Theory of Negative Dispersion of Sound Velocity in Liquid by Double Relaxation Model

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§ 1. Introduction

It has been shown that there are two different experimental results about the dispersion of sound velocity in some kinds of liquid. Benedek et al. show the existence of the negative dispersion of sound velocity[1], while Tiganov shows that the negative dispersion of sound velocity does not appear[2]. Like this, expermenital results obtained by some workers are often different from that obtained by the other. However, it is important to remember that the single relaxation theory which contains only a single slow internal process of relaxation has no solution of the negative dispersion of sound velocity. Recently it has been shown that the experimental values for the absorption of sound wave for some liquids (e.g. chloroform) can not be explained accurately by this single relaxation theory[6].

The aim of this paper is to calculate the dispersion of sound velocity and the absorption coefficient of sound wave using the double relaxation theory which contains two different processes of relaxation simultaneously, instead of the single relaxation theory.

In sec. 2 let us review shortly the experimental and theoretical situations of Rayleigh-Brillouin light scattering up to the present day. In sec. 3 we shall calculate the dispersion of sound velocity and the absorption coefficient of sound wave by the use of the double relaxation theory under some assumptions. Lastly we shall compare the results obtained with the experimental values given by some workers.

§ 2. The review of Rayleigh-Brillouin scattering

a) Intensity

In 1922 Brillouin predicted only a doublet (Brillouin doublet or Brillouin components

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)[3]. But the experimental observation by Gross in 1930, and subsequently by many others observed not only Brillouin components but also a central unshifted component in the spectrum of the scattered light[4]. In 1934 Landau and Placzek explained the origin of the unshifted central component that it comes from the local fluctuation of entropy which does not propagate in normal liquid, while the Brillouin doublet comes from the adiabatic (isentropic) pressure fluctuation (called sound wave). Landau and Placzek used the fluctuation theory in adiabatic and reversible process to calculate the $I_C/2I_B$ (I_C, I_B are total intensity of central component and Brillouin components respectively) and obtained the well-known relation $I_C/2I_B = \gamma - 1$[5]. But this ratio gives the greater value than the experimental value on account of many basic assumptions; i.e., pressure fluctuation is adiabatic and reversible, and the fluctuations of the thermodynamic quantities are independent on the frequency of their instantaneous change. In actual processes in liquid, we can not expect that the pressure fluctuation is reversible if the hypersonic attenuation exists, at the same time, many thermodynamic variables should exhibit the dependency on frequency of their fluctuation[7]. Moreover, if molecules have the internal degrees of freedom, non-adiabatic pressure fluctuation become effective[8]. In order to avoid the above mentioned limitation of thermodynamic consideration and to extend the theory applicable to the more general case, we can derive the intensity distribution from the microscopic point of view based on the statistical mechanical theory of the scattering of light. The intensity distribution is formulated by the correlation function of the molecular density in liquid which is in turn related with the kinetic character of molecules[9].

b) Frequency shift of Brillouin components

The origin of Brillouin components is explained as follows: In liquid, there exist thermally excited sound waves and the incident light couples with the one of them. On the scattering process of light, the conservation laws of momentum and energy must hold:

$$k = k' + q$$
$$\omega = \omega' \pm \Omega$$

here ω, ω' and Ω are the frequency of incident light, scattered light and sound wave, respectively. Then frequency shift of Brillouin components is given $\Omega = |\omega - \omega'|$.

The sign + corresponds to sound wave creation giving rise to Stokes line. The sign - corresponds to sound wave annihilation giving rise to anti-Stokes line. Brillouin scattering can be regarded as first order Raman scattering by acoustic wave, while usual Raman scattering in liquid is due to molecular vibrations the shift of which are much larger than the Brillouin shift.

The acoustic wave frequency $\Omega/2\pi \sim 10^9$ c.p.s., while the light frequency $\omega/2\pi \sim 10^{15}$ c.p.s., then $\omega'/\omega \sim 1 \pm 10^{-6} \approx 1$. The dispersion relation for photon is given by

$$\omega = c'|k| = \frac{c}{n} |k| \qquad (2.3)$$

here c', c and n are velocity of light in liquid, that of vacuum and the index of

refraction for an optically isotropic liquid, respectively.

