

ACOUSTIC PROPERTIES OF SELECTED LIQUIDS OVER THE ULTRA- AND HYPERSONIC RANGES**MIKOŁAJ ŁABOWSKI**

Acoustics Department, UAM (60-769 Poznań, ul. Matejki 48/49)

The paper presents the results of the investigation of the ultra- and hypersonic properties of some selected liquids. It also analyses the temperature variation of the ratio $I_c/2I_{MB}$ in the fine structure of the Rayleigh line of scattered light. On the basis of the results of the experimental investigation it is shown that the following relaxation processes occur in the liquids investigated: rotational isomerism in *n*-hexane and iso-octane; structural relaxation in isobutyric acid and vibrational relaxation in *n*-heptane. It has been observed that no acoustic dispersion occurs in nitroethane below a frequency of ~ 4 GHz.

1. Introduction

Numerous association processes and the formation of complexes by hydrogen bonds or other weak chemical interactions, the rotation of isomers, and also the excitation and inactivation of intramolecular vibrations, occur in the liquid phase with characteristic times of the order of 10^{-6} to 10^{-12} s [1]. Such fast reactions have recently been successfully investigated, using acoustic spectroscopy methods which give information on the dispersion of velocity and absorption of ultra- and hypersonic waves propagating in liquid media. These are relaxation methods.

The condition for an acoustic method to be used in the investigation of the relaxation process mechanisms in liquid media is that the acoustic waves used in the investigations should have periods T close to the relaxation time, τ , of the process investigated. Thus the investigation of the nature of different relaxation processes having different relaxation times, should be performed over a wide frequency range.

The information obtained on a given relaxation process becomes the fuller, the broader the frequency spectrum of the ultrasonic waves used in the investigations becomes.

Very often only one normal reaction can be observed to occur in a system, and the spectrum of acoustic relaxation times is reduced to one value of τ [1]

$$\frac{\alpha}{f^2} = \frac{A}{1 + (\omega\tau)} + B \quad (1)$$

and

$$\varepsilon = \frac{Av_0}{2\pi\tau} \quad (2)$$

where A is a parameter depending on the equilibrium characteristics.

The constants A and B are determined from the experimental shape of the dispersion curve, expressing the dependence of the quantity α/f^2 on $\log f$.

The following method is usually used for determination of the value of the quantities A , B , the relaxation time τ and the relaxation force ε . Using the quantity α/f^2 , we check if for some frequencies in the dispersion range the frequency dependence of the quantity α/f^2 is, within the errors of measurement and calculation, described by expression (1). The values of A , B , and τ are then determined. Using the values found for A , B , τ and ε , the values of the relaxation force ε is calculated from formula (2).

The quantities A , B , τ , and ε are characteristic of relaxation processes occurring in liquids. These processes include for example: vibrational relaxation, structural relaxation, and rotational isomerism.

By investigation of the temperature dependence of the quantities A , ε , η_v/η_s , it is possible to identify the type of process involved [1], since these quantities decrease as temperature increases in the case of rotational isomerism, and increase in the case of vibrational relaxation. In the case of structural relaxation, the quantities A and ε decrease, while η_v/η_s is practically independent of temperature.

The velocity and absorption coefficient of the acoustic waves were measured at three independent measuring positions at frequencies from 20 MHz to 150 MHz, and 300 MHz to 1000 MHz, using suitable pulse methods involving resonance and non-resonance excitation of piezoelectric transducers, and also at hypersonic frequencies, from the displacement and half power width of the Mandelshtam-Brillouin components in the fine structure of the Rayleigh lines scattered light.

The propagation velocity and absorption coefficient of hypersonic waves, and the $I_c/2I_{MB}$ ratio were determined at different temperatures from photo-electrical recordings of the fine structure of Rayleigh light scattering. The liquids used in the investigation were additionally cleaned chemically and distilled. The degree of purity was controlled through measurements of refractive index and density.

An exact description of the measuring apparatus and methodology is given elsewhere [2, 18].

The values of α_{kl}/f^2 , the coefficient of volume viscosity, and the coefficient of adiabatic compressibility were calculated from the formulae:

$$\frac{\alpha_{kl}}{f^2} = \frac{8}{3} \frac{\pi^2 \eta_s}{\rho v_0^3}, \quad (3)$$

$$\eta_v = \frac{A \rho v_0^3}{2\pi^2}, \quad (4)$$

$$\beta = \frac{1}{\rho v^2}, \quad (5)$$

where η_s is the coefficient of shear viscosity, and ρ is density. A maximum absorption on the wavelength, μ_{\max} , at a frequency $f = f_m$ was found from the expression

$$\mu_{\max} = \frac{A v f_m}{2}. \quad (6)$$

The ratio of the total intensity of the central line to the total intensity of the Mandelshtam-Brillouin lines was determined for all the media investigated according to the formula (given in [3])

$$\left(\frac{I_c}{2I_{MB}} \right)_{\text{total}} = \left(\frac{I_c}{2I_{MB}} \right)_{\text{max}} \frac{\delta\nu_c}{\delta\nu_{MB}}, \quad (7)$$

where $\delta\nu$ and $\delta\nu_{MB}$ are the half power widths of the central and lateral components.

