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PROPAGATION OF HYPERSONIC WAVES IN SOME VISCOUS FLUIDS

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Taking advantage of Mandelsztam-Brillouin dispersion the velocity of propagation and the absorption coefficient of hypersonic waves at a frequency of 8 GHz have been measured in the following viscous fluids: castor oil, paraffin oil, diethylene glycol, triethylene glycol and polyethylene glycol. The results obtained are discussed on the basis of relaxation theory and ISAKOVICH-CZABAN theory.

1. Introduction

An analysis of the results of measurements of the velocity of propagation and the absorption coefficient of hypersonic waves in very viscous fluids [4, 5, 8, 9] leads to the conclusion that relaxation theory [6] cannot be applied to them since there is considerable divergence between the experimental data and theoretical predictions. In the hypersonic region this divergence is even more conspicuous. However, the range of experimentation at very high frequencies is relatively small because of unfavourable conditions and this hinders comparison between theory and experiment.

By virtue of the very high absorption, the only proper method of investigating the propagation of hypersonic waves in very viscous fluids is an examination of the spectrum of molecular light diffusion. As a result of the scattering of photons by thermal phonons there appear, in the spectrum of scattered light, components of changed frequency, the so called Mandelsztam-Brillouin components [1]. The vibration frequencies of these components are shifted relative to the primary light frequency by an amount given by the formula

$$f = 2nf_0\frac{v}{c}\sin\frac{\theta}{2}, \tag{1}$$

where f_0 is the primary frequency of the light wave, n — coefficient of light refraction, v — velocity of hypersonic wave, c — velocity of light, θ — scattering angle.

The absorption of hypersonic waves which always occurs in a medium causes a certain broadening of components of the fine structure

$$\delta\nu = \frac{\alpha v}{\pi c}, \tag{2}$$

where $\delta\nu$ denotes the half width expressed in wave numbers, and α the coefficient of absorption of hypersonic wave.

Thus, if we measure the distance between the fine structure components and their half widths, we can evaluate from formulae (1) and (2) the velocity of propagation and the coefficient of absorption of the hypersonic waves.

In this paper – by taking advantage of this method – the velocity of propagation and absorption coefficient of hypersonic waves in castor oil, paraffin oil, di-, tri- and polyethylene glycol have been measured. The in measurements were made at a frequency of 8 GHz in the temperature interval from –40 to 90°C. To obtain more extensive information, measurements in the ultrasonic region, at a frequency of 3 MHz, were also made. The results obtained will be discussed on the basis of existing theories of the propagation of waves in fluids.

2. System for examination of the fine structure components

Fig. 1 shows the measuring system. The ligt source is a ruby laser with $\lambda = 6943$ Å; pulse power $M = 100$ M·W; time of duration $t = 3 \times 10^{-8}$ s. The half width of the emission line was smaller than 0.01 cm^{-1}. The laser light wa ocused by means of a lens of focal length $f = 5$ cm, in a vessel containing thef fluid to be tested. The vessel had a length $l = 200$ mm, and a diameter $d = 40$ cm. The purity of the fluids to be tested was determined by measurement of the coefficient of light refraction, density and viscosity. The measurement results are given in Table 1.

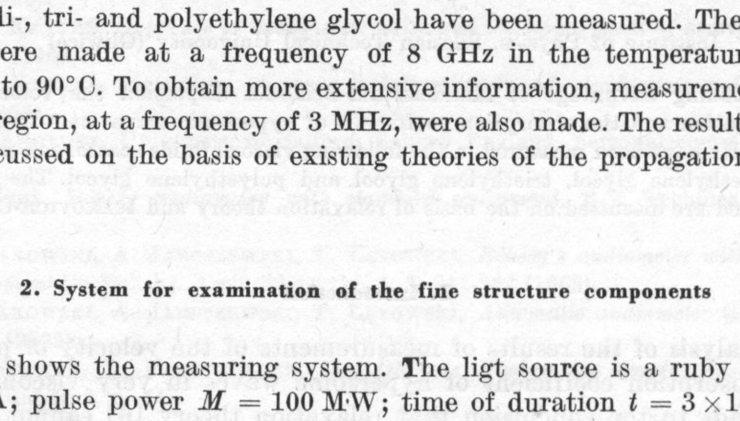

Fig. 1. Diagram of the Mandelsztam-Brillouin testing system

– ruby laser, *2* – vessel containing tested liquid, *3* – cooling, *4* – mat plate, *5* – Fabry-Perot interferometer, *6* – photographic film