Thus we conclude that

$$|k'| \sim |k| \quad \text{or} \quad |k-k| = 2|k| \; sin \frac{\theta}{2} \qquad (2.4)$$

here $\theta = k \wedge k'$. Eq. (2.2), Eq. (2.4) and the dispersion relation for phonon

$$\Omega = v \mid q \mid \qquad (2.5)$$

lead to the well-known results:

$$\Omega = 2n \; \omega \frac{v}{c} \; sin \frac{\theta}{2}$$

$$\text{or} \quad \omega - \omega' = \pm 2n \; \omega \; \frac{v}{c} \; sin \frac{\theta}{2} \qquad (2.5)$$

where v, velocity of sound wave, is the function of Ω. The form $v(\Omega)$ can be derived from the relaxation theory. Indeed, the relaxation theory with single relaxation time τ_s gives

$$\frac{v^2 - v_0^2}{v_\infty^2 - v_0^2} = \frac{\Omega^2 \; \tau_s^2}{1 + \Omega^2 \tau_s^2} \qquad (2.7)$$

Recently new experimental technique using injected phonons of well defined frequency has been developed. Using this new technique, the frequency dependence of phonon velocity is measured over wide range for which usual ultrasonic and Brillouin scattering measurements can be carried out[11].

c) The width of Rayleigh and Brillouin components

The origin of width of Rayleigh component is attributed to the decay of entropy fluctuation and that of Brillouin components is to the decay of phonons (decay of pressure fluctuation). Assuming the damping form of amplitude of pressure fluctuation as $e^{-\alpha x}$, the width of Brillouin components is given by

$$\delta \nu_{M.B.} = \frac{\alpha v}{\pi c}. \qquad (2.8)$$

From the hydrodynamic consideration which is applicable only for low frequency, total absorption coefficient of sound wave α is obtained as

$$\alpha = \frac{\Omega^2}{2 v_0^3} \left\{ \frac{4}{3} \eta + \eta' + \kappa \left(\frac{1}{C_v} - \frac{1}{C_p} \right) \right\} \equiv \alpha_\eta + \alpha_{\eta'} + \alpha_\kappa \qquad (2.9)$$

here v_0 is sound velocity at low frequency limit, η, η' are shear and bulk viscosity coefficient, which are assumed to frequency independent[13], α_η, $\alpha_{\eta'}$ and α_κ mean the absorption coefficients due to shear viscosity, bulk viscosity and thermal conduction, respectively. Eq. (2.9) is not applicable to the case where strong absorption exists, because we used the nondamping plane wave as a zeroth order basis on the way deriving this

formula. The same formula about the absorption coefficient is obtained by the study of the correlation function of the density fluctuation[12]. The extension of Eq. (2.9) was done by L. I. Mandel'shtam and M. I. Leontovich who considered the frequency dependence of bulk viscosity and calculated it according to the relaxation theory which they developed[13]. Their result about the frequency dependent of bulk viscosity is expressed as

$$\eta'(\Omega) = \tau_s \, \rho \frac{v_\infty^2 - v_0^2}{1 - i\Omega\tau_s} \qquad\qquad (2.10)$$

here ρ is density of liquid, τ_s is single relaxation time of some physical quantity, v_∞ is phase velocity of sound wave at high frequency limit. Then the absorption coefficient due to bulk viscosity is

$$\alpha_{\eta'} = \frac{\Omega^2 \, \tau_s \, \{v_\infty^2/v_0^2 - 1\}}{2v_0(1 + \Omega^2\tau_s^2)} \,. \qquad\qquad (2.11)$$

Recently R. D. Mountain calculated the relaxation time of shear viscosity. But its relaxation time is very small in usual liquid. We expect that this effect will be occur at only very high frequency[14]. R. M. Herman showed that the total absorption coefficient α vanishes in pure sound wave field in which κ, thermal coefficient of expansion, is equal to zero although η and η' do not necessarily vanish based on a quantum statistical theory in liquid[15].

As a matter of fact the accuracy of data published for the width of Brillouin components is not so high. The relative errors are about 20% in the experimental data using the laser source combined with the Fabri-Perot interferometer, the resolving power of this device $\nu/\Delta\nu$ does not exceed about 10^6. Recently optical heterodyne spectrometer has been developed where the resolving power $\nu/\Delta\nu \sim 10^{14}$. Experimental data with more accuracy will be expected to be published in future by these improved technique.

On the other hand the width of Rayleigh component Γ is obtained from the equation of thermal conduction $\rho C_p \dfrac{\partial T}{\partial t} = \kappa \Delta T$ and $T(r, t) \sim e^{-\Gamma t} e^{iqr}$ which is non-propagating temperature wave. The result is

$$\Gamma = \frac{\kappa q^2}{\rho C_p} \,. \qquad\qquad (2.12)$$

This formula is expected to describe well the actual experimental results so far as we can be confined with the low frequency region in considering the Rayleigh component. Recently J. B. Lastovka and G. B. Benedek tested the Eq. (2.12) using a laser light source and a high-sensitivity optical-heterodyne spectrometer. Their result was in good agreement with Eq. (2.12) for toluene.