The results are presented below of the acoustic investigation of the following selected pure liquids: isobutyric acid, *n*-heptane, *n*-hexane, iso-octane, nitroethane in both ultrasonic and hypersonic ranges.

The selection of the different objects for the investigation was caused, among other things, by the fact that they are components of the critical mixtures [19]. In addition, an attempt was made to show that the thermodynamics of irreversible processes, as developed by MASON [1], provides a sufficiently universal method and which can be used for the explanation of the molecular mechanisms of reactions occurring in different liquids.

2. Measurement results and their interpretation

2.1. Isobutyric acid. The acoustic properties of isobutyric acid were investigated in both ultra- and hypersonic ranges. As an illustration, Fig. 1 shows the original recordings of the fine structure of the Rayleigh line of light scattered at an angle $\theta = 90^\circ$ in isobutyric acid at selected temperatures. The values of $\Delta\nu$, f , $I_c/2I_{MB}$ are shown at the side of each recording, while the dependence of $I_c/2I_{MB}$ on temperature is plotted in Fig. 2.

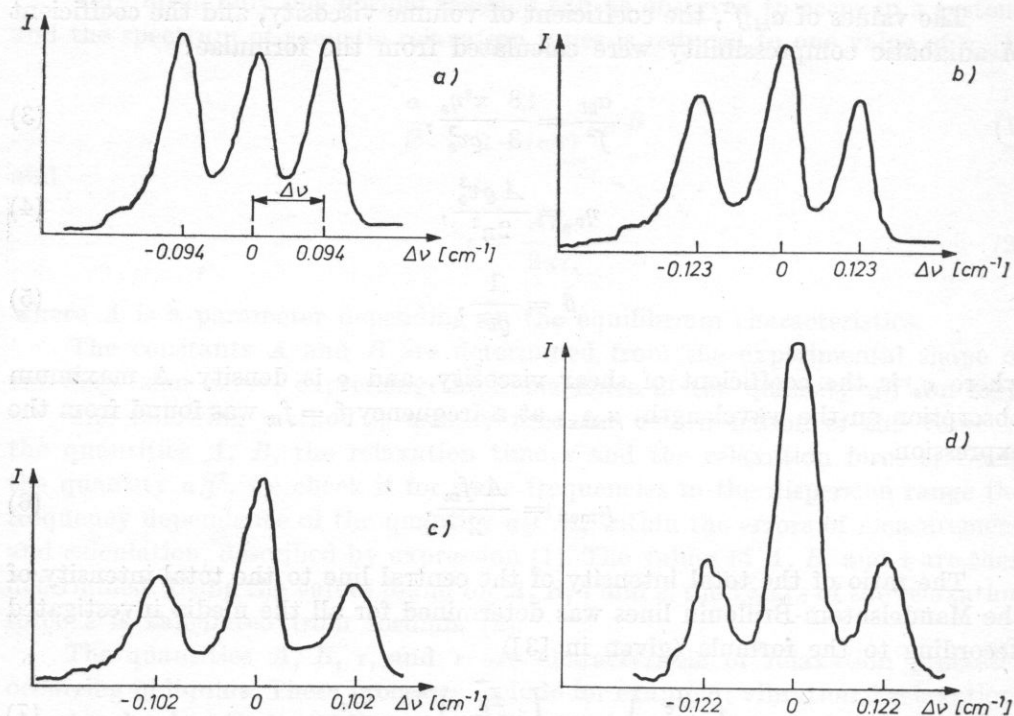


Fig. 1. Variation behaviour in the original recordings of the fine structure of the Rayleigh line of light scattered at an angle $\theta = 90^\circ$ for isobutyric acid at different temperatures

- a) $t = 75^\circ\text{C}$, $\Delta\nu = 0.094 \text{ cm}^{-1}$, $f = 2.8\text{GHz}$, $I_c/2I_{MB} = 0.459$;
 b) $t = 45^\circ\text{C}$, $\Delta\nu = 0.123 \text{ cm}^{-1}$, $f = 3.69\text{GHz}$, $I_c/2I_{MB} = 0.686$;
 c) $t = 25^\circ\text{C}$, $\Delta\nu = 0.102 \text{ cm}^{-1}$, $f = 3.07\text{GHz}$, $I_c/2I_{MB} = 0.982$;
 d) $t = 18^\circ\text{C}$, $\Delta\nu = 0.122 \text{ cm}^{-1}$, $f = 3.68\text{GHz}$, $I_c/2I_{MB} = 1.220$.

The results obtained for the absorption coefficient of the ultrasonic waves of varying frequencies at temperatures of 10° , 20° , 30° and 40°C are shown graphically in Figs. 3 and 4.

