



# Research of the temperature dependence of the vibrational relaxation time in liquids based on a mechanism based on Frenkel's idea of the nature of the thermal motion of molecules

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## Abstract

The processes of energy exchange between different degrees of freedom of liquid molecules play an important role in the study of a large number of physical and chemical phenomena, and the theoretical and experimental study of such phenomena has both scientific and practical significance. In the theoretical and experimental study of vibrational relaxation processes in liquids, models of binary collisions and collective interactions of the probability and time of perturbations of molecules are widely used. The first of these mechanisms ignores the collective interaction of molecules in liquids, while the connection of the necessary relaxation parameters during the study based on the second does not correspond to the experiment. For this reason, the question is raised about the introduction of a new model of the relaxation process in liquids. This article also discusses one of the theoretical works in this direction.

**Keywords:** Relaxation, Molecular vibrations, Vibrational relaxation, Transition probability, vibrational relaxation mechanism, vibrational - propagation energy exchange, stage of molecular jumps, oscillator, Debye vibrations, Debye frequency, activation energy, number of jumps per unit time - frequency, vibrational relaxation time.

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## Main part.

The question of calculating the probability of awakening and deactivation of internal molecular vibrations in liquids is one of the very difficult ones. In a strict consideration of this issue, such calculations require a quantum mechanical description. In [1-4] it is emphasized that the solution of this problem based on classical methods also correctly reflects the results obtained when solving the problem based on quantum mechanics methods in fundamental cases. The

results obtained in the study of VT exchange, discussed in the previous paragraph, emphasize that with proper modeling of the process, satisfactory results can be obtained even when described by the classical method.

Based on the mechanism of vibrational relaxation, we believe that intensive VT-exchange in molecular collisions occurs at the stage of molecular jumps. In this process, it is clearly seen that along with the awakening of the intense translational motion of one or



more particles, the awakening of Debye oscillations occurs.

Similarly, the reverse process occurs, i.e. the process of occurrence of fluctuations in the VT exchange. [12]

To estimate (calculate) the probability of an exchange of Watts occurring with the participation of Debye oscillations, consider the following model process. We study the collision process of two harmonic oscillators, one of which simulates a molecule and the other a Debye oscillation. In the process of deactivation of high-frequency molecular vibrations, the increased part of the relative energy of the center of gravity of the oscillators is a part of the translational motion energy concentrated in colliding oscillators

$$\begin{aligned} \ddot{x}_1 + \omega^2 x_1 &= \frac{1}{m_1} \Phi_1(t, x_1, x_2) \\ \ddot{x}_2 + \Omega^2 x_2 &= \frac{1}{m_2} \Phi_2(t, x_1, x_2) \end{aligned} \quad (1)$$

Here  $x_1, x_2$  the internal molecular coordinates of the first and second oscillator, respectively,

$m_1, m_2, \omega, \Omega$  effective weights of oscillator, respectively and  $\mu$  – private frequencies

$\Phi_1(t, x_1, x_2)$   $\Phi_2(t, x_1, x_2)$  - the forces arising from the interaction of oscillators, and these forces  $\Phi_1(t, x_1, x_2)$  and  $\Phi_2(t, x_1, x_2)$ , we can determine based on the following considerations. Assuming that collisions of two oscillators are similar to collinear collisions, we can write it down as follows.

$$V(r, x_1, x_2) = W_0 \exp[-\alpha(z - \lambda_1 x_1 - \lambda_2 x_2)] \quad (2)$$

In (2)  $r$  – the coordinate of the relative motion of the centers of gravity of the oscillators,  $\alpha^{-1}$  -the radius of intermolecular interaction,  $\lambda_1$  and  $\lambda_2$  -the parameters that determine the position of the center of gravity in the molecules. In collinear collisions of diatomic homonuclear

molecules, designations in the form of  $\lambda_1 = \lambda_2 = \frac{1}{2}$ ,  $W_0 = \frac{\mu\omega^2}{2}$  were used for the above parameters.