§ 3. Double Relaxation Theory

The origins of the dispersion of sound velocity and the absorption of sound wave are attributed to many processes; e. g., relaxation processes associated with the energy transfer from translational to vibrational degrees of freedom, configurational relaxation or another some processes through the intermolecular potential. Although the question about which process is dominant is very difficult to be determined completely, the first process mentioned above seems to be dominant for some kinds of liquid, CH_2Cl_2 for example, because for this liquid Eq. (2.11) agrees with the experimental results very well. But for chloroform the deviation of Eq. (2.11) from the experimental results is observed[17]. Some other difficulties of relaxation theory with single relaxation time were pointed in sec. 1. These difficulties may be removed in somewhat degree, if we consider the relaxation process with multi-relaxation times. In general, however, the calculation with multi-relaxation times is very complicated and parameters which will appear can not be fully determined by the simple consideration. In the following, for simplicity, we are concerned with only two relaxation processes which arise from the time delay in the redistribution of energy between the vibrational and translational degrees of freedom of molecules. The relaxation times to be considered characterize the rate of establishment of the internal equilibrium corresponding to the instantaneous value of pressure. According to our model, the pressure of liquid in sound wave field now be regarded as a function of density ρ and the value of some physical quantities ξ_1, ξ_2 which characterize the macroscopic state of the liquid concerned and also of entropy S;

$$P = P\ (\rho,\ \xi_1,\ \xi_2,\ S) \tag{3. 1}$$

In the first order approximation, the rate equations are established as

$$\dot{\xi}_i = -\frac{\xi_i - \xi_{i0}}{\tau_i}\ (i=1,2) \tag{3. 2}$$

Here, ξ_{i0} is the equilibrium value of ξ_i which varies in time and determined by density and entropy. In this approximation pressure fluctuation is entropy independent because the first order derivative of entropy with respect to ρ and ξ_i is zero. We divide ξ'_{i0} and ξ'_i into two parts;

$$\xi_{i0} = \xi_{i00} + \xi'_{i0} \qquad \text{and} \qquad \xi_i = \xi_{i00} + \xi'_i$$

here, $\dot{\xi}_{i00} = 0$ and ξ'_{i0}, ξ'_i vary with time associated with the change of pressure. Let us assume that $\rho(t) \sim e^{-i\Omega t}$ Further, we assume the time dependence of ξ'_i, ξ'_{i0} as ξ'_i $(\sim \xi'_{i0}) \sim e^{-i\Omega t}$ corresponding to the density fluctuation. Substituting this time dependence of ξ^i and ξ_{i0} into Eq. (3.2), we obtain

$$\xi_i' = \frac{\xi_i'^0}{1 - i\Omega\tau_i} \tag{3.3}$$

Then, v_Ω^2 is written as

$$v_\Omega^2 = \frac{\partial P}{\partial \rho} = \left(\frac{\partial P}{\partial \rho}\right)_{\xi_1,\xi_2} + \left(\frac{\partial P}{\partial \xi_1}\right)_{\rho,\,\xi_2} \cdot \left(\frac{\partial \xi_1}{\partial \rho}\right) + \left(\frac{\partial P}{\partial \xi_2}\right)_{\rho,\,\xi_1} \cdot \left(\frac{\partial \xi_2}{\partial \delta}\right)$$

$$= \left(\frac{\partial P}{\partial \rho}\right)_{\xi_1,\xi_2} + \left(\frac{\partial P}{\partial \xi_1}\right)_{\rho,\,\xi_2}\left(\frac{\partial \xi_1}{\partial \xi_1'^{10}}\right)\left(\frac{\partial \xi_1'^{10}}{\partial \rho}\right) + \left(\frac{\partial P}{\partial \xi_2}\right)_{\rho,\,\xi_1}\left(\frac{\partial \xi_2}{\partial \xi_2'^{0}}\right)\left(\frac{\partial \xi_2'^{0}}{\partial \rho}\right)$$

$$= \left(\frac{\partial P}{\partial \rho}\right)_{\xi_1,\xi_2} + \frac{1}{1 - i\Omega\tau_1}\left(\frac{\partial P}{\partial \xi_1}\right)_{\rho,\,\xi_2}\left(\frac{\partial \xi_{10}}{\partial \rho}\right) + \frac{1}{1 - i\Omega\tau_2}\left(\frac{\partial P}{\partial \xi_2}\right)_{\rho,\,\xi_1}\left(\frac{\partial \xi_{20}}{\partial \rho}\right)$$

$$= v_\infty^2 + \frac{1}{1 - i\Omega\tau_1}\,a_1 + \frac{1}{1 - i\Omega\tau_2}\,a_2 \tag{3.4}$$