The spectrum of scattered light was analyzed with the aid of a Fabry-Perot interferometer IT51-30. The distance between interferometer mirrors was 0.6 cm, and this corresponds to the scatter region $\delta\nu = 0.83\ \mathrm{cm}^{-1}$. A chamber with a focal length of $f = 2015$ mm produced an image on the photographic

Table 1. Density, refraction coefficient and viscosity of tested fluids
(t = 20° C)

Substance	$\varrho\left[\frac{\text{g}}{\text{cm}^3}\right]$	n	η [poise]
Paraffin oil	0.807	1.4421	2.21
Castor oil	0.963	1.4490	8.85
Diethylene glycol	1.118	1.4475	0.63
Triethylene glycol	1.127	1.4578	0.83
Polyethylene glycol	1.203	1.4621	0.92

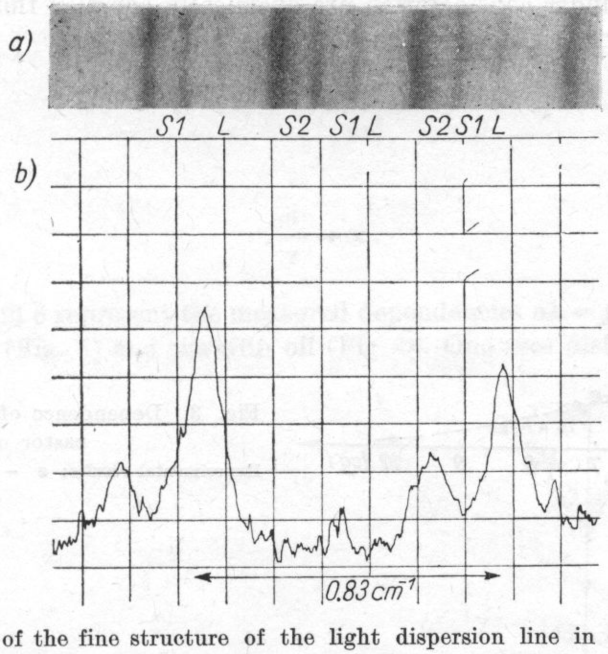

Fig. 2. Spectrum of the fine structure of the light dispersion line in triethylene glycol:
a) Fabry-Perot interferogram, b) microphotogram
L – laser radiation, $S1$, $S2$ – stokes components

plate. The linear dispersion was of the order 10^7. All measurements were made at the angle of 180°. The accuracy of measurement was approximately two seconds of arc. The photographs obtained were analyzed by means of a microphotometer. Fig. 2 shows an example of the lines of the fine structure obtained.

3. Results and discussion of the measurements

An analysis of the dependence of the absorption coefficient in the ultra-hypersonic region in castor and paraffin oil points to departures from a relaxation theory with one or two times of relaxation and a complexity of relaxation processes. Figs. 3 and 4 show the dependence of a/f^2 on $\log f$ for the above mentioned oils. Within the whole frequency region a square dependence of absorption coefficient is not observed. The curves of α/f^2 all have almost uniform slope, without points of inflexion which would have been indicative of a determined time of relaxation for a given mechanism. We should rather consider a superposition of a great number of relaxation processes, both of shear and volumetric viscosity over a wide range of frequencies. Evidence for the occurrence of volumetric viscosity relaxation is given by the fact that at frequencies above 10 MHz the total absorption coefficient for both oils is smaller than the value of this coefficient in stokes. The calculation of these relaxation times, as also the explanation of their molecular system is very difficult especially because the substances under consideration are chemically complex fluids that possess molecules of various sizes and shapes.

