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# Complex non-exponential form of damped vibrations in uniaxially oriented polymeric materials with one mechanical degree of freedom

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**Abstract.** In the present paper the relaxation properties of uniaxially oriented polymer threads are investigated. Vibrational relaxation in mechanical system with one degree of freedom is analyzed. This system consists of a heavy weight suspended on a light thread made of a uniaxially oriented polymer material.

The authors experimentally confirmed the existence of beats for a number of polymer materials (polycapromide, SVM, Terlon, Armos, etc.) at a certain static load level below the glass transition temperature. A physical explanation is given for the existence of the second vibration mode.

On the one hand, elastic oscillations occur in the system, on the other hand, according to the barrier model, oscillations of the occupation numbers of energy levels take place. Thus, another vibration mode arises, associated with highly elastic deformation, since highly elastic deformation is determined by the occupation numbers of the corresponding states. At close frequencies the beat effect is observed.

**Keywords:** uniaxially oriented polymeric materials, longitudinal low amplitude oscillations, highly elastic deformation, constitutive equation, beats

## Introduction

The authors studied the elastic-relaxation properties of uniaxially oriented polymer threads in the dynamic mode of deformation. A theoretical explanation of the beat's occurrence is proposed.

In the second half of the last century the effects of occurrence of vibrational relaxation in some systems were discovered. For instance, the phenomenon of the occurrence of current oscillations in a homogeneous multi-valley semiconductor placed in a strong electric field was discovered—the Gunn effect (Gunn 1963). In chemistry, it is the Belousov-Zhabotinsky reaction (Belousov 1982; Zhabotinskii 1974)—a class of chemical reactions occurring in an oscillatory mode, in which some reaction parameters (color, concentration of components, temperature, etc.) change periodically, forming a complex spatio-temporal structure of the reaction medium. In the present article vibrational relaxation in a mechanical system with one degree of freedom is analyzed. This system consists of a heavy weight suspended on a light thread made of a uniaxially oriented polymer material below the glass transition temperature.

Previously, in (Romanova et al. 2000; 2005; 2007b; Rymkevich et al. 2014) for a number of studied polymer threads (SVM, lavsan, terlon, etc.), it was found that in a certain range of mechanical stresses

(or levels of static deformation) a complex non-exponential form of damped oscillations or beats schematically presented in Fig. 1 is observed, which is difficult to explain using the traditional description.

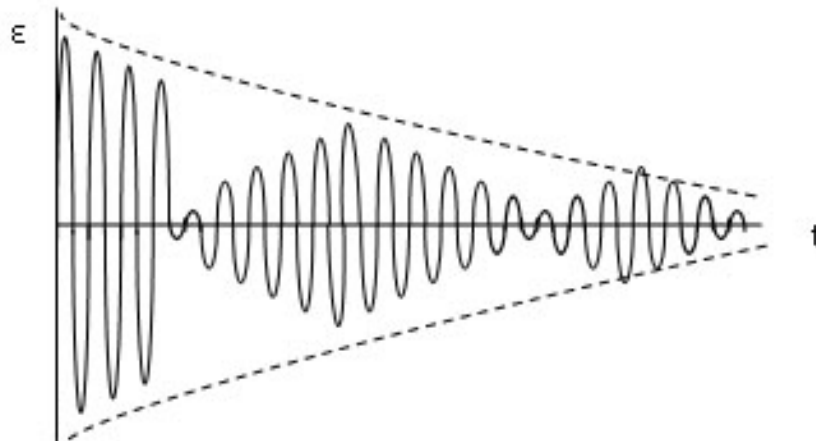


Fig. 1. Beatings (Amplitude-Modulated Free Oscillations)