Here,

$$v_\infty^2 = \left(\frac{\partial P}{\partial \rho}\right)_{\xi_1,\xi_2}, \quad a_1 = \frac{\partial P}{\partial \xi_1}\frac{\partial \xi_{10}}{\partial \rho} = \frac{\partial P}{\partial \xi_1}\left(\frac{\partial \xi_1}{\partial \rho}\right)_{eq}$$

and

$$a_2 = \left(\frac{\partial P}{\partial \xi_2}\right)\frac{\partial \xi_{20}}{\partial \rho} = \frac{\partial P}{\partial \xi_2}\left(\frac{\partial \xi_2}{\partial \rho}\right)_{eq}$$

and $\left(\dfrac{\partial \xi_i}{\partial \rho}\right)_{eq}$ means the derivatives of ξ_i with respect to density for the process which is so slow that the i-th physical quantity remains in equilibrium with the external degree of freedom of molecules.

While

$$v_0^2 = \left(\frac{\partial P}{\partial \rho}\right)_{eq} = \left(\frac{\partial P}{\partial \rho}\right)_{\xi_1,\xi_2} + \left(\frac{\partial P}{\partial \xi_1}\right)_{\rho,\,\xi_2}\left(\frac{\partial \xi_1}{\partial \rho}\right)_{eq} + \left(\frac{\partial P}{\partial \xi_2}\right)_{\rho,\,\xi_1}\left(\frac{\partial \xi_2}{\partial \rho}\right)_{eq}$$

$$= v_\infty^2 + a_1 + a_2 \tag{3.5}$$

From Le Chatelier's principle, it is evident that

$$\left(\frac{\partial P}{\partial \rho}\right)_{\xi_1,\xi_2} > \left(\frac{\partial P}{\partial \rho}\right)_{eq} \qquad \text{or} \qquad v_\infty^2 > v_0^2.$$

The Eq.(3.4) is combined with Eq. (3.5) to give

$$v_\Omega^2 = \frac{1}{1 - \Omega^2\tau_1\tau_2 - i\Omega(\tau_1 + \tau_2)}\{v_0^2 - \Omega\tau_1\tau_2 v_\infty^2 - i\Omega(\tau_1 + \tau_2)v_\infty^2 - i\Omega(\tau_2 a_1 + \tau_1 a_2)\} \tag{3.6}$$

We may assume that $v_\infty < 1.1 v_0$ i.e., the greatest dispersion is 10% at most, then $v_\infty^2 - v_0^2 < 2.1 \times 10^9$ (cm/sec)^2 using the value $v_0 \sim 10^5$ cm/sec.

Moreover we assume $a_1 \sim a_2$, then $|a_1|$ and $|a_2| < 10^9 < v_\infty^2 \approx 10^{10}$ (cm/sec)^2 at hypersonic region. Thus we can omit $\tau_1 a_1$ and $\tau_2 a_2$ in Eq. (3.6) to get

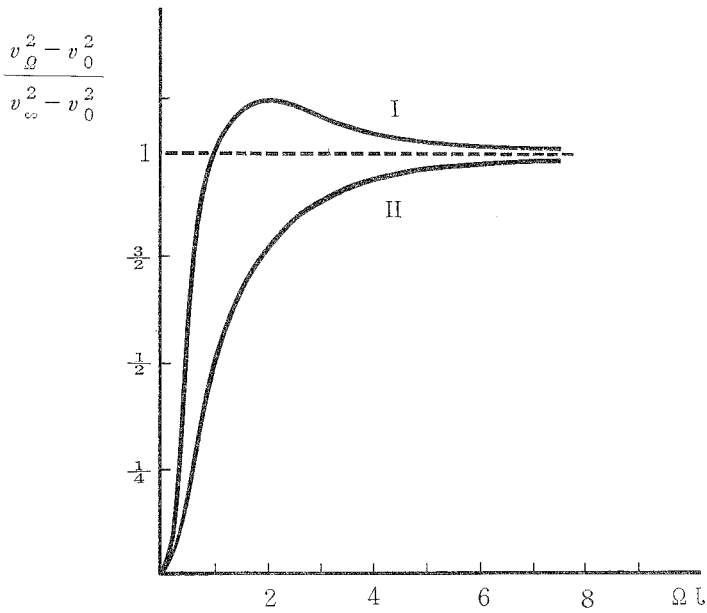$$\frac{v_\Omega^2 - v_0^2}{v_\infty^2 - v_0^2}$$

Fig. 1   The dispersion of sound velocity

I : The dispersion of sound volocity obtained from the double relaxation theory,
II : The dispersion of sound velocity obtained from the single relaxation theory,