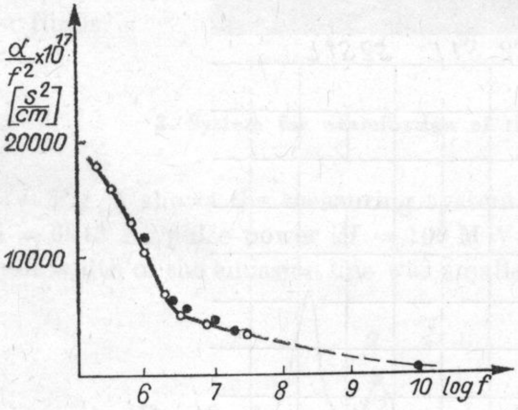

Fig. 3. Dependence of α/f^2 on $\log f$ for castor oil

Experimental results: ● – by author, ○ – [7]

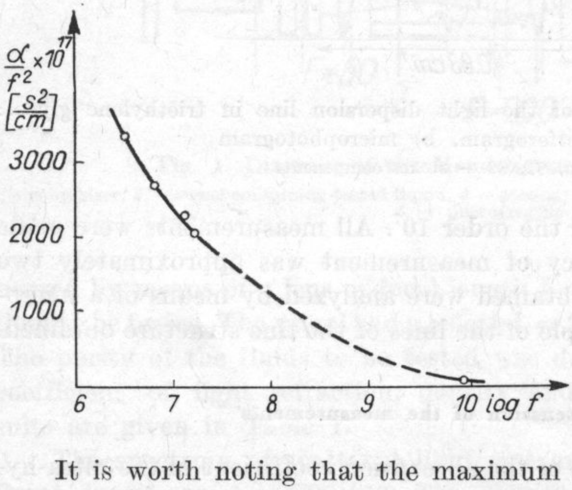

Fig. 4. Dependence of α/f^2 on $\log f$ for paraffin oil

It is worth noting that the maximum of the curve $\alpha\lambda$ as a function of τ should occur for $\tau = 1/\omega$, i.e. at a point at which the dependence of α/τ versus τ is reduced to a half of its maximum value. This is the result of the following considerations.

The value of α/τ — according to relaxation theory — is given by the formula

$$\frac{\alpha}{\tau} = \frac{\omega^2 (v_\infty^2 - v_0^2)}{(1 + \omega^2 \tau^2) \cdot 2 v_0^3}, \tag{3}$$

where τ is the time of the relaxation, v_0 and v_∞ the velocities of wave propagation at very small and infinitely large frequencies respectively.

Depending on the value of $\omega\tau$ the formula can take various forms:

for $\omega\tau \ll 1$

$$\frac{\alpha}{\tau} = \frac{\omega^2 (v_\infty^2 - v_0^2)}{2 v_0^3}, \tag{4}$$

for $\omega\tau = 1$

$$\frac{\alpha}{\tau} = \frac{\omega^2 (v_\infty^2 - v_0^2)}{4 v_0^3}, \tag{4a}$$

for $\omega\tau \gg 1$

$$\frac{\alpha}{\tau} \to \infty. \tag{4b}$$

Figs. 5 and 6 represent the measured dependencies $\alpha\lambda = f(\tau)$ and $\alpha/\tau = f(\tau)$ for castor oil (Fig. 5) and paraffin oil (Fig. 6). One sees distinctly the shift of the maximum of the curve towards higher values of τ for both fluids. At the same time it is assumed, as is often the case, that the time of relaxation is proportional to the viscosity and inversely proportional to temperature,

$$\tau = B \frac{\eta}{T}, \tag{5}$$

where B is a constant.