The deformation curve in Fig. 1 can be represented at least as the result of the addition of two vibration modes. The question is why the second mode of oscillations appears in a mechanical system that formally has one degree of freedom. There are at least two explanations for this phenomenon. The first one is that the material under consideration obeys non-linear differential equations. The second one is that there are “hidden” degrees of freedom in the system. The authors adhere to the second explanation, because nonlinear equations should be linearized when studying low-amplitude oscillations. From the point of view of mechanics, the method of integral transformations based on the Boltzmann-Volterra equations when describing the mechanical properties of highly oriented polymers is often used (Gorshkov et al. 2004; Makarov et al. 2015; Romanova et al. 2007a; Stalevich et al. 2005). The authors of this article had previously shown that the fact of the presence of beats requires the existence of an oscillatory relaxation core, which, in principle, does not contradict the laws of mechanics (Rabotnov 1988), but requires additional explanations. Various rheological models of uniaxially oriented polymeric materials were considered in (Gorshkov et al. 2015; Rymkevich et al. 2021). In particular, a rheological model of polymer filament with hidden degrees of freedom was proposed in (Gorshkov et al. 2023). The described model consists not only of a spring and a damper, but also of a spiral visco-elastic element connected in series to them (Fig. 2).

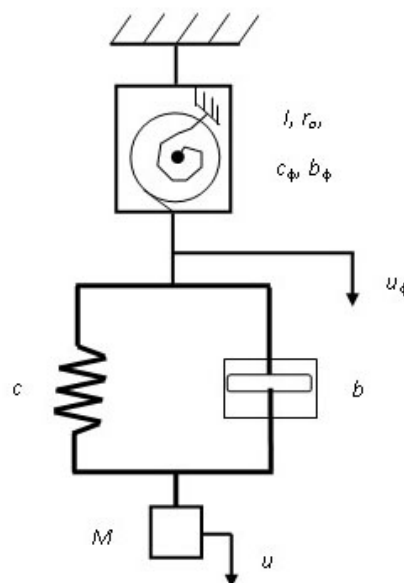


Fig. 2. Structural rheological model of a polymer thread with a spiral visco-elastic element

This mechanical model is a system with two degrees of freedom. One degree of freedom corresponds to translational motion  $u$ , the other one to rotational (rotary) motion  $\varphi$ . Rotational motion (rotational degree of freedom) is a hidden from the “macro-observer” form of motion. The transformation of the rotational motion of the ball into translational movement ensures the occurrence of an oscillatory beating mode. This rheological model can sufficiently describe the experimental results but does not provide any physical explanation. The authors propose to consider the complex and diverse supramolecular structure of oriented polymer materials as a set of different groups of macromolecules (clusters) in one of the possible stable states, separated by energy barriers (Gorshkov et al. 2013; Romanova 2007b).

As it was shown in (Rymkevich 2018), regardless of the conformational models considered, all allowed highly elastic (conformational) states differ from each other only in the heights and widths of the energy barriers between them. Therefore, the following assumptions for describing oriented polymer materials are accepted:

(a) polymer macromolecules contain groups of molecules in different stable states separated by energy barriers;

(b) the elastic part of the deformation obeys Hooke’s law

$$x = \frac{\sigma}{E_0},$$

where  $E_0$  is the true elastic modulus.

Here and further, we assume that the total deformation of a polymer material can be represented as the sum of two terms—elastic ( $x$ ) and highly elastic ( $\varepsilon_{he}$ ), i. e.:

$$\varepsilon = x + \varepsilon_{he}.$$

Groups of macromolecules that can change their conformations during deformation will be called active conformational elements (ACE). We present active conformational elements with two stable energy states in the form of a model depicted in Fig. 3.

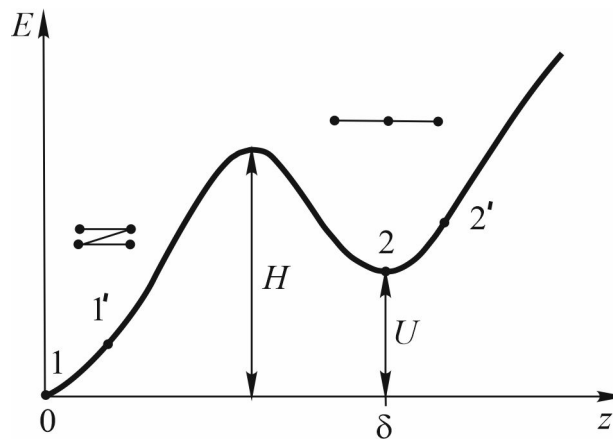


Fig. 3. Energy diagram of ACE as a function of cluster size for two stable states