$\triangle S / \triangle d$

○ : CH_2Cl_2
◎ : $C_6H_5CH_3$
▣ : $CHCl_3$
⊠ : CS_2
△ : C_6H_6
⊗ : CCl_4

$\Omega / 2\pi$

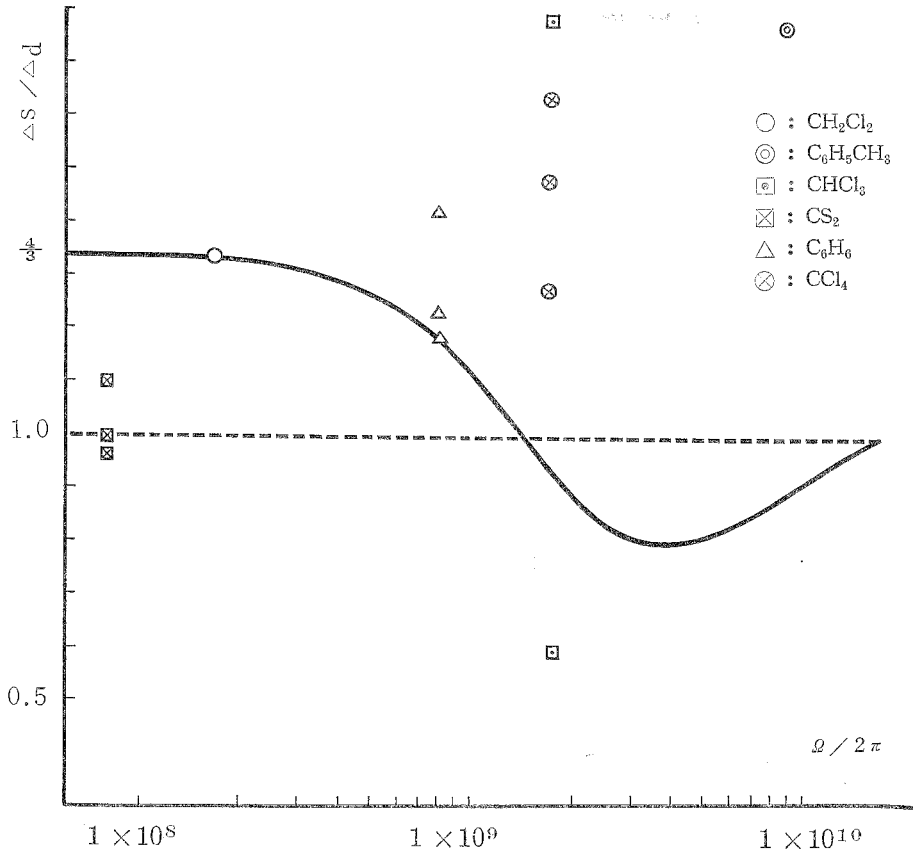1×10^8 1×10^9 1×10^{10}

Fig. 2   The solid curve represents Eq. (3. 12).
The dotted line shows the values given by (18)

$$v_\Omega^2 = \frac{v_0^2 - \Omega^2 \tau_1 \tau_2 v_\infty^2 - i\Omega(\tau_1 + \tau_2)v_\infty^2}{1 - \Omega^2 \tau_1 \tau_2 - i\Omega(\tau_1 + \tau_2)} \qquad (3.7)$$

Eq. (3.7) is transformed into

$$\frac{v_\Omega^2 - v_0^2}{v_\infty^2 - v_0^2} = -\frac{\Omega^2 \tau_1 \tau_2 - \Omega^4 \tau_1^2 \tau_2^2 - \Omega^2(\tau_1 + \tau_2)^2}{(1 - \Omega^2 \tau_1 \tau_2)^2 + \Omega^2(\tau_1 + \tau_2)^2} \qquad (3.8)$$

Let us assume $\tau_1 = \tau_2 \equiv \tau_d$ for simplicity, then Eq. (3.8) now leads to a simple form

$$\frac{v_\Omega^2 - v_0^2}{v_\infty^2 - v_0^2} = \frac{\Omega^2 \tau_d^2 (\Omega^2 \tau_d^2 + 3)}{(1 + \Omega^2 \tau_d^2)^2} \qquad (3.9a)$$

On the other hand single relaxation theory gives

$$\frac{v_\Omega^2 - v_0^2}{v_\infty^2 - v_0^2} = \frac{\Omega^2 \tau_s^2}{1 + \Omega^2 \tau_s^2}. \qquad (3.9b)$$

The curves of (3.9b) and (3.9a) versus $\Omega\tau$ are presented in Fig. 1.

