Ultrasonic attenuation and dispersion in deuterium-neon gas mixture

R. G. HARLOW and R. A. BACON (GUILDFORD)

MEASUREMENTS of the ultrasonic absorption and dispersion in deuterium–neon gas mixtures have been made at temperatures of 293 K, 473 K and 873 K using a non-interferometric technique. With an ultrasonic frequency of 1 MHz, and using deuterium mole fractions of 1.0, 0.75, 0.5, 0.25 and 0.1, each experiment covered a range of frequency upon pressure ratios of from 1 MHz/atm to about 100 MHz/atm. Corrections for the diffraction effects that occur in disc to disc coupling have been calculated using a model based upon numerical integration, that allows the transmitter to have a nonuniform amplitude distribution, and the receiver to have a nonuniform amplitude sensitivity. This feature was necessary because of the long quartz buffer rods required to transmit the ultrasound into and out of the high temperature environment of the gas under test.

Wykonano pomiary ultradźwiękowej absorpcji i dyspersji w mieszaninach gazowych deuteru i neonu w temperaturach 293, 473, 673 i 873°K posługując się techniką interferometryczną. Przy częstości ultradźwiękowej 1 MHz i posługując się ułamkami molowymi deuteru 1,0, 0,75, 0,5, 0,1, w każdym doświadczeniu uwzględniono zakres stosunków częstości do ciśnienia zmieniający się od 1 MHz/atm do ok. 100 MHz/atm. Poprawki wynikające ze współoddziaływania między poszczególnymi krążkami określono posługując się modelem opartym na całkowaniu numerycznym, co pozwalało uwzględnić nierównomierny rozkład amplitudy w nadajniku i nierównomierny rozkład czułości w odbiorniku. Była to własność istotna ze względu na długie pręty kwarcowe potrzebne do przenoszenia ultradźwięków do wnętrza i na zewnątrz obszarów wysokotemperaturowych w badanym gazie.

Проведены измерения ультразвуковой абсорбции и дисперсии в газовых смесях дейтерия и неона в температурах 293, 473, 673 и 873° К, послуживаясь интерферометрической техникой. При ультразвуковой частоте 1 Мгц и послуживаясь молярными дробями дейтерия 1,0, 0,75, 0,5 и 0,1, в каждом эксперименте учтен интервал отношений частот к давлению, исменяющийся от 1 Мгц/атм до примерно 100 Мгц/атм. Поправки, вытекающие из взаимодействия между отдельными кружками, определены послуживаясь моделью опирающейся на численном интегрировании, что позволило учитывать неравномерное распределение амплитуды в передатчике и неравномерное распределение чувствительности в приемнику. Это свойство существенно из-за длинных кварцевых стержней, необходимых для переноса ультразвуков внутрь и вне высокотемпературных областей в исследуемом газе.

1. Experimental

THE TECHNIQUE used involves passing short bursts of ultrasound through a sample of the required gas mixture and measuring the propagation parameters, from which the rotational relaxation times may be derived. The main features of the experimental arrangement are shown in schematic form in Fig. 1. The synthetic quartz buffer rods a with the PZT5 piezoelectric transducers b fixed to their outer ends are supported by the stainless steel end flange assemblies c. The stainless steel cylinder d of the test chamber is mounted within a cylindrical furnace (not shown) and is connected to the end flanges by means of stainless steel bellows e. Titanium steel encased thermocouples f are used for the furnace temperature control, and for monitoring the gas temperature.
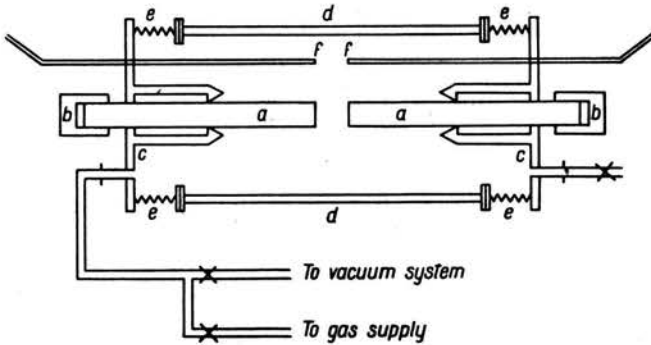
FIG. 1. Schematic diagram of the test chamber, a — synthetic quartz buffer rods, b — piezoelectric transducers in housings, c — stainless steel end flanges and buffer rod supports, d — stainless steel cylinder, e — stainless steel bellows, f — thermocouple probes.