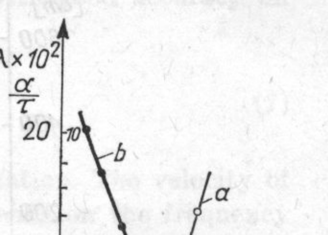

Fig. 5. Dependence of $\alpha\lambda$ (*a*) and α/τ (*b*) on $\log \eta/T$ for castor oil

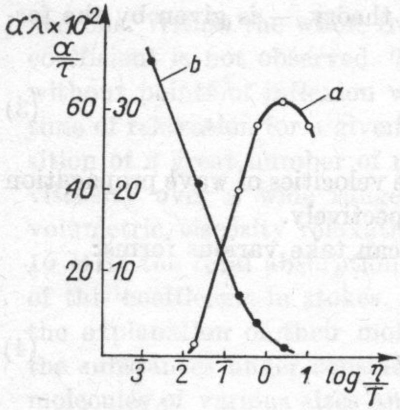

Fig. 6. Dependence of $a\lambda$ (*a*) and a/τ (*b*) on $\log \eta/T$ for paraffin oil

The frequency dependence of the absorption coefficient for triethylene glycol (Fig. 7) is similar to that for castor and paraffin oil. However, it should be noted that for frequencies $f < 15$ MHz the curve is very steep, and this may indicate the appearance in this area of relaxation process of duration $\tau = 10^{-7}$s or of several processes of very similar relaxation times. Above a frequency of 8 MHz, the total absorption coefficient is smaller than the values expressed in stokes, thus already at this frequency shear viscosity relaxation occurs.

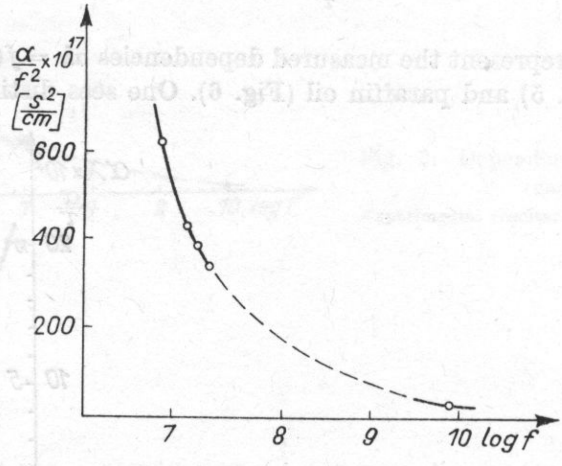

Fig. 7. Dependence of a/f^2 on $\log f$ for triethylene glycol

Velocity measurements as a function of temperature, both in ultra- and hypersonic regions, are very interesting for many reasons. The dependence of the velocity of ultrasonic waves on the temperature in paraffin oil ($f = 3$ MHz) is shown in Fig. 8. At the first sight it seems that this curve is a curve of the relaxation type. However, a thorough analysis of this dependence indicates that this conclusion is inaccurate.

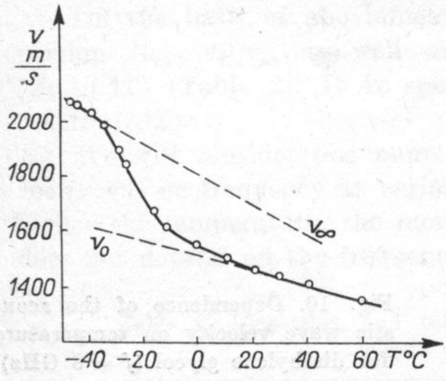

Fig. 8. Dependence of the acoustic wave velocity on temperature for paraffin oil ($f = 3$ MHz)

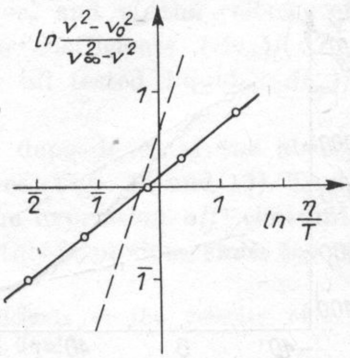

Fig. 9. Dependence of $\ln (v^2 - v_0^2)/(v_\infty^2 - v^2)$ on $\ln \eta/T$ for paraffin oil