It can be seen from Figs. 3 and 4 that acoustical relaxation occurs within the investigated region of frequencies and temperatures. It can also be seen from the character of the curves for the dependence of the quantity a/f^2 on $\log f$, that this is the second range of acoustic relaxation for isobutyric acid.

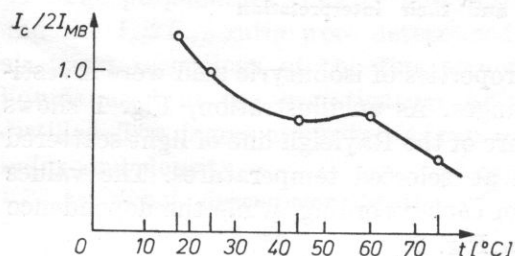


Fig. 2. Dependence of $I_c/2I_{MB}$ on temperature for isobutyric acid

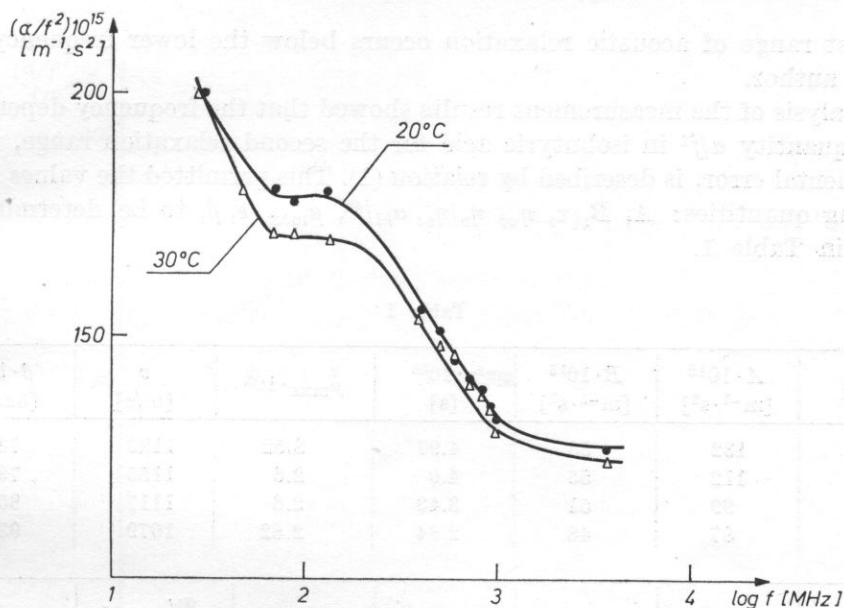


Fig. 3. Dependence of α/f^2 on $\log f$ for isobutyric acid at temperatures of 20°C and 30°C

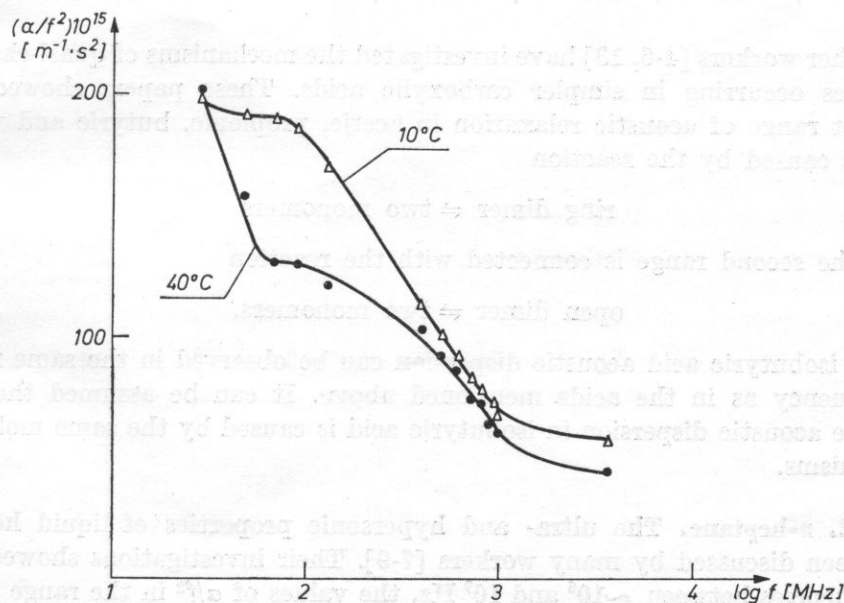


Fig. 4. Dependence of α/f^2 on $\log f$ for isobutyric acid at temperatures of 10°C and 40°C

The first range of acoustic relaxation occurs below the lower frequency used by the author.

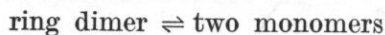
Analysis of the measurement results showed that the frequency dependence of the quantity α/f^2 in isobutyric acid for the second relaxation range, within experimental error, is described by relation (1). This permitted the values of the following quantities: A , B , τ , η_v , η_v/η_s , α_{kl}/f^2 , μ_{\max} , ε , β , to be determined as shown in Table 1.