Measurements are made by observing the changes in both the amplitude and the phase of the received signal as the path length in the gas is slowly increased by withdrawing the transmitting buffer rod and transducer. The data obtained consists of a set of amplitude measurements, and a set of position measurements for the transmitting buffer rod, both at received signal phase intervals of π.

2. Diffraction corrections

In the past [1] these data sets have been used directly to calculate the absorption and the velocity, but it is now well established that the diffraction effects that occur in the transmission of sound from a disc to a disc should be taken into account. SEKI, GRANATO and TRUELL [2] and others [3] have investigated the form of the transfer function for plane piston-like transducers transmitting through a non-absorbing medium (see for example curves A in Fig. 2). Due to the use of the buffer rods, it was felt that it was unlikely that the amplitude and phase would be constant across the transmitting face or that the amplitude and phase sensitivity would be constant across the receiving face.

To resolve this question, each rod was set up as the transmitter, and the amplitude and phase distributions across each rod's end face were measured. It was found that the phase differences were negligeable but that the radially symmetric amplitude distributions were as shown in Fig. 3. No appropriate theory could be found for the coupling of a non-piston-like source to a receiver of nonuniform amplitude sensitivity through an absorbing medium, and so a computer model of the situation was developed. In Fig. 2, curves A show the transfer function generated by the model for piston-like transducers in a non-absorbing medium, which is in agreement with RHYNE's recent analytical treatment [4]. Curves B show the effect of a highly absorbing medium on the transfer function, and this is in agreement with the calculations of KHIMUNIN [5] for this case. Curves C are the generated transfer function using the measured transmitter and receiver amplitude functions with a non-absorbing medium. Measurements are made at transducer separations of up to 25 mm. and it can be seen that the corrections in this region can be signifi-
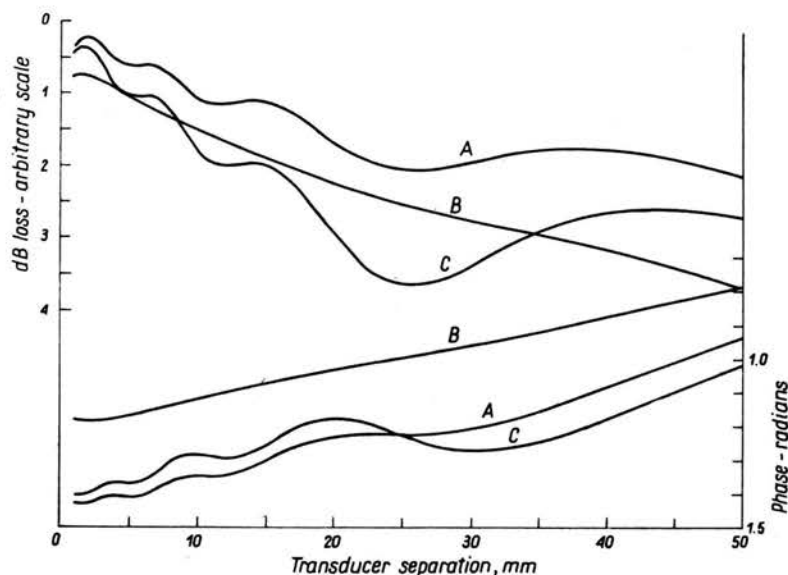
FIG. 2. Disc to disc transfer functions generated by computer model. Curves A — piston-like transducers in a non-absorbing medium, B — piston-like transducers in a highly absorbing medium, C — transducers with measured amplitude functions in a non-absorbing medium.
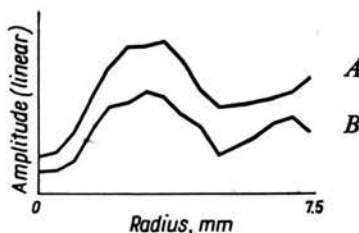

FIG. 3. Amplitude distribution along a radius of the buffer rod ends. Curve A — rod used as transmitter, B — rod used as receiver.

cantly different from those for the simple piston-like transducer case. Thus for each experimental arrangement approximate values of the velocity and absorption were obtained and used to generate correction data by which the experimental values were modified, before calculating the final velocity and absorption.