The discussion will use the well-known formula of relaxation theory

$$\frac{v^2 - v_0^2}{v_\infty^2 - v^2} = \omega^2 \tau^2. \tag{6}$$

In Fig. 9 this dependence is shown by means of a broken line. Experimental data are presented by dots. They arrange themselves with good accuracy on the curve

$$\frac{v^2 - v_0^2}{v_\infty^2 - v^2} = \omega \tau, \tag{7}$$

but this does not result clearly from the theory of relaxation. The velocity of acoustic waves (as does the absorption coefficient) depends on the frequency and on the temperature. In fact, in all formulae there is the product $\omega\tau$. It has already been stated that $\tau \sim \eta/T$. Thus if, with the temperature change from -30°C, the viscosity would have changed by four orders, one can speak of very high and very low frequencies for this case. In Fig. 8, v_0 and v_∞ denote only extrapolated velocities for very large and very small values.

Figs. 10 and 11 show the dependence of the velocity of hypersonic waves as a function of temperature for diethylene and polyethylene glycol, and Figs. 12 and 13 the dependence of $\log(v^2 - v_0^2)/(v_\infty^2 - v^2)$ on $\log \eta/T$. When making hypersonic measurements by Brillouin scatter method, a change in temperature also (slightly) changes the frequency. However, because of very large changes in the value of τ, ω can be assumed constant to a good approximation. For these liquids too there occurs great divergence from the predictions of relaxation theory. Broken lines in Figs. 12 and 13 represent the dependence $(v^2 - v_0^2)/(v_\infty - v^2) = \omega^2\tau^2$ (relaxation theory). The experimental points, as for paraffin oil, arrange themselves on the straight line $(v^2 - v_0^2)(v_\infty^2 - v^2) = \omega\tau$.

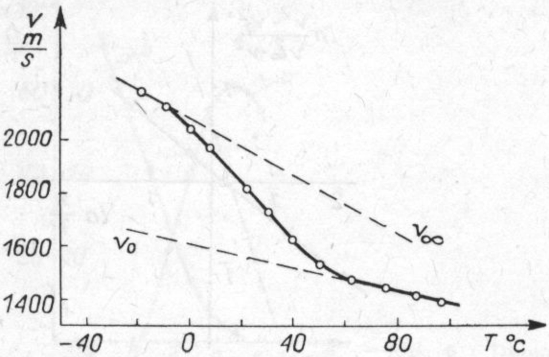

Fig. 10. Dependence of the acoustic wave velocity on temperature for diethylene glycol ($f = 8$ GHz)

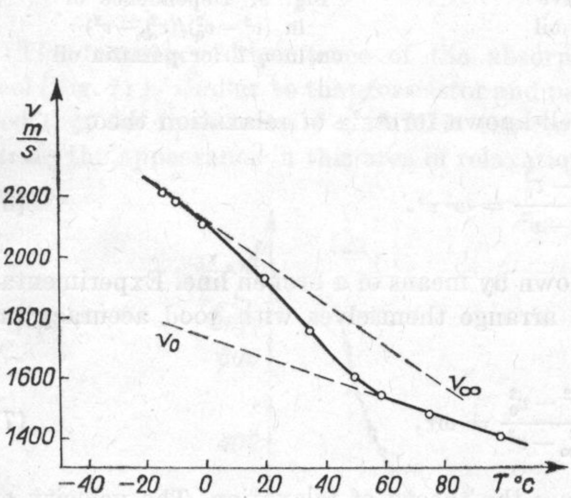

Fig. 11. Dependence of the acoustic wave velocity on temperature for polyethylene glycol ($f = 8$ GHz)

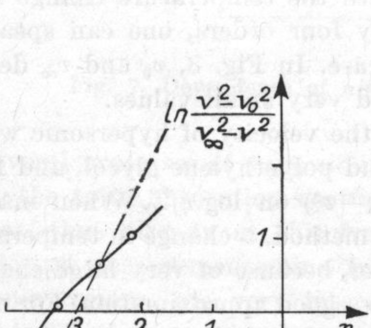

Fig. 12. Dependence of $\ln (v^2-v_0^2)/(v_\infty^2-v^2)$ on $\log \eta/T$ for diethylene glycol