Table 1

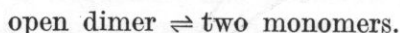
t [°C]	$A \cdot 10^{15}$ [m ⁻¹ ·s ²]	$B \cdot 10^{15}$ [m ⁻¹ ·s ²]	$\tau \cdot 10^{10}$ [s]	$\mu_{\max} \cdot 10^2$	v [m/s]	$\beta \cdot 10^6$ [bar ⁻¹]
10	132	58	4.97	2.52	1193	73.6
20	112	55	4.0	2.6	1155	79.5
30	99	51	3.43	2.6	1117	85.5
40	87	48	2.84	2.62	1079	93.0

t [°C]	$\varepsilon \cdot 10^2$	η_s [cP]	η_v [cP]	η_v/η_s	$\frac{\alpha_{kl}}{f^2} \cdot 10^{15}$ [m ⁻¹ ·s ²]	ρ [g/cm ³]
10	1.62	1.568	11	7	25.2	0.9592
20	1.66	1.318	8.35	6.5	23.8	0.9504
30	1.66	1.129	6.6	5.85	22.7	0.9414
40	1.67	0.98	5.2	5.3	21.9	0.9325

Other workers [4-6, 13] have investigated the mechanisms of quasi-chemical processes occurring in simpler carboxylic acids. These papers showed that the first range of acoustic relaxation in acetic, propionic, butyric and valeric acids is caused by the reaction



while the second range is connected with the reaction



In isobutyric acid acoustic dispersion can be observed in the same ranges of frequency as in the acids mentioned above. It can be assumed therefore that the acoustic dispersion in isobutyric acid is caused by the same molecular mechanisms.

2.2. *n*-heptane. The ultra- and hypersonic properties of liquid heptane have been discussed by many workers [7-9]. Their investigations showed that at frequencies between $\sim 10^6$ and 10^9 Hz, the values of α/f^2 in the range of the measurement error do not depend, within experimental error, on the frequency, and exceed the values of the quantity α_{kl}/f^2 by more than a factor of five.

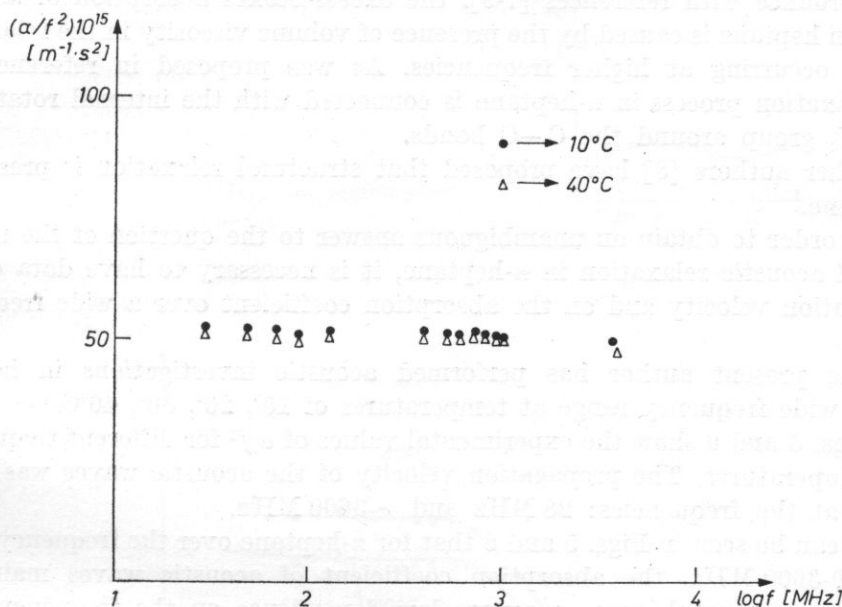


Fig. 5. Dependence of α/f^2 on $\log f$ for *n*-heptane at temperatures of 10°C and 40°C