3. Classical absorption calculations

The results of measurements made on pure neon gas at 200°C are shown in Fig. 4 plotted in the dimensionless form used by GREENSPAN [6]. The solid lines represent the theoretical values of the velocity and absorption, and the triangles and circles are the experimental points. In this case of a pure monatomic gas at low frequencies, the absorp-
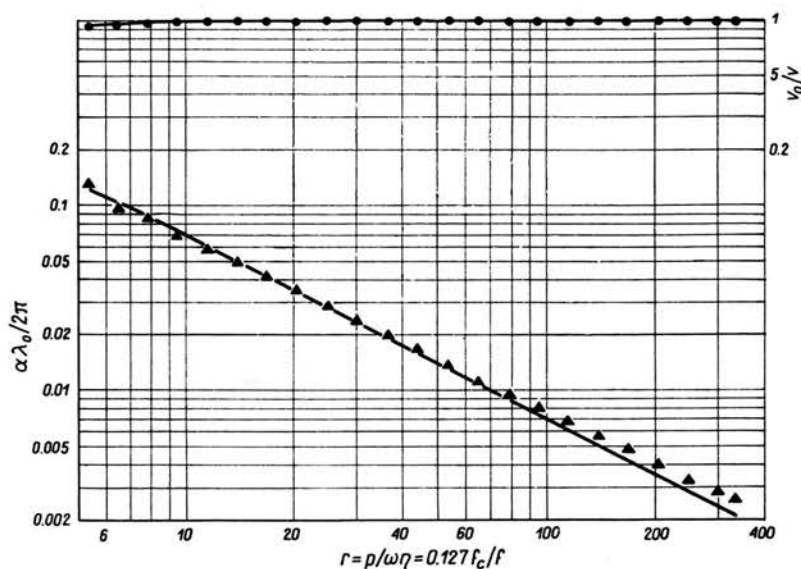
FIG. 4. Neon at 200°C. Propagation parameters as functions of r, Triangles — from experimental absorption, circles — from experimental velocity, solid lines — Burnett solution.

tion can be calculated in at least two different but equivalent ways. The classical absorption α_{cl} [7] in terms of the viscosity η and the thermal conductivity K is given by

$$\alpha_{cl}\lambda = \frac{2\pi^2}{\gamma}\frac{f}{p}\left[\frac{4}{3}\eta + \frac{(\gamma-1)K}{c_p}\right],$$

where f is the frequency, $\gamma = c_p/c_v$, c_p, c_v are the specific heats, p is the pressure, and λ is the wavelength.

Alternatively, the GREENSPAN form of the BURNETT expression [6] is

$$k^6\left(-\frac{21}{125}\frac{1}{r^4} - \frac{18}{25}\frac{i}{r^3}\right) + k^4\left(-\frac{97}{50}\frac{1}{r^2} + \frac{9}{10}\frac{i}{r}\right) + k^2\left(1 + \frac{23}{10}\frac{i}{r}\right) + 1 = 0,$$

$$k = \frac{\alpha\lambda_0}{2\pi} + i\frac{v_0}{v},$$

$$r = \frac{p}{\eta 2\pi f},$$

where λ_0 is the low frequency wavelength, v_0 and v are the low frequency and normal velocities.

These are equivalent expressions for values of $r \gg 10$, but for $r < 10$ the Burnett solution departs from the linear classical solution.

For a pure diatomic gas the Burnett theory has been used by GREENSPAN [6] to predict the non-molecular part of the absorption by using a multiplier to modify the value of r so as to form an effective r_e to be used in the calculation. The effect of this can be seen in Fig. 5 where the theoretical absorption curve has been clearly shifted slightly in the