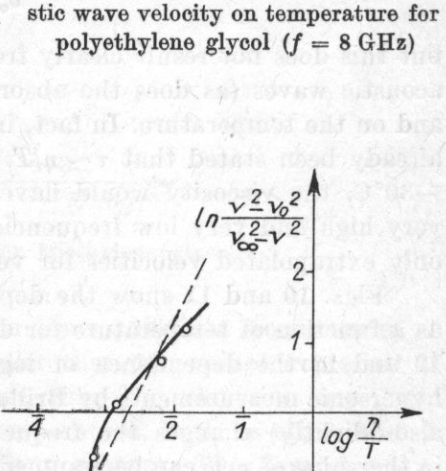

Fig. 13. Dependence of $\ln (v^2-v_0^2)/(v_\infty^2-v^2)$ on $\log \eta/T$ for polyethylene glycol

On the basis of above-mentioned values v_0 and v_∞ the velocity of dispersion $(v_\infty - v_0)/v_\infty$ as well as temperature coefficients $(\Delta v_\infty)/(\Delta T)$ and $(\Delta v_0)/(\Delta T)$ (Table 2). It is specific that for all tested liquids $(\Delta v_\infty)/(\Delta T) > (\Delta v_0)/(\Delta T)$.

We will consider one more form for the dependence of the absorption coefficient on frequency at various temperatures (Figs. 14 and 15). Thus, the higher the temperature, the more nearly is the expression α/f^2 constant and does not depend on the frequency. However, this is obvious since the higher

Table 2. Dispersion and temperature coefficients of the velocity of acoustic waves for tested liquids

Substance	$\frac{v_\infty - v_0^2}{v_0}$	$\frac{\Delta v_\infty}{\Delta T}\left[\frac{\text{m}}{\text{s deg}}\right]$	$\frac{\Delta v_0}{\Delta T}\left[\frac{\text{m}}{\text{s deg}}\right]$
Paraffin oil	0.28	−5.4	−2.1
Diethylene glycol	0.26	−5.8	−2.3
Polyethylene glycol	0.19	−7.0	−3.3

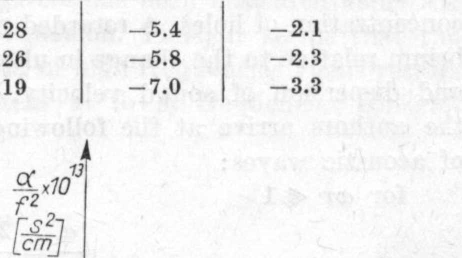

Fig. 14. Dependence of α/f^2 on frequency for triethylene glycol at various temperatures

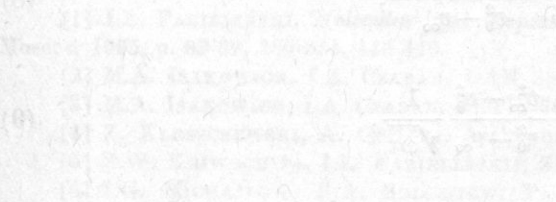

Fig. 15. Dependence of α/f^2 on frequency for paraffin oil at various temperatures

the temperature, the less viscous the fluid becomes. It can therefore be expected that the formulae of relaxation theory will be satisfied since the latter predicts that α/f^2 does not depend on frequency.

At present the most authoritative theory of acoustic wave propagation in fluids of high viscosity is regarded to be the theory developed by **Isakovich** and **Czaban** [2, 3], also called non-local theory.