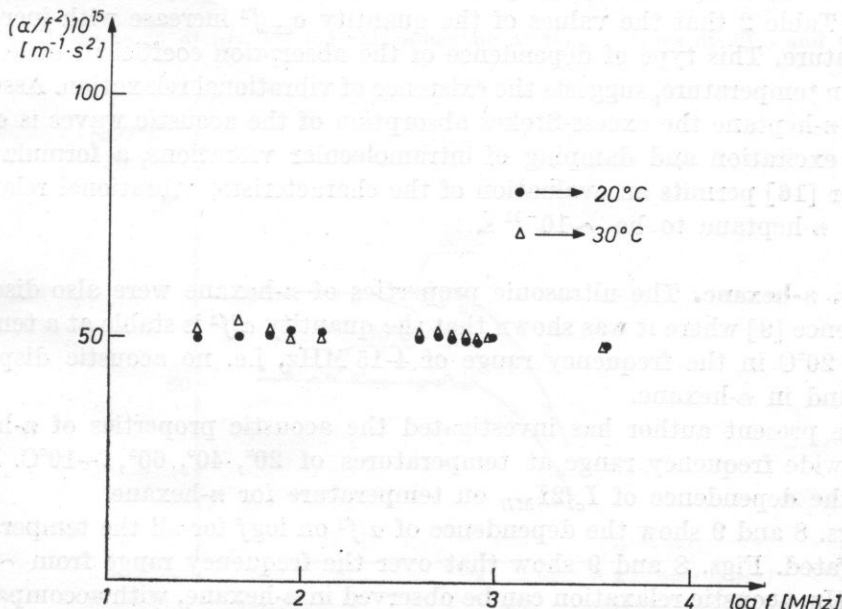


Fig. 6. Dependence of α/f^2 on $\log f$ for *n*-heptane at temperatures of 20°C and 30°C

In accordance with references [7-9], the excess-Stokes absorption of acoustic waves in heptane is caused by the presence of volume viscosity in the relaxation process occurring at higher frequencies. As was proposed in reference [7], the relaxation process in *n*-heptane is connected with the internal rotation of the CH₃ group around the C—C bonds.

Other authors [8] have proposed that structural relaxation is present in *n*-heptane.

In order to obtain an unambiguous answer to the question of the mechanism of acoustic relaxation in *n*-heptane, it is necessary to have data on the propagation velocity and on the absorption coefficient over a wide frequency range,

The present author has performed acoustic investigations in heptane over a wide frequency range at temperatures of 10°, 20°, 30°, 40°C.

Figs. 5 and 6 show the experimental values of α/f^2 for different frequencies and temperatures. The propagation velocity of the acoustic waves was determined at the frequencies: 28 MHz and ~ 3600 MHz.

It can be seen in Figs. 5 and 6 that for *n*-heptane over the frequency range of ~ 20 -3600 MHz, the absorption coefficient of acoustic waves maintains, within experimental error, a square law dependence on the frequency.

The experimental values of α_{ex}/f^2 , v_0 , α_{kl}/f^2 , η_s , η_v , η_v/η_s , $\alpha_{\text{ex}}/\alpha_{kl}$, and also the values of C_p , C_v and the total specific heat of vibration C_{opt}^* , are shown in Table 2.

The values of C_{opt} are found from the data on the normal frequencies of vibration of the molecules, using the Planck-Einstein formula [1]. It can be seen in Table 2 that the values of the quantity α_{ex}/f^2 increase with increasing temperature. This type of dependence of the absorption coefficient of acoustic waves on temperature, suggests the existence of vibrational relaxation. Assuming that in *n*-heptane the excess-Stokes absorption of the acoustic waves is caused by the excitation and damping of intramolecular vibrations, a formula given in paper [16] permits an evaluation of the characteristic vibrational relaxation time in *n*-heptane to be $\sim 10^{-11}$ s.

2.3. *n*-hexane. The ultrasonic properties of *n*-hexane were also discussed in reference [9] where it was shown that the quantity α/f^2 is stable at a temperature of 20°C in the frequency range of 4-15 MHz, i.e. no acoustic dispersion was found in *n*-hexane.

The present author has investigated the acoustic properties of *n*-hexane over a wide frequency range at temperatures of 20°, 40°, 60°, -10°C. Fig. 7 shows the dependence of $I_c/2I_{MB}$ on temperature for *n*-hexane.

Figs. 8 and 9 show the dependence of α/f^2 on $\log f$ for all the temperatures investigated. Figs. 8 and 9 show that over the frequency range from ~ 20 to 3500 MHz, acoustic relaxation can be observed in *n*-hexane, with accompanying weak dispersion of the wave propagation velocity: ~ 0.012 . The value of

Fig. 7. Dependence of $I_c/2I_{MB}$ on temperature for *n*-hexane

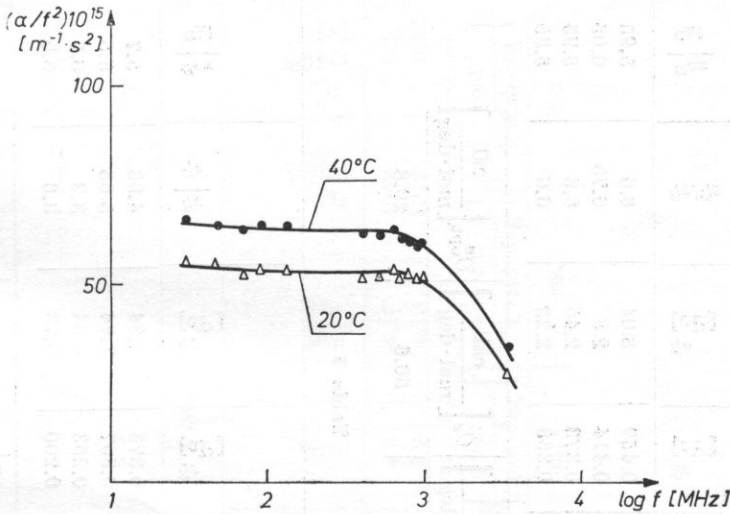
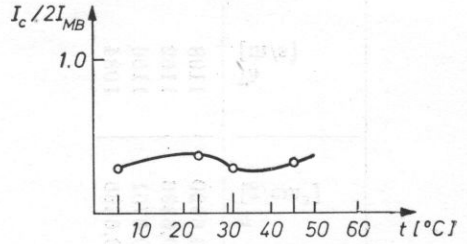