The reduced mass of colliding molecules is denoted by the letter  $\mu$ , the forces  $\Phi_1(t, x_1, x_2)$  and  $\Phi_2(t, x_1, x_2)$  are defined as a derivative obtained from the interaction potential of two oscillators by the internal molecular coordinates of the oscillators.  $r$  – an exact function of time is calculated. [18,19]

$$\Phi_1 = -\frac{\partial V}{\partial x_1}, \quad \Phi_2 = -\frac{\partial V}{\partial x_2} \quad (3)$$

that have not yet had time to pass into low-frequency Debye oscillations. Thus, the proposed model, in addition to the energy exchange known to us between internal molecular vibrations and the phonon subsystem, also takes into account a new factor, that is, the transfer of energy even to advanced degrees of freedom of the closest partners. Such energy transfer is associated with the excitation and deactivation of the limiting amplitude oscillations and formally cannot be considered in terms of phonon cross-sections.[12-13]

The equations of motion of two interacting harmonic oscillators are written as [13].



Comparing the amplitudes of excitation of oscillations at the lower (lower) levels of the excited state with the radius of the intermolecular interaction, it is seen that their magnitude is much smaller than the radius of the intermolecular interaction,  $\alpha x_1 \ll 1, \alpha x_2 \ll 1$ . Hence (2) can be expressed as.

$$V(r, x_1, x_2) = W_0 \exp[-\alpha r] \sum_{m,n} \frac{C_{m+n}^n}{(m+n)!} (\alpha \lambda_1 x_1)^m (\alpha \lambda_2 x_2)^n \quad (4)$$

Here  $C_{m+n}^n = \frac{(m+n)!}{n!}$  are binomial coefficients, and  $r = r(t)$  is determined from the equation of motion of the center of gravity of the oscillators without oscillation.

Suppose that at the initial moment of time the molecular oscillator is not excited, while the Debye oscillator is excited and has an energy of  $E_0$ , and the coordinates are determined as follows.  
 $x_1(-\infty) = 0, \dot{x}_1(-\infty) = 0$

$$E_0 = q\hbar\Omega., \quad \left. \frac{m_2}{2} (\dot{x}_2^2 + \Omega^2 x_2^2) \right|_{t \rightarrow -\infty} = E_0 \quad (5)$$

In the process of collapse, the increase in the energy of the oscillatory motion of the first oscillator to  $\Delta E$  occurs with the loss of the second oscillator  $q$  quantum, and the appearance of  $\Delta E$  is determined as follows.

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$$\Delta E = \frac{1}{2m_1} \left| \int_{-\infty}^{\infty} \Phi_1 \exp(-i\omega t) dt \right|^2 \quad (6)$$

In (6) in the formula  $\Phi_1(t, x_1, x_2)$  only  $m = 1, n = q$  terms should be preserved. Here the appearance of  $\Phi_1(t, x_1, x_2)$  function is written as follows.

$$\Phi_1 = \frac{1}{(q+1)!} C_{q+1}^q \lambda_1 \alpha x_1 W_0 (\alpha \lambda_2 x_2)^q Sh^2 \left( \frac{\alpha \nu t}{2} \right) \quad (7)$$

$x_2$  at zero approximation, i.e. it is necessary to take the following form,  
 $x_2 = x_2^0 [\exp(i\Omega t) + \exp(-i\Omega t)]$ , (8)

Here: 
$$x_2^0 = \left( \frac{E_0}{2m_2\Omega^2} \right)^{\frac{1}{2}}$$

taking into consideration (5) – (8) formulas, for  $\Delta E$  the following formula can be written.  
 (indicator of exponent  $(\omega + q\Omega)$  will not be considered.

$$\Delta E = \frac{2\pi^2 \lambda_1^2 \lambda_2^{2q} \alpha^{2(q-1)} \mu^2 \varepsilon_0^q}{(q!)^2 2^q m_1 m_2^q \Omega^{2q} \Omega^{2q}} \frac{(\omega - q\Omega)^2}{Sh^2 \left[ \frac{\pi(\omega - q\Omega)}{\alpha \nu} \right]} \quad (9)$$



When the  $q > 1$  condition is met in (4)  $n < q$  terms also contribute to  $\Delta E$ .

The contribution of these additional terms is in the terms that are formed due to the connection of  $\Phi_2$  with  $x_2$ .

The contribution of these terms when the condition  $\Omega_D \tau_{m\dot{y}} \gg 1$  is fulfilled (this condition is fulfilled well) is considered quite small. ( $\Omega_D$  is the characteristic of the Debye frequency,  $\tau_{m\dot{y}}$  is the duration of the collision).