In the energy diagram (Fig. 3), these two stable states with energies  $E_1 = 0$  and  $E_2 = U$  are respectively separated by a potential barrier of height  $H > 0$ . We will call state 1 “conditionally collapsed”, and state 2—“conditionally expanded”. In general case the value of the energy gap can be either greater or less than zero and depends on the type of conformational states. During the transition from state 1 to state 2, the geometry of the ACE changes, which is accompanied by absolute deformation  $\delta > 0$ . And the third assumption:

(c) all ACEs are fully described by three numerical characteristics: barrier height  $H$ , energy difference  $U$  and deformation quantum  $\delta$ .

An attempt was proposed in (Golovina et al. 2022) to explain the observed complex amplitude modulation using a two-stable state theory within the framework of the assumptions proposed above. But such a theory leads to a nonlinear differential equation, which does not allow us to give a complete explanation of the indicated phenomenon of amplitude modulation within the framework of the observed frequencies. Therefore, the authors propose to consider a theory with three stable states.

### Constitutive equation for the three-level model

Let us consider a model with three stable energy states (Fig. 4).

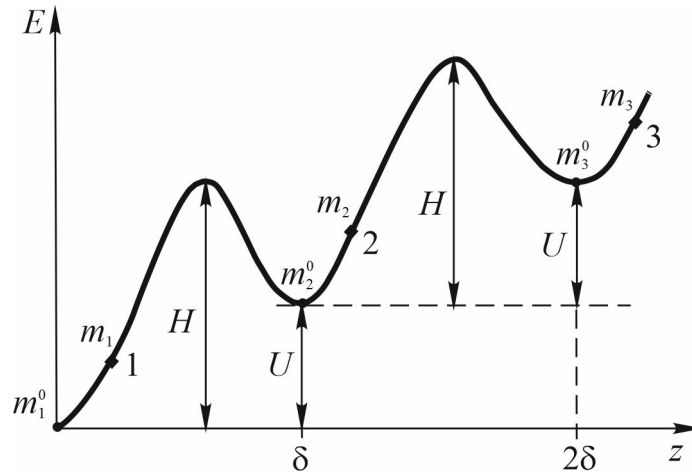


Fig. 4. Energy diagram of ACE as a function of cluster size for three stable states

A forced orientation of macromolecules takes place in the amorphous layers of a highly oriented polymer material under external load. Then, according to the barrier theory, groups (clusters) of macromolecules move from one stable state to another. It should be noted that in the absence of load (elastic deformation), the stable states are points with the number of clusters per unit length  $m_1^0$ ,  $m_2^0$  and  $m_3^0$ . The loaded state becomes stable under external load (points 1, 2 and 3 with occupation numbers  $m_1^0$ ,  $m_2^0$  and  $m_3^0$ )—Fig. 4. And according to the barrier theory, we can write a balance equation for these numbers:

$$\begin{cases} \frac{dm_1}{dt} = -m_1W_{\rightarrow} + m_2W_{\leftarrow} \\ \frac{dm_2}{dt} = -m_2(W_{\rightarrow} + W_{\leftarrow}) + m_1W_{\rightarrow} + m_3W_{\leftarrow} \\ \frac{dm_3}{dt} = -m_3W_{\leftarrow} + m_2W_{\rightarrow} \end{cases} \quad (1)$$

The state of an ideal highly oriented polymer material, consisting only of clusters of one type with an elastic modulus  $E_p$ , is characterized by the barrier height  $H$  (corresponding to the necessary energy reserve for another rotation of a chain link), the energy gap width  $U$  (corresponding to the difference in energies of two neighboring stable states), the quantum deformation value  $\delta$  and parameter  $\bar{\gamma}$ —the so-called structure-sensitive element, depending on the type of material (Romanova et al. 2005; Rymkevich 2018). In practice,  $U$  mostly takes negative values. We will measure all quantities  $H$  and  $U$  in kelvins as is customary in molecular physics.