Fig. 8. Dependence of α/f^2 on $\log f$ for *n*-hexane at temperatures of 20°C and 40°C

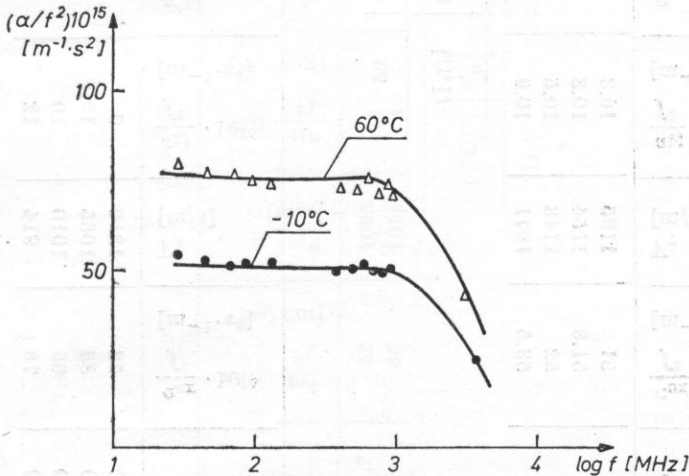


Fig. 9. Dependence of α/f^2 on $\log f$ for *n*-hexane at temperatures of 60°C and -10°C

Table 2

t [°C]	$\frac{f}{x_{ep}} \cdot 10^{15}$	$[z^s \cdot I - w]$	$\frac{f}{v} \cdot 10^{15}$	$[z^s \cdot I - w]$	$\frac{f}{v} \cdot 10^{15}$	$[D^s]$	$\frac{f}{v} \cdot 10^{15}$	$[D^s]$	$\frac{f}{v} \cdot 10^{15}$	s_{μ}/a_{μ}	$\frac{17D}{x_{ep}}$	$[\frac{z^s w \partial}{\partial}]$	$[\frac{s}{w}]$ φ_0
10	51	10.3	40.7	0.459	3.04	6.6	5.95	0.6920	1198				
20	51.8	10.3	41.5	0.414	2.8	6.75	6.05	0.6836	1162				
30	52	10.5	41.5	0.373	2.45	6.6	5.95	0.6751	1156				
40	53.5	10.9	42.6	0.338	2.22	6.6	5.95	0.6665	1084				

t [°C]	C_p $[\frac{\text{cal}}{\text{mol} \cdot \text{deg}}]$	C_v $[\frac{\text{cal}}{\text{mol} \cdot \text{deg}}]$	C_{opt}^* $[\frac{\text{cal}}{\text{mol} \cdot \text{deg}}]$
25	53.4	50.6	29.8

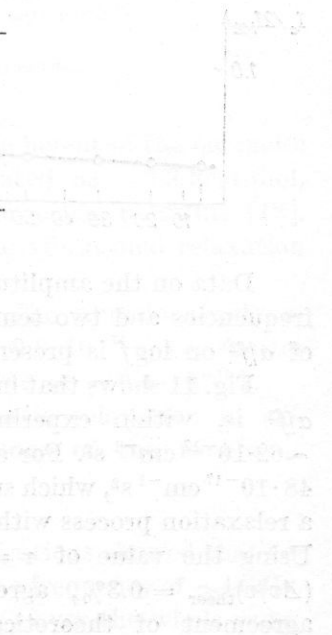
Table 3

$[C_0] t$	$\frac{f}{x_{ep}} \cdot 10^{15}$	$[z^s \cdot I - w]$	$\frac{f}{v} \cdot 10^{15}$	$[D^s]$	$\frac{f}{v} \cdot 10^{15}$	$[D^s]$	$\frac{f}{v} \cdot 10^{15}$	$[D^s]$	$\frac{f}{v} \cdot 10^{15}$	s_{μ}/a_{μ}	$\frac{17D}{x_{ep}}$	$[\frac{s}{w}]$ φ_0	$[\frac{s}{w}]$ $\varphi_0 \cdot 10^{1.2}$
-10	52	9	43	0.373	1.84	4.93	5.7	1220	0.81				
20	55	10.1	44.9	0.307	1.74	5.65	5.5	1078	0.80				
40	65	10	55	0.253	0.84	3.3	6.5	1009	0.76				
60	78	12	66	0.200	0.73	3.6	6.5	919	0.69				

t [°C]	C_p $[\frac{\text{cal}}{\text{mol} \cdot \text{deg}}]$	C_v $[\frac{\text{cal}}{\text{mol} \cdot \text{deg}}]$	C^* $[\frac{\text{cal}}{\text{mol} \cdot \text{deg}}]$
20	46.3	35.8	24.6