direction of decreasing r. The experimental absorption is greater than the classical theoretical value shown because of the additional absorption due to the rotational degrees of freedom of the deuterium molecules. Similarly the departure of the velocity from the theoretical values at high frequencies is due to the loss of the rotational component of the specific heat.
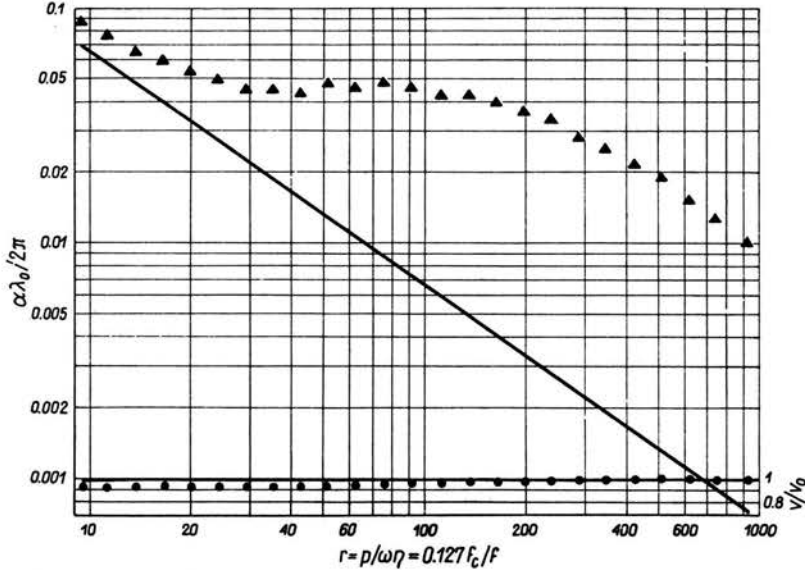

Fig. 5. Deuterium at 200°C. Propagation parameters as functions of r. Triangles — from experimental absorption, circles — from experimental velocity, solid lines — modified Burnett solution.
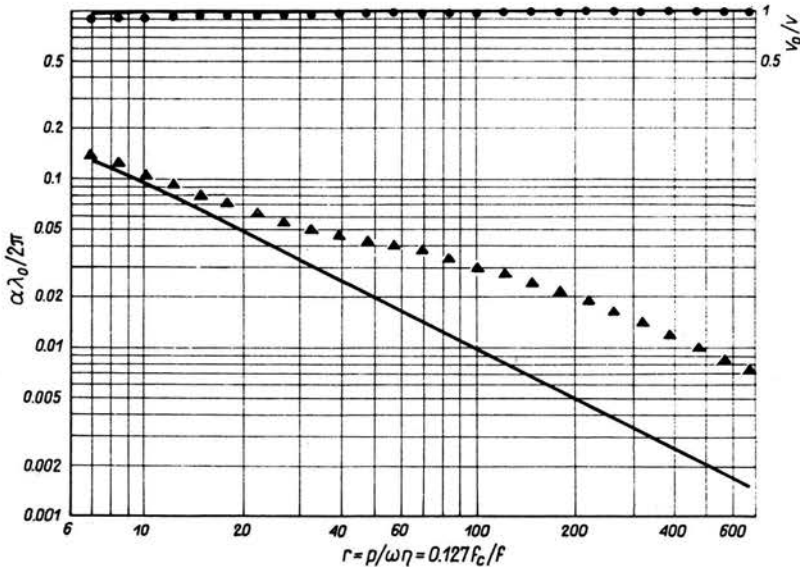

Fig. 6. 50/50 deuterium/neon mixture at 20°C. Propagation parameters as function of r. Triangles — from experimental absorption, circles — from experimental velocity, solid lines — modified Burnett solution.

In the case of a gas mixture the expression for the classical absorption acquires an additional term due to the interdiffusion of the two gas species. In the form given by Köhler [8] the expression for the classical absorption becomes

$$\alpha_{cl}\lambda = \frac{2\pi^2}{\gamma}\frac{f}{p}\left[\frac{4}{3}\eta + \frac{(\gamma-1)K}{c_p} + \frac{\gamma^2 x_1 x_2 p D_{12}}{v^2}\left\{\frac{M_2-M_1}{M} + \frac{\gamma-1}{\gamma}\frac{k_T}{x_1 x_2}\right\}^2\right],$$

where $x_1 x_2$ are the mole fractions of the two gas species, v is the sound velocity in the mixture, $M_1 M_2$ are the molecular weights of the two components and $M = M_1 x_1 + M_2 x$, D_{12} is the binary diffusion coefficient and k_T is the diffusion ratio.

This term has the effect of increasing the theoretical absorption and we have modified the Burnett solution in a manner similar to that employed for the case of the pure diatomic gas. The value of r used in the Burnett calculation is modified by a multiplier chosen such that the low frequency Burnett absorption is the same as that obtained from Kohler's expression. This is illustrated in Fig. 6 for a 50/50 mixture of deuterium and neon.