In system of equations (1), the change in the number of clusters in the  $N$  stable state will be determined by the number of clusters moving from this state to the right (see Fig. 4) with probability  $W_{\rightarrow}$  per second, and the number of clusters moving from this state to the left with probability  $W_{\leftarrow}$  per second.

Let us assume that this system of clusters obeys Boltzmann statistics, according to which the probability of a transition from state 1 to state 2 (and similarly the probability from state 2 to state 1) is determined by the heights of the energy barriers  $H_{12}$  and  $H_{21}$ , respectively:

$$P_{12} = P_0 \cdot \exp(-H_{12}/T),$$

$$P_{21} = P_0 \cdot \exp(-H_{21}/T).$$

The number of transitions per second (fraction) is denoted as:

$$W_{\rightarrow} = \nu_0 \cdot \exp(-H_{\rightarrow}/T),$$

$$W_{\leftarrow} = \nu_0 \cdot \exp(-H_{\leftarrow}/T),$$

where  $\nu_0$  is the frequency of approach to the barrier per second.

During the deformation of material, the height of the barrier in the direction of orientation decreases by the value proportional to the square of the mechanical elastic stress (for example, rotation of Kuhn segment from one stable state to another). Therefore, the value  $H_{\rightarrow}$  is assumed to be equal to:

$$H_{\rightarrow} = H - \bar{\gamma}x^2,$$

where  $x$  stands for the value of elastic deformation ( $x = \sigma/E_0$ ).

As can be seen from Fig. 4,  $H_{\leftarrow} = H - U + \bar{\gamma}x^2$ .

As is customary in thermodynamics, it is more convenient to use the reduced energy values. Thus, we introduce the following notation:  $H^* = H/T$ ,  $U^* = U/T$ ,  $\gamma = \bar{\gamma}/T$  and  $\exp(U^*) = A$ . Here are the possible variants:  $A < 1$  (if a more stable state is a more oriented one—which is the most common in practice),  $A \approx 1$  (if the energies of these states are approximately the same), and  $A > 1$  (in the opposite case). Therefore:

$$\begin{aligned} W_{\rightarrow} &= \nu_0 \cdot \exp(-H_{\rightarrow}^*) = \nu_0 \cdot \exp(-H_{\rightarrow}/T) = \nu_0 \cdot \exp(-H/T + \gamma x^2) = \\ &= \nu_0 \cdot \exp(-H^*) \cdot \exp(\gamma x^2) = \frac{1}{\tau_0} \exp(-H^*) \cdot \exp(\gamma x^2), \end{aligned}$$

$$\begin{aligned} W_{\leftarrow} &= \nu_0 \cdot \exp(-H_{\leftarrow}^*) = \nu_0 \cdot \exp(-H_{\leftarrow}/T) = \nu_0 \cdot \exp(-H/T + U/T - \gamma x^2) = \\ &= \nu_0 \cdot \exp(-H^*) \cdot \exp(U^*) \cdot \exp(-\gamma x^2) = \frac{1}{\tau_0} \exp(-H^*) \cdot \exp(U^*) \cdot \exp(-\gamma x^2). \end{aligned}$$

Most polymer materials that are oriented (deformed) in this way differ only in barrier heights, deformation quantum and number of clusters. That is why it is convenient to introduce the characteristic relaxation time. In system (1), it is expedient to switch to dimensionless time  $\tau = t/\tau_{\delta}$ , where  $\tau_{\delta} = \tau_0 \cdot \exp(H^*)$  is the relaxation time,  $\tau_0 = 1/\nu_0$  is the material constant characterizing the average transition time with barrier height  $H \rightarrow 0$ . Then after introducing the new notation a more compact representation is obtained:

$$R_+ = W_{\rightarrow} \cdot \tau_{\delta} = \frac{1}{\tau_0} \exp(-H^*) \cdot \exp(\gamma x^2) \cdot \tau_0 \cdot \exp(H^*) = \exp(\gamma x^2), \quad (2)$$

$$\begin{aligned} R_- &= W_{\leftarrow} \cdot \tau_{\delta} = \frac{1}{\tau_0} \exp(-H^*) \cdot \exp(U^*) \cdot \exp(-\gamma x^2) \cdot \tau_0 \cdot \exp(H^*) = \\ &= \exp(U^*) \cdot \exp(-\gamma x^2) = A \cdot \exp(-\gamma x^2). \end{aligned} \quad (3)$$

Further we will take into account that  $R_+ \cdot R_- = A$ .