Table 4

t [°C]	$\frac{f_z}{\rho_{ex}} \cdot 10^{12}$	$[\omega_{-1} \cdot s^2]$	Λ^0 [s/w]	$\frac{f_z}{\rho_{ex}} \cdot 10^{12}$	$[\omega_{-1} \cdot s^2]$	$\frac{f_z}{\rho} \cdot 10^{12}$	$[\omega_{-1} \cdot s^2]$	$\frac{v_a}{v_l}$	$\frac{\rho_{ex}}{\rho}$	$\frac{v_a}{v_l}$	$\frac{v_a}{v_l}$	$\frac{v_a}{v_l} \cdot 10^2$	$\frac{v_a}{v_l} \cdot 10^2$
1.7	62		1053	15		47		4.3	4.13			10.7	6.9
40	66		1014	16		50		3.9	4.15			10.3	6.4
t [°C]	C_p [cal/mol·deg]	C_v [cal/mol·deg]	C_p^0 [cal/mol·deg]	C_p^0 [cal/mol·deg]	C_v^* [cal/mol·deg]	C_p^0 [cal/mol·deg]	C_v^* [cal/mol·deg]						
31.7	57.6	45.9	46.7	38.2									



$(\Delta v/v)_{\text{theor}} \sim 0.0101$, which agrees with the experimental value, within experimental error. Thus the relaxation process observed can be described with one value of τ .

The acoustic parameters are given in Table 3.

According to previous investigations [10], associated complexes do not exist in *n*-hexane at the expense of hydrogen bonds.

In the liquid and gaseous states, *n*-hexane takes the form of a mixture of rotational isomers. Answering the question as to which of the possible rotational isomers are actually present in the medium investigated, involves considerable experimental difficulties.

Calculations made using the Herzfeld formula given in reference [17] showed that the vibrational relaxation in *n*-hexane has a time of $\sim 0.6 \cdot 10^{-11}$ s, i.e. it is shorter by an order of magnitude than the experimentally found value of $\tau = 0.8 \cdot 10^{-10}$ s. Thus the relaxation process observed in *n*-hexane in the frequency range of $\sim 10^7$ - $3.5 \cdot 10^9$ Hz and in the temperature range investigated is, in all probability, caused by rotational-isomeric changes in the molecules of *n*-hexane.

Using the values of τ , the height of the energetic barrier between the rotational configurations was evaluated as ~ 3.5 kcal/mol.

2.4. Iso-octane. The acoustic properties of iso-octane were investigated over the frequency range from ~ 20 - 3400 MHz by pulse and optical methods. Fig. 10 shows the dependence of $I_c/2I_{MB}$ on temperature.

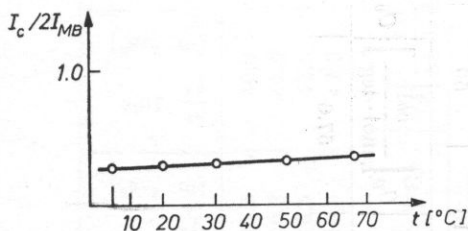


Fig. 10. Dependence of $I_c/2I_{MB}$ on temperature for iso-octane

Data on the amplitude absorption coefficient of acoustic waves at different frequencies and two temperatures are shown in Fig. 11 where the dependence of a/f^2 on $\log f$ is presented graphically.

Fig. 11 shows that in iso-octane over the frequency range of ~ 20 - 1000 MHz, a/f^2 is, within experimental error, independent of the frequency, being $\sim 62 \cdot 10^{-17}$ $\text{cm}^{-1} \text{s}^2$. For a hypersonic frequency of ~ 3.4 GHz a/f^2 decreases to $48 \cdot 10^{-17}$ $\text{cm}^{-1} \text{s}^2$, which suggests that in the frequency range of ~ 20 MHz- 3.4 GHz a relaxation process with a relaxation time of $\sim 0.4 \cdot 10^{-10}$ s occurs in iso-octane. Using the value of $\tau = 0.4 \cdot 10^{-10}$ s, $(\Delta v/v)_{\text{theor}}$ was calculated. The result, $(\Delta v/v)_{\text{theor}} = 0.3\%$, agrees with the observed value $(\Delta v/v)_{\text{ex}} = 0.2\%$. This agreement of theoretical and experimental values of $\Delta v/v$ suggests that

the acoustic relaxation observed in iso-octane over the investigated frequency range is characterized by one value of τ . All the parameters defined are shown in Table 4.

According to earlier investigations [11], associated complexes with vibrational relaxations occurring at frequencies higher than $\sim 10^{10}$ Hz do not occur in liquid iso-octane.

