/p = 335$ corresponding to, say, $p = 10^{-2}$ atm. and $f = 3.35$ mc and at $T = 305.1^\circ\text{K}$

$$\frac{\Delta V}{V_0} = 20.8 \%$$

in contrast to the 3% obtained by Tsien and Schamberg. Of course, for $\Delta V/V_0$ so large the next term in the development will not be negligible. To get the next term in the velocity one would have to calculate $p^{(4)}$ and $q^{(4)}$.

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