It is necessary to average  $\Delta E$  over the Maxwell distribution of velocities and over all frequencies of thermal oscillations, i.e.

$$\Delta \bar{E}(q) = \int_0^\infty \int_0^{\Omega_D} \Delta E f(v) D(\Omega) dv d\Omega \quad (10)$$

Here:  $f(v) = \frac{v^3}{2} \left( \frac{\mu}{\kappa T} \right)^2 \exp\left(-\frac{\mu v^2}{2kT}\right)$

$D(\Omega)$  – is the spectral density of normal oscillations, which is convenient for simplicity to choose

according to the formula determined by Debye, presented in the form  $D(\Omega) = \frac{3\Omega^2}{\Omega_D^3}$  below. [10-

15]

The characteristic Debye frequency is obtained in the following form.

$$\Omega_D = \frac{2\pi \cdot 10,4}{\alpha\beta} \sqrt{\frac{\varepsilon}{M} \left(1 - \frac{5}{12}\right)} \quad (11)$$

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is in the form, its expression is determined based on the cellular model of the fluid. When moving from  $\Delta \bar{E}(q)$  to the probability of VT –exchanging, we use the following equality. [14-16]

$$\Delta \bar{E}(q) = \sum n \hbar \omega P_{0n}^{(q)}, \quad (12)$$

here  $P_{0n}^q$ , here  $0 \Rightarrow n$  - the probability of an oscillatory transition is assumed that  $q$  – Debye quanta are simultaneously absorbed during this transition.

$$P_{0n}^{(q)} \ll P_{01} \quad \text{By erda: } P_{01}^q = \frac{\Delta E(q)}{\hbar \omega}$$

At the same time,  $0 \Rightarrow n$ , which occurs by absorption of  $q$  – Debye quanta, we obtain the following expression for the probability of VT –exchange:

$$P_{10}^{(q)} = \frac{3}{2\Omega_D^2} \left( \frac{\mu}{kT} \right)^2 \int_0^\infty \int_0^{\Omega_D} \frac{\Delta E}{\hbar \omega} v^3 \exp\left(-\frac{\mu v^2}{2kT}\right) \Omega^2 d\Omega dv \quad (11)$$



(2.2.11) when calculating the integral,  $\frac{\pi(\omega - q\Omega)}{\alpha\nu} \gg 1$  conditions are checked and the adiabaticity of the collision is determined. As a result of calculating the integral (11), we obtain the following expression for the probability of  $VT$ -exchange associated with the simultaneous passage of  $q$  Debye quanta.

$$P_{10}^{(q)} = G \left\{ \frac{1}{3B_0} \left[ (\omega - q\Omega_D)^{\frac{10}{3}} \exp \left[ -3B_0(\omega - q\Omega_D)^{\frac{2}{3}} \right] - \omega^{\frac{10}{3}} \exp \left( -3B_0\omega^{\frac{2}{3}} \right) \right] \right\} +$$

$$+ \frac{1}{3B_0^2} \left[ (\omega - q\Omega_D)^{\frac{8}{3}} \exp \left[ -3B_0(\omega - q\Omega_D)^{\frac{2}{3}} \right] - \omega^{\frac{8}{3}} \exp \left( -3B_0\omega^{\frac{2}{3}} \right) \right] +$$

$$+ \frac{1}{6B_0^3} \left[ (\omega - q\Omega_D)^2 \exp \left[ -3B_0(\omega - q\Omega_D)^{\frac{2}{3}} \right] - \omega^2 \exp \left[ -3B_0\omega^{\frac{2}{3}} \right] \right]$$

$$G = 36 \sqrt{\frac{2}{3}} \frac{\mu^{\frac{5}{2}} \pi^{\frac{7}{2}} \alpha^{2q-3} \lambda_1^2 \lambda_2^{2q} (\hbar q)^q}{q(q!)^2 2^q m_1 m_2^q \omega (kT)^{\frac{1}{2}} \hbar \Omega_D^{q+1}} \quad B_0 = \left( \frac{\mu \pi^2}{2\alpha^2 kT} \right)^{\frac{1}{3}} \quad (12)$$

The calculation of  $P_{10}^{(q)}$  was carried out within the framework of a one-dimensional collision model, without taking into account the influence of gravitational forces and anharmonism.

The main proportion (12) to the value of the probability of  $VT$ -exchange is added by the values in the first two brackets of the formula, while the value of  $P_{10}^{(q)}$  decreases sharply with an increase in the value of  $q$ , [14] that is,  $P_{10}^{(0)} \gg P_{10}^{(1)}, P_{10}^{(0)} \gg P_{10}^{(2)}$ .