It can be seen in Table 4 that the value of η_v/η_s is not the same at both temperatures, since it is lower for the higher temperature. This permits the assumption that in iso-octane, as in other paraffins [12], the acoustic dispersion is connected with rotational-isomeric transformations of the iso-octane molecules.

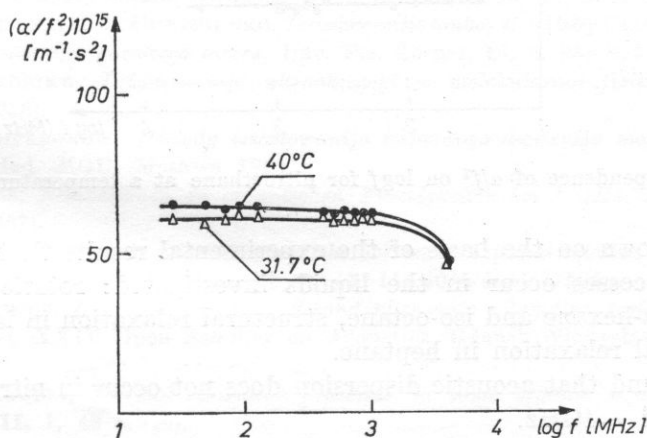


Fig. 11. Dependence of α/f^2 on $\log f$ for iso-octane

Then, using the experimentally found value of τ , the height of the energetic barrier between the two configurations can be evaluated as ~ 3.3 kcal/mol, which agrees with the values obtained by PIERCY for other paraffins [12].

On the other hand, it is interesting to evaluate the vibrational relaxation time.

The values of C_p , C_v , C_p^0 , C_{opt}^* are shown in Table 4. The vibrational relaxation time τ , obtained for iso-octane was found to be $\sim 0.6 \cdot 10^{-11}$ s, i.e. shorter by an order of magnitude than the experimental value of $\sim 0.4 \cdot 10^{-10}$ s.

This circumstance confirms the thesis that acoustic relaxation in iso-octane is caused by rotational-isomeric transformations of the iso-octane molecules.

2.5. Nitroethane. The ultra- and hypersonic investigations showed that acoustic dispersion does not occur in pure nitroethane below a frequency of ~ 4 GHz. Fig. 12 shows graphically the dependence of α/f^2 on $\log f$ over the whole range

of frequencies investigated, at a temperature $t = 31.7^\circ\text{C}$. It can be seen that within experimental error, the absorption coefficient of the acoustic waves is proportional to the square of the frequency, and the values of α/f^2 are relatively low. Fig. 13 shows the dependence of $I_c/2I_{MB}$ on temperature.

The values of A , B , τ , η_v , η_v/η_s , ε , β , μ_{\max} , v_0 , v_n , α_{ex}/f^2 , α_{kl}/f^2 , Δv , δv_{MB} , $I_c/2I_{MB}$, have been determined for all the liquids investigated.

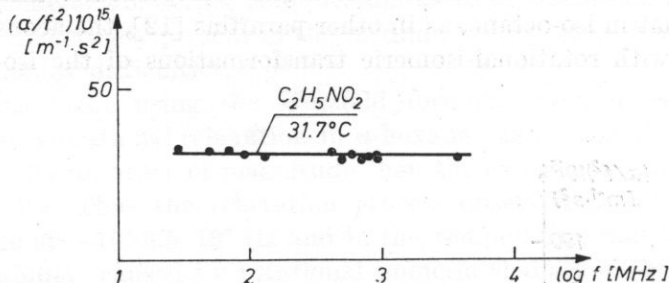


Fig. 12. Dependence of α/f^2 on $\log f$ for nitroethane at a temperature of 31.7°C

It was shown on the basis of the experimental results that the following relaxation processes occur in the liquids investigated: rotational isomerism relaxation in *n*-hexane and iso-octane, structural relaxation in isobutyric acid, and vibrational relaxation in heptane.

It was found that acoustic dispersion does not occur in nitroethane below a frequency of $\sim 4\text{GHz}$.

Recordings of the Mandelshtam-Brillouin spectra and measurements of the absorption coefficient and propagation velocity of ultrasonic waves were made in Prof. M. I. SHAKHPARONOV'S Solutions Laboratory, at the M. W. Lomonosov University in Moscow, in Dr. O. I. ZINOVYEV'S section, to whom the author wishes to express his gratitude.

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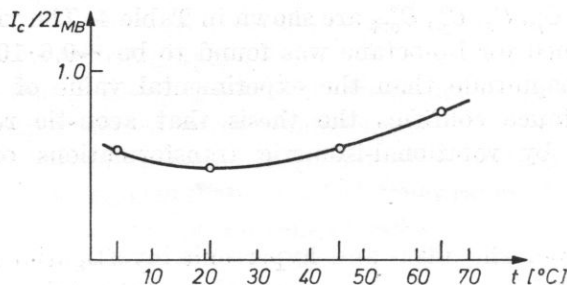


Fig. 13. Dependence of $I_c/2I_{MB}$ on temperature for nitroethane

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