According to this curves it is seen that in some frequency range $v_{\Omega'} < v_\Omega$ (for $\Omega' > \Omega$) holds using double relaxation theory while the case is not obtained from the single relaxation theory. From the above result, it is shown that the negative dispersion of sound velocity is allowed in the case of our model. Eq. (3.9a) and Eq. (3.9b) are linearised with respect to v assuming small dispersion, i.e., $v \sim v_0$ or $v_0 + v \sim 2v_0$;

$$\Delta_d \equiv \frac{v_\Omega - v_0}{v_0} = \frac{\Omega^2 \tau_d^2 \{v_\infty^2/v_0^2 - 1\}(\Omega^2 \tau_d^2 + 3)}{2(1 + \Omega^2 \tau_d^2)^2}, \qquad (3.10a)$$

$$\Delta_s \equiv \frac{v_\Omega - v_0}{v_0} = \frac{\Omega^2 \tau_s^2 \{v_\infty^2/v_0^2 - 1\}}{2(1 + \Omega^2 \tau_s^2)}. \qquad (3.10b)$$

Then

$$\frac{\Delta_s}{\Delta_d} = \left(\frac{\tau_s}{\tau_d}\right)^2, \qquad \frac{(1 + \Omega^2 \tau_2^d)^2}{(1 + \Omega^2 \tau_2^s)(3 + \Omega^2 \tau_2^d)} \qquad (3.11)$$

If we set

$$2\tau_d = \tau_s, \qquad (3.12)$$

$$\frac{\Delta_s}{\Delta_d} = 4 \frac{(1 + \frac{\Omega^2 \tau_s^2}{4})^2}{(1 + \Omega^2 \tau_s^2)(3 + \frac{\Omega^2 \tau_s^2}{4})}$$

The curve Eq. (3.12) is presented in Fig.2. This curve shows that for a certain frequency range the dispersion given by this model is smaller than the dispersion by single relaxation theory. The systematically larger dispersion by an amount which appear to be outside the range of experimental error than the predicted dispersion was pointed out by J. E. Piercy and G. R. Hanes[18]. Absorption coefficient due to bulk viscosity defined by $\alpha_{\eta'} = I_m k$ is obtained from (3.4) where k is obtained by the relation, $\Omega = v(\Omega)\, k$.

But, instead of the direct calculation, we start with the result obtained by the single relaxation theory and transfer this result to our case by appropriate replacements of parameters. From the single relaxation theory we obtain

$$v_{\Omega}^2 = v_{\infty}^2 + \frac{a_1}{1 - i\Omega\tau_s}, \quad v_{\infty}^2 + a_1 = v_0^2$$

then

$$v_{\Omega}^2 = \frac{v^2 + a_1 - i\Omega\tau_s v_{\infty}^2}{1 - i\Omega\tau_s} . \tag{3. 13}$$

On the other hand, from the double relaxation theory we obtain

$$v_{\Omega}^2 = \frac{v_{\infty}^2(1 - \Omega^2\tau_1\tau_2) + a_1 + a_2 - i\Omega(\tau_1 + \tau_2)v_{\infty}^2}{1 - \Omega^2\tau_1\tau_2 - i\Omega(\tau_1 + \tau_2)} . \tag{3. 14}$$

here we omitted the cross terms $a_1\tau_2$ and $a_2\tau_1$.

Comparing (3.13) with (3.14) we can find the rule of the correspondence as follows,

1) In the first terms of numerator and denomerator 1 must be replaced by $1 - \Omega^2\tau_1\tau_2$.

2) a_1 must be replaced by $a_1 + a_2$.　　　　　　　　　　　　　　(3. 15)

3) τ_1 must be replaced by $\tau_1 + \tau_2$.

Eq. (3.13) lead to the well known expression for

$$\alpha_{\eta'} = I_m k = \frac{\Omega^2\tau_s\{v_{\infty}^2/v_0^2 - 1\}}{2v_0(1 + \Omega^2\tau_s^2)} . \tag{3. 16}$$

Here we assumed small dispersion i.e. $v_{\infty}^2/v_0^2 \sim 1$ in the last step in Eq. (3.16). By Fabelinskii et al. and by L. V. Lanshina et al. Eq. (3.16) is used to explain the experimental data for absorption of sound wave [17,19].

According to (17) Eq. (3.16) explains the experimental result very well for CH_2Cl_2 in spite of the fact that molecule CH_2Cl_2 has many vibrational modes and that among these modes the pair satisfying $C_\nu/C_{\nu'} \sim 1$ exists, here C_ν, $C_{\nu'}$ are vibrational specific heat associated with τ and τ' mode respectively.

This suggests that two internal processes of relaxation are concerned with two

modes of vibration which have different vibrational specific heat each other. Here we assume that we can set as

$$\frac{\tau_1}{\tau_2} = \frac{C_1}{C_2} = n \qquad (3.17)$$

where C_1 and C_2 two arbitrary vibrational specific heats of molecules.