And the probability of passing  $P_{10}^{(q)}$  is determined by the following formula, adjusted for the orientation coefficient and the influence of gravitational forces.

$$P_{10}^{(q)} = \frac{3}{\Omega_D^3} \int_0^{\Omega_D} P_{10}^{(q)}(\Omega) \Omega^2 d\Omega \quad (13)$$

Here  $P_{10}^{(q)}(\Omega)$  is defined for polyatomic molecules based on a generalization of the formula obtained by Tankosh. Generalizing the Tankosh formula for the case under consideration, i.e. studying the interaction of two oscillators, it is assumed that one of the oscillators simulates the Debye oscillation and, in addition,

Thus, in the proposed mechanism of  $VT$ -exchange in liquids, along with binary collisions, awakenings (deactivation) of effectively collective vibrational modes are also taken into account, and for probability the formula  $(\Omega_D)$  is obtained, which is a function of  $P_{10}^{(q)}(\Omega)$ . On this basis, the dependences of the  $VT$ -exchange on the density of the liquid and temperature are obtained. [20-23]



The mechanism of vibrational relaxation under consideration is based on the fact that in the process in which molecular jumps of  $VT$ -exchange occur, and as a result of this, there is a simultaneous awakening of Debye vibrations associated with the awakening of degrees of freedom that do not occur in one or more particles of the medium.

With an increase in the density of the medium, the Debye frequency increases, which leads to a decrease in the energy difference between internal molecular vibrations and the quanta of Debye vibration, which leads to an increase in the probability of  $VT$ -exchange. [12-14]

When calculating the probability of  $VT$ -exchange in liquid hydrogen ( $H_2$ ) at a temperature of  $T = 293$  K, the formula (6) was used. In addition, the temperature dependence of the probability of  $VT$ -exchange was studied. With an increase in temperature, an increase in the probability of  $VT$ -exchange was also observed.[18-19]

The temperature dependence of the probability of  $VT$ -exchange is well studied in the eyes.

The connection of  $LnP_{10}^{(q)}(\Omega)$  with  $T^{-\frac{1}{3}}$  has the form of a straight line, the switching mechanism of  $VT$ -exchange differs from the Landau–Lifshitz model in the model we are considering. For this

reason, the connection  $LnP_{10}^{(q)}(\Omega)$  with  $T^{-\frac{1}{3}}$  should consist of a curve, we believe that even in the liquid state,  $H_2$  comes out close to a straight line, based on the properties of gaseous hydrogen, that is, its curvature is very small, and the density of the gas is relatively small.

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To calculate the relaxation time of oscillations, it is necessary to know the number of jumps of liquid molecules per unit of time  $Z$  or the average value of the residence time of the molecule in the bound (settled) state

$$\tau_{VT} = [ZP_{10}(1 - e^{-\theta})]^{-1} \quad (14)$$

$$\tau = \tau_0 \exp\left(\frac{\Delta W}{kT}\right) \quad (15)$$

Here  $\tau_0$  is the half-life of the molecule oscillation around the equilibrium state, and  $\Delta W$  is the activation energy in the jump. Calculation of  $\tau$  requires calculation of activation energy. To quantify the activation energy, we use the Frenkel–Andrade formula.

$$\eta = C \exp\left(\frac{\Delta W}{kT}\right) \quad (16)$$

Here the coefficient up to the exponent  $C$  - is weakly related to temperature and density. Equation (15) was obtained by Frenkel in the process of processing ideas about the thermal motion of liquid molecules, this equation, although it is a negative equation, correctly explains the experimental results, therefore, to calculate  $\Delta W$  with great accuracy. gives an opportunity.

In the question we are considering, we are convinced that the fact that we applied Frenkel's ideas about the thermal motion of liquid molecules to the calculation of the vibrational relaxation



time in liquids is of great importance, having studied the specifics of the thermal motion of molecules and applying patterns within one model to determine the probability of VT –exchange  $P_{10}^{(q)}(\Omega)$ , as well as the number of jumps Z.[13-16]

For liquid hydrogen, the viscosity coefficient has been determined by the following formula.

$$\eta = \eta_0(T) + A(\rho) \exp\left(\frac{B(\rho)}{T}\right) \eta \quad (17)$$

Here  $\eta_0(T)$  will depend only on temperature at low densities.

$$\eta_0(T) = 8,5558 \left[ \frac{T^{\frac{3}{2}}}{T + 19,55} \right] \left[ \frac{T + 650,39}{T + 1175,9} \right] \quad (18)$$

$A(\rho)$  and  $B(\rho)$  will be determined as follows.