4. Molecular absorption

The same data is used in Fig. 7 to plot the excess absorption (i.e. the experimental values minus the classical values), and the velocity (rather than the dispersion) against frequency/pressure ratio. In addition a theoretical absorption curve for a single relaxation process has been fitted to the experimental points by a least squares method, and is shown by the solid line. The theoretical velocity for this relaxation process is shown by
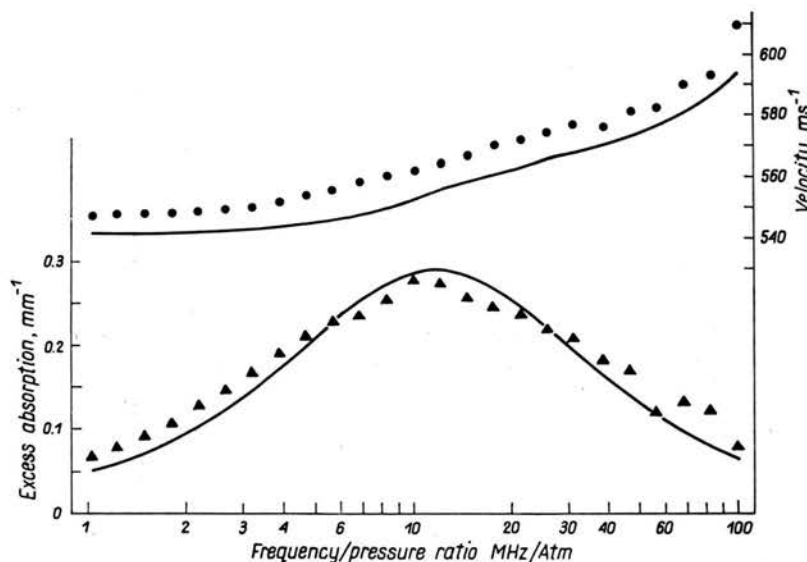

Fig. 7. 50/50 deuterum/neon mixture at 20 C. Excess absorption and velocity as function of frequency/pressure ratio. Triangles — excess experimental absorption over modified Burnett, circles — experimental velocity, solid lines — theoretical single relaxation process absorption and velocity fitted to excess absorption only.

the upper full line, this velocity being that predicted by the parameters found by fitting the absorption curve. These theoretical curves are given by [7]:

$$\frac{v^2}{v_0^2} = 1 + \frac{(C_p - C_v)C'}{C_p(C_v - C')} \cdot \frac{(\omega\tau(C_v - C')/C_v)^2}{1 + (\omega\tau(C_v - C')/C_v)^2},$$

$$\alpha_{mol}\lambda = \pi \frac{v^2}{v_0^2} \frac{(C_p - C_v)C'}{(C_p - C')C_v} \frac{\omega\tau(C_p - C')/C_p}{1 + (\omega\tau(C_p - C')/C_p)^2},$$

where C_p, C_v are the molar specific heats, C' is the molar specific heat of the relaxation process, $\omega = 2\pi f$ and τ is the relaxation time of the process.

Note that the (unexplained) difference between the theoretical velocity and the measured velocity is nearly constant at about 1%. It should also be noted that it is expected that there will be several relaxation processes occurring so that the absorption curve for the single process would be expected to have too large a maximum compared to the experimental points.

Figure 8 shows a similar plot of the data from a 50/50 deuterium/neon mixture at 626°C and it clearly shows two possibly related problems that are besetting the analysis. The first is that at high temperatures and at high frequency/pressure ratios, the measured absorption is sometimes less than the theoretical value (even without molecular processes). The other is that the measured velocity appears to increase with the frequency/pressure ratio at a significantly higher rate than can be accounted for by relaxation alone. The hypothesis that both of these effects could be the result of differential absorption of the two gas species by the chamber walls is currently under investigation.
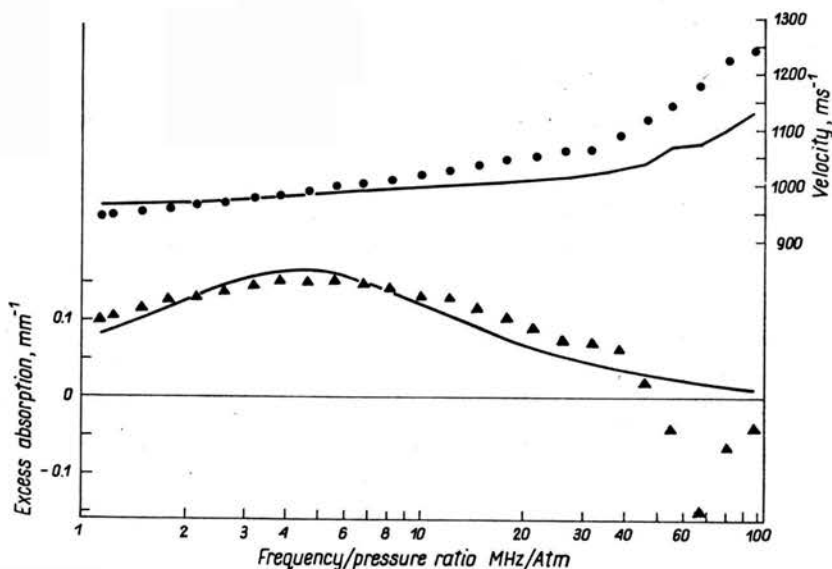