At the root of this theory lies the assumption that a viscous fluid is an inhomogeneous two-phase medium. Each of the phases of this medium has, apart from normal thermodynamic parameters, two more parameters which determine the concentration of holes. At equilibrium the concentration for both phases is equal. The motion of an acoustic wave causes disturbance of the medium, and thus also the change of parameters that determine the concentration of holes. The return to equilibrium is effected by a diffusion of holes, the time of which depends on the diffusion coefficient and the size of the phase with the smaller concentration of holes. A retarded return of the medium to the state of equilibrium relative to the change in ultrasonic wave pressure causes the absorption and dispersion of sound velocity. Having justified the above assumptions, the authors arrive at the following expression for the absorption coefficient of acoustic waves:

for $\omega\tau \ll 1$

$$\frac{\alpha}{\omega} = \frac{2}{5}\,\frac{\omega\tau\,(v_\infty^2 - v_0^2)}{v_0^2 - v_\infty}; \tag{8}$$

for $\omega\tau \gg 1$

$$\frac{\alpha}{\omega} = \frac{3}{8}\,\frac{v_\infty^2 - v_0^2}{v_0^2 - v_\infty}\,\frac{1}{\sqrt{\omega\tau}}, \tag{9}$$

where

$$\tau = \frac{5}{3}\,\frac{v_\infty^2}{\varrho v_0^2}\,\frac{\eta + \frac{3}{4}\eta}{v_\infty^2 - v_0^2}.$$

It can be seen from formula (8) (for $\omega\tau \ll 1$) that this theory, like relaxation theory, assumes that α/f^2 is independent of frequency, but this does not agree with the experimental results obtained.

For high frequencies, it is hard to analyze the dependence of the absorption coefficient on the frequency since with the aid of Mandelsztam-Brillouin scatter method only one experimental point is obtained. It is true that by changing the temperature, we can get a change of frequency but this is very small. It was thus possible to compare experimental data with formula (9) at only one frequency. Table 3 contains values of the absorption coefficient evaluated from formula (9) (column 4), and measured (column 5), for castor oil, paraffin oil and triethylene glycol. It can be seen that the agreement between experimental and theoretical values of the absorption coefficient is satisfactory.

Table 3. Theoretical and experimental values of absorption coefficient of hypersonic waves

Substance	$T[^\circ\mathrm{C}]$	$f\times10^{-9}[\mathrm{s}^{-1}]$	$\alpha\times10^{-3}$ (theory) $[\mathrm{cm}^{-1}]$	$\alpha\times10^{-3}$ (exp.) $[\mathrm{cm}^{-1}]$
Castor oil	20	8.20	5.3	24
Paraffin oil	20	7.71	24	20
Triethylene glycol	20	8.46	15	21

It can be concluded that the theory of Isakovich and Czaban assumes that the ratio $(v^2-v_0^2)/(v_\infty^2-v^2)$ is proportional to $\omega\tau$, and that this is in agreement with the experimental results described above. Similar conclusions can also be draw from reference [10] where the velocity of propagation of hypersonic waves in glyceril triacetate and propylene glycol has been measured using the Mandelsztam-Brillouin method of forced dispersion. Thus, it seems that the theory of Isakovich and Czaban in the region of high frequencies yields results in good agreement with experiment, whereas at lower frequencies considerable discrepancies occur.

References

[1] I.L. Fabieliński, *Molecular light dispersion* [in Russian], Publ. House «Nauka» Moscow 1965, p. 82-89, 285-324, 440-448.

[2] M.A. Isakowich, I.A. Czaban, DAN SSSR, **165**, 299 (1965).

[3] M.A. Isakowich, I.A. Czaban, ZETF, **50**, 1343 (1966).

[4] Z. Kleszczewski, A. Opilski, Archiwum Akustyki, **4**, 4 (1969).

[5] S.W. Kriwochiża, I.L. Fabielińskij, ZETF, **50**, 3 (1966).

[6] I.G. Michajłow, B.A. Sołowiew, P. Syrmikow, *Fundamentals of molecular acoustics* [in Russian], Publ. House «Nauka», Moscow, p. 235-282.

[7] I.G. Michajłow, L.I. Sawina, *Applications of molecular acoustics* [in Russian], MOPI, vol. **4** (1957), p. 85-93.

[8] M.S. Piesin, I.L. Fabieliński, DAN SSSR, **129**, 299 (1959).

[9] M.S. Piesin, I.L. Fabieliński, DAN SSSR, **135**, 114 (1960).

[10] I. Zlatin, S.W. Kriwochiża, I.L. Fabieliński, ZETF, **56**, 4, 1186 (1969).

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