We denote the derivative of a certain value  $X$  with respect to a dimensionless parameter  $\tau$  as  $\overset{\circ}{X} = \frac{d}{d\tau}$ .

Taking into consideration the introduced notation when switching to dimensionless time, we obtain a system of equations (4) equivalent to system (1):

$$\begin{cases} \overset{\circ}{m}_1 = -m_1 R_+ + m_2 R_- \\ \overset{\circ}{m}_2 = -m_2(R_+ + R_-) + m_1 R_+ + m_3 R_- \\ \overset{\circ}{m}_3 = -m_3 R_- + m_2 R_+ . \end{cases} \quad (4)$$

Taking into account the normalization condition (law of conservation of the cluster number):

$$m_0 = m_1 + m_2 + m_3 . \quad (5)$$

Here  $m_0$  is the total number of clusters per length of the given polymeric sample.

The magnitude of the highly elastic part of the deformation can be represented as the difference between the total  $\varepsilon$  and elastic deformation  $x$ :

$$\varepsilon_{he} = \varepsilon - x = \delta[m_2 - m_2^0] + 2\delta[m_3 - m_3^0] = \delta(m_2 + 2m_3) - \Delta_0, \quad (6)$$

where  $\Delta_0 = \delta(m_2^0 + 2m_3^0)$  is the initial deformation.

Let us express  $m_1$  from the normalization condition (5) and substitute it into (4), thereby eliminating  $m_1^0$  from the system of equations:

$$\begin{cases} \overset{\circ}{m}_2 = -m_2(R_+ + R_-) + (m_0 - m_2 - m_3)R_+ + m_3 R_- = -m_3(R_+ - R_-) + m_0 R_+ - m_2(2R_+ + R_-) \\ \overset{\circ}{m}_3 = -m_3 R_- + m_2 R_+ . \end{cases} \quad (7)$$

Let us write down the first derivatives of  $R_+$  and  $R_-$ :

$$\overset{\circ}{R}_+ = R_+ \cdot 2\gamma x \overset{\circ}{x} = R_+ \alpha, \text{ where } \alpha = 2\gamma x \overset{\circ}{x} \quad (8)$$

$$\overset{\circ}{R}_- = -R_- \alpha . \quad (9)$$

Let us write the first derivative of the highly elastic part of the deformation with respect to the parameter  $\tau$ :

$$\begin{aligned} \varepsilon_{he}^{\circ} &= \delta[\overset{\circ}{m}_2 + 2\overset{\circ}{m}_3] = \delta\left[\{m_0 R_+ - m_2(2R_+ + R_-) - m_3(R_+ - R_-)\} + 2m_2 R_+ - 2m_3 R_-\right] = \\ &= \delta[m_0 R_+ - m_3(R_+ + R_-) - m_2 R_-] . \end{aligned} \quad (10)$$

Let us write the second derivative of the highly elastic part of the deformation with respect to the parameter  $\tau$ :

$$\varepsilon_{he}^{\circ\circ} = \delta\left\{-\left[\overset{\circ}{m}_3(R_+ + R_-) + \overset{\circ}{m}_2 R_-\right] + \alpha[m_0 R_+ - m_3(R_+ - R_-) + m_2 R_-]\right\} . \quad (11)$$

After substituting  $\overset{\circ}{m}_2$  and  $\overset{\circ}{m}_3$  from system of equations (7) and performing algebraic transformation we obtain

$$\varepsilon_{he}^{\circ\circ} = \delta\left\{\alpha[m_0 R_+ - m_3(R_+ - R_-) + m_2 R_-] - [m_0 A - m_2(A + R_-^2 - R_+^2) - m_3 \cdot 2A]\right\} . \quad (12)$$

In expressions (6) and (10) we select the terms with unknown values and obtain a system of equations with these two unknowns:

$$\begin{cases} \delta(m_2 + 2m_3) = \varepsilon_{he} + \Delta_0 \\ \delta[m_2 R_- + m_3(R_+ + R_-)] = \delta m_0 R_+ - \varepsilon_{he}^{\circ} \end{cases} \quad (13)$$

Solving system (13), we obtain expressions for  $m_2$  and  $m_3$ .