Let us transform Eq. (3.16) to the case where two different processes of relaxation occur simultaneously using Eq. (3.15), Eq. (3.17) and the assumption of small dispersion. Then we obtain

$$\alpha_{\eta'} = \frac{\Omega^2 \tau_2 (n+1) \{v_\infty^2/v_0^2 - 1\}}{2v_0 \{(1 - n\Omega^2 \tau_2^2)^2 + \Omega^2 \tau_2^2 (n+1)^2\}}. \qquad (3.18)$$

Recalling that total absorption coefficient α is equal to $\alpha_\eta + \alpha_{\eta'} + \alpha_k$, we obtain finally the form of $\alpha(\Omega)$ for this case (assuming that absorption due to thermal diffusion is negligible being compared with α_η and $\alpha_{\eta'}$).

$$\frac{\alpha}{\Omega^2} = \frac{A}{(1 - n\Omega^2 \tau_2^2)^2 + \Omega^2 \tau_2^2 (n+1)^2} + B \qquad (3.19)$$

Here $A \equiv \dfrac{2(n+1)\{v_\infty^2/v_0^2 - 1\}}{2v_0}$ characterizes the absorption of sound wave at low

frequency and

$B \equiv \dfrac{2\eta}{3v_0^3 \rho}$ characterizes the absorption of sound wave at high frequency. Shear

viscosity assumed to be constant. From Eq. (3.19) we obtain

$$\frac{\alpha_0'}{\Omega_0^2} \Big/ \frac{\alpha_h'}{\Omega_h^2} = (1 - n\Omega_h^2 \tau_2^2)^2 + \Omega_h^2 \tau_2^2 (n+1)^2. \qquad (3.20)$$

Here

$$\frac{\alpha_0'}{\Omega_0^2} = \lim_{\Omega \to 0} \frac{\alpha_{\eta'}}{\Omega^2}$$

and α_h'/Ω_h^2 is the value of $\alpha_{\eta'}/\Omega^2$ at fixed hypersonic frequency Ω_h.

On the other hand, usual relaxation theory with a single relaxation time gives

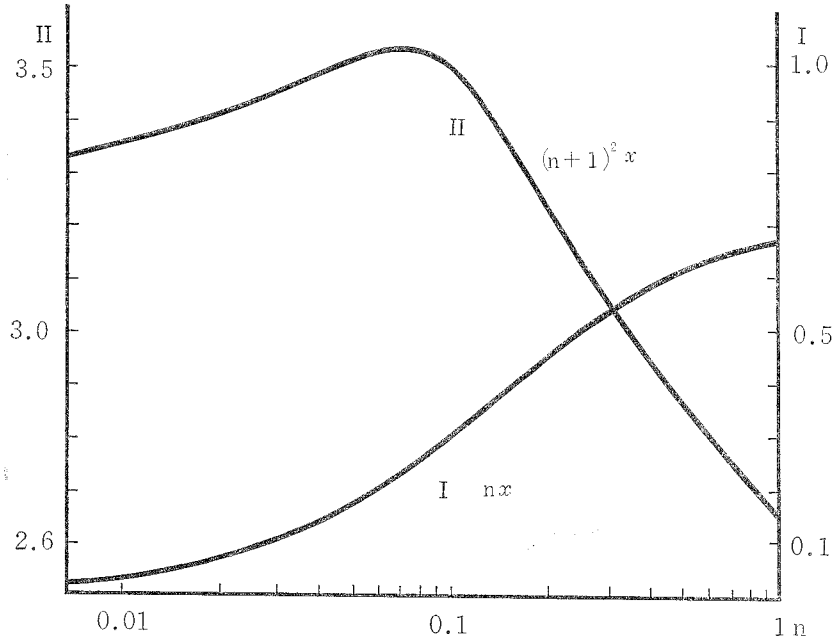$$\frac{\alpha}{\Omega^2} = \frac{a}{1 + \Omega^2 \tau_s^2} + b. \qquad (3.21)$$
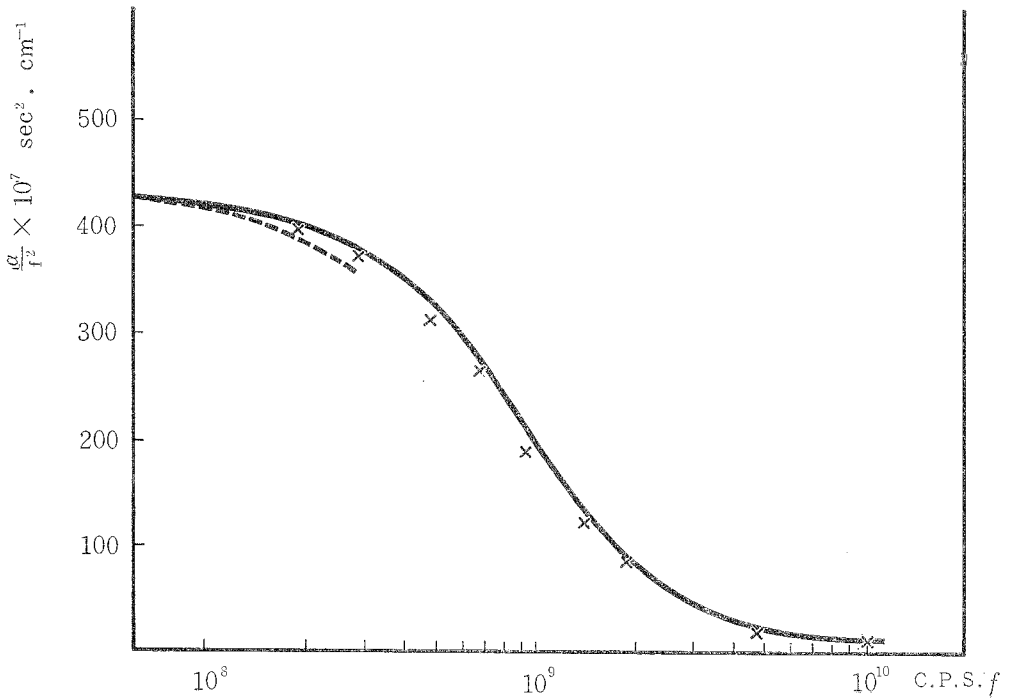
Fig. 3 nx and $(n+1)^2x$.