$$\ln A(\rho) = 5,7694 + \ln \rho + 65,0 \rho^{\frac{3}{2}} - 6,110^{-5} e^{12,79\rho} \quad (19)$$

$$B(\rho) = T_0 \left[ 10,0 + 7,2 \left[ \left( \frac{\rho}{0,07} \right)^6 - \left( \frac{\rho}{0,07} \right)^{\frac{3}{2}} \right] \right] - 17,63 \exp \left[ -58,75 \left( \frac{\rho}{0,07} \right) \right] \quad (19)$$

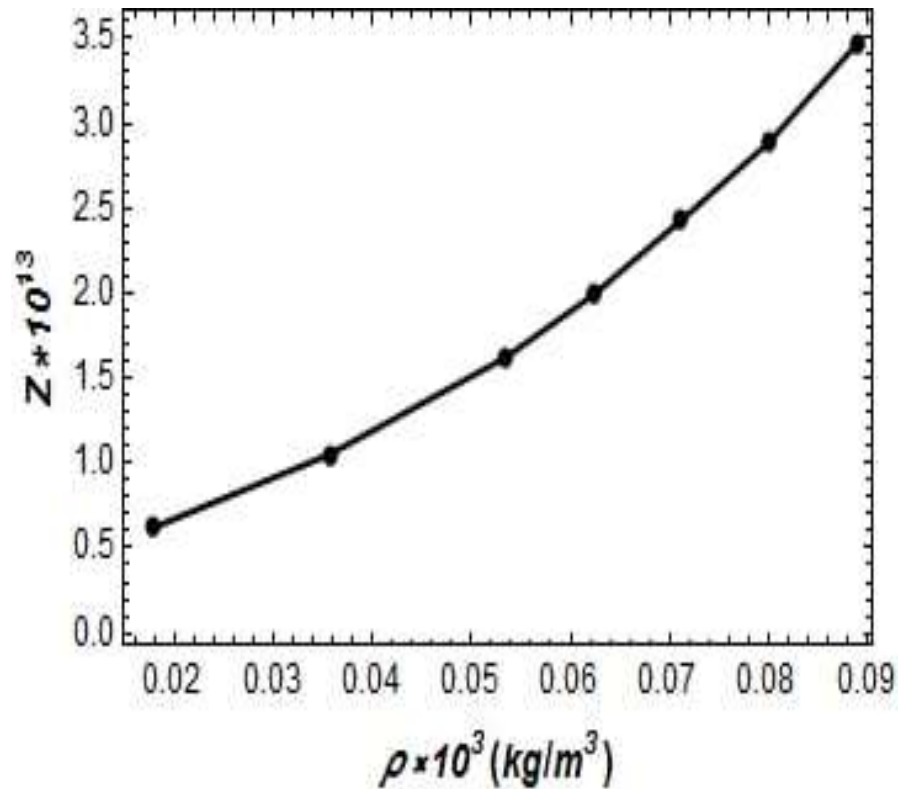
Here  $T_0 = 1,0$   $B(\rho) = \frac{\Delta W}{\kappa} \quad (20)$

The binding of the activation barrier  $\frac{\Delta W}{\kappa}$  to the density was determined by the viscosity coefficient

$\eta$  out of  $\frac{1}{T}$ . As the density increases, the value of  $\Delta W$  nomotons increases. The value of  $\tau_0$  – parameter decreases monotonically with increasing density.

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1-graph. Graph of the dependence of the frequency density  $Z$  (number of jumps per unit of time) for liquid hydrogen

At low indicators of densities  $Z$  increases, in liquids at high indicators of densities  $Z$  non-monotonically decreases.

The relaxation time of vibrations in liquids is determined by the formula

$$\tau_{VT}^{\lambda} = \frac{\tau_0 \exp\left(\frac{\Delta W}{kT}\right)}{\sum_q^n P_{10}^q \left[1 - \exp\left(-\frac{\hbar\omega}{kT}\right)\right]} \quad (2.3.9)$$