Fig. 8. 50/50 deuterium/neon mixture at 600°C. Excess absorption and velocity as function of frequency/pressure ratio. Triangles — excess experimental absorption over modified Burnett, circles — experimental velocity, solid lines — theoretical single relaxation process absorption and velocity fitted to excess absorption only.

7*

5. Conclusion

Measurements have been made on mixtures of deuterium and neon at four different concentrations at the nominal temperatures of 20°C, 200°C and 400°C and for three different concentrations at 600°C. For each set of absorptions obtained, the specific heat and the relaxation time of a hypothetical single relaxation process was obtained as described above. The specific heats thus obtained are plotted in Fig. 9 (a), (b), (c) and (d) for the 15 mixtures and also for four additional sets of measurements made on pure deuterium.
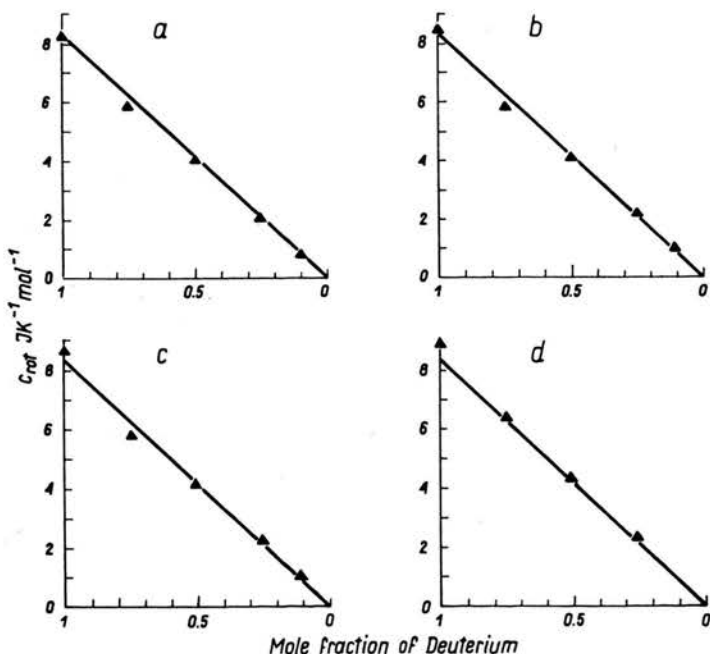

FIG. 9. Rotational specific heats found by fitting a single relaxation process to the experimental excess absorption data for mixtures of deuterium and neon at (a) 20°C, (b) 208°C, (c) 420°C, (d) 626°C.

The fit of this preliminary analysis appears to us to be quite reasonable, particularly when it is remembered that as the temperature is increased, the number of relaxation processes involved will also increase so that the single process approximation will become progressively less valid.

References

1. S. J. BLACKER, Ph. D. Thesis, University of Surrey, 1977.
2. H. SEKI, A. GRANATO, R. J. TRUELL, J. Acous. Soc. Am., 28, 230–238, 1956.
3. M. B. GITIS, A. S. KHIMUNIN, Sov. Phys. Acous., 14, 413–431, 1969.
4. T. L. RHYNE, J. Acous. Soc. Am., 61, 318–324, 1977.

5. A. S. Khimunin, Acustica, **27**, 173–181, 1972.
6. M. Greenspan, J. Acous. Soc. Am., **31**, 155–160, 1959.
7. K. I. Herzfeld, T. A. Litovitz, *Absorption and dispersion of ultrasonic waves*, Academic Press, 1959.
8. M. Kohler, Annal der Phys., **39**, 209–225, 1951.

DEPARTMENT OF PHYSICS
UNIVERSITY OF SURREY, ENGLAND.