Fig. 4   The curve of α/f^2.

Here

$$a \equiv \frac{\tau_s \{v_\infty^2/v_0^2 - 1\}}{2v_0} = \frac{\alpha_0}{\Omega_0^2} - b,$$

$$b \equiv B, \tag{3.22}$$

and

$$\frac{\alpha_0'}{\Omega_0^2} \Big/ \frac{\alpha_h'}{\Omega_h^2} = 1 + \Omega_h^2 \tau_z^2 .$$

Eq.(3.21) gives only one curve once the values of a, α_h/Ω_h^2 and b are fixed. But unfortunately, the curves for chloroform under these conditions does not agree with the plots of α/Ω^2 given by ultrasonic measurement; that is, Eq. (3.21) does not show so large dispersion as that obtained from an experiment at ultrasonic region for the value of τ_s given by Eq. (3.22).

On the other hand (3.19) has a flexibility by changing the values of n even if A, α_h/Ω_h^2 and B are fixed. Two curves of Eq. (3.19) and Eq. (3.21) are presented in Fig. 4.

From the experimental values for the absorption of ultrasonic sound wave for $CHCl_3$,

$$\alpha_0'/f_0^2 \times 10^{17} = 400 sec^2. \ cm^1$$

On the other hand,

$$\alpha_h'/f^2 \times 10^1 = 25 sec^2. \ cm^1 \qquad \text{at } f_h = \frac{\Omega_h}{2\pi} = 3.4 \times 10^9 \text{c.p.s.}$$

Then Eq. (3.19) reduce to

$$16 = (1 - 4.55 nx)^2 + 4.55(n+1)^2 x,$$

$$x \equiv (10^{10} \tau_2)^2$$

Then

$$nx = \frac{(n + \frac{1}{n}) + (n + \frac{1}{n})^2 + 59.6}{9.05} \quad \cdots\cdots\cdots\cdots\text{curve I}$$

and

$$(n+1)^2 x = \frac{16 - (1 - 4.55 nx)^2}{4.55} \quad \cdots\cdots\cdots\cdots\text{curve II}$$

Maximum value of $(n+1)^2 \ x$ is given by $nx = 0.22$ which corresponds to

$$n = 0.88, \ \tau_2 \sim \sqrt{3} \times 10^{-10} \ sec.$$

From our consideration by the use of double relaxation theory, we obtain

$$\frac{\alpha}{f^2} = \frac{400}{(1 - 2.4 \times 10^{-21} \Omega^2)^2 + 3.51 \times 10^{-20} \Omega^2} + 10 + \cdots$$

Some values of α/f^2 are shown by the mark $\times$ in Fig.4. Here we used the value of $\tau_2 = \sqrt{3} \times 10^{-10}$ *sec.* The solid line shows the curve of $\alpha/f^2 \times 10^{17}$ versus f obtained from the single relaxation theory. The broken line shows the results obtained from the ultrasonic measurement.

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