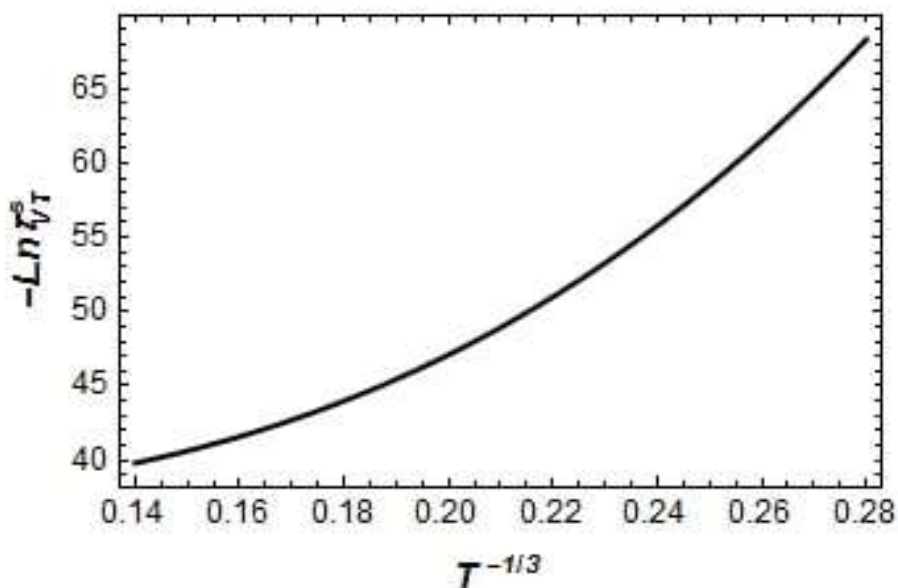
The experimental study of the vibrational relaxation time was studied first using ultrasound methods,

and then using the Brillouin method. In experimental studies, the binding of  $\frac{1}{\tau_{VT}^{\lambda}}$  with a density of

$\rho$  was first studied. The binding of  $\frac{1}{\tau_{VT}^{\lambda}}$  with  $\rho$  increased with increasing density.







Graph 2. Graph of the dependence of the logarithm of the relaxation time of oscillations in

liquids on temperature  $T^{-\frac{1}{3}}$ .

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In our proposed model, the relaxation time of vibrations in liquids is determined by the formula (2,3.9) given above. The logarithmic values of the relaxation time of oscillations  $T^{-\frac{1}{3}}$  out of  $Ln \tau_{VT}$ , at which a curvilinear connection is obtained, are associated with a change in the density of the liquid.

The literature indicates that  $T^{-\frac{1}{3}}$  bonds  $Ln \tau_{VT}$  in gases are rectilinear. The above changes in liquids, that is, the result of calculations programmed on a computer, can also be justified by the fact that the mechanism of interaction in liquids differs from the mechanism of interaction in gases.

Based on the above points, we formulate the following main conclusions.

**Main conclusions.**

1. The theories of vibrational relaxation in liquids were critically studied and, on this basis, the mechanism of vibrational relaxation used in the study of vibrational relaxation in liquids and the vibrational –progressive energy exchange based on it were analyzed. It was

noted that the basic ideas of the mechanism of binary collisions can also be used in liquids, given that in liquids, energy exchange at low densities occurs in the same way as in gases, that is, in paired collisions.

2. Taking into account the specifics of liquids when calculating the relaxation time of vibrations in liquids, the concepts of collision frequency or effective number of collisions were introduced. It is concluded that it is necessary to use a new mechanism for their calculation.

3. As a result of the analysis of the vibrational relaxation model based on Frenkel's ideas about the thermal nature of liquid molecules, formulas were derived for calculating the vibrational relaxation time for liquid hydrogen, the probability of



vibrationally progressive energy exchange and the number of jumps of molecules per unit of time or the probability of VT-energy exchange, based on the solution of the corresponding equations and calculated on the basis of these formulas in computer programming.

4. For liquid hydrogen, a graph of the dependence of the number of jumps per unit of time on the density of  $Z$  was constructed. Analysis of the graph shows that the activation energy increased unevenly with an increase in

density  $\frac{\Delta W}{\kappa}$ , and with a decrease in  $\tau_0$ , an increase in  $Z$  was observed, which confirmed its suitability for experiments.

4. With an increase in the density of the liquid, the relative role of VT-energy exchange, which occurs simultaneously with the awakening of  $q$  - Debye quanta, changes. At low densities  $q=0$ , mainly  $0 \Rightarrow 1$  vibrational–progressive transitions play a key role, at high densities the proportion of transitions  $q=1$  increases, the hypothesis that transitions  $q=2$  will always be small has been tested, the results of theoretical studies have shown that they practically coincide with the experiment.

5. The dependence of the probability of VT- energy exchange and the relaxation time of oscillations on temperature is studied. The results obtained in the form of a graph showed the accuracy of the change in the probability of VT- energy exchange in density, as well as a reduction in the error of the results due to the improvement of computing devices.

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