Finally, by means of algebraic transformations and substituting expressions for  $m_2$  and  $m_3$ , we obtain the following second-order differential equation:

$$\begin{aligned} \varepsilon_{he}^{\circ\circ} + \varepsilon_{he}^{\circ} \left[ R_+ - R_- - \frac{\alpha(R_+ + R_-)}{R_+ - R_-} \right] + \varepsilon_{he} \left[ R_+^2 + R_-^2 + A + \frac{2A\alpha}{R_+ - R_-} \right] - \\ - \delta m_0 (2R_+^2 + A) + \Delta_0 (R_+^2 + R_-^2 + A) + \alpha \delta m_0 \cdot 2A - \frac{\alpha \Delta_0 \cdot 2A}{R_+ - R_-} = 0. \end{aligned} \quad (14)$$

Since equation (14) is a differential equation of the second order in time, the solution of this equation in certain cases leads to an oscillatory relaxation mode. Thus, even in a system with one external degree of freedom, when the sample is periodically loaded with a certain frequency, interaction between this external and natural frequency is possible. This fact is supported by experiments on the study of low-amplitude longitudinal vibrations on highly oriented polymer threads in (Gorshkov 2004; Stalevich et al. 2005).

### Separation of the static and dynamic parts of highly elastic deformation in the basic constitutive equation

Let us present equation (14) in the following form:

$$\begin{aligned} (R_+ - R_-) \varepsilon_{he}^{\circ\circ} + \varepsilon_{he}^{\circ} \left[ (R_+ - R_-)^2 - \alpha(R_+ + R_-) \right] + \\ + \varepsilon_{he} \left[ (R_+^2 + R_-^2)(R_+ - R_-) + A(R_+ - R_-) + 2A\alpha \right] = \\ = \delta m_0 (2R_+^2 + A)(R_+ - R_-) - \Delta_0 (R_+^2 + R_-^2 + A)(R_+ - R_-) - \\ - 2\delta m_0 \alpha A (R_+ - R_-) + 2A\alpha \Delta_0. \end{aligned} \quad (15)$$

In the sequel, we will separate the static and dynamic components of this equation. Let us separate the static and dynamic components from the highly elastic part of deformation:

$$\varepsilon_{he} = (\varepsilon^0 - x^0) + (\theta - y) = \varepsilon_{he}^0 + \mu,$$

where  $\varepsilon^0$ ,  $x^0$ ,  $\varepsilon_{he}^0$  are static components of full, elastic and highly elastic deformation; let us denote the dynamic part of highly elastic deformation as  $\mu = (\theta - y)$ .

Taking into account that  $x = x^0 + y$ , expressions for  $R_+$  and  $R_-$  take the form:

$$\begin{aligned} R_+ &= \exp(\gamma x^2) = \exp[\gamma(x^0 + y)^2] = (1 + 2\gamma x^0 y) \cdot \exp[\gamma(x^0)^2], \\ R_- &= A \cdot \exp(-\gamma x^2) = A \cdot \exp[-\gamma(x^0 + y)^2] = (1 - 2\gamma x^0 y) \cdot A \cdot \exp[-\gamma(x^0)^2]. \end{aligned}$$

Let us introduce the notation:  $R_+^0 = \exp[\gamma(x^0)^2]$ ;  $R_-^0 = A \cdot \exp[-\gamma(x^0)^2]$ ;

$$v = 2\gamma x^0 y; \quad \alpha = 2\gamma x \dot{x} = 2\gamma x^0 \dot{y} = \dot{v}.$$

Now let us transform the factors at  $\varepsilon_{he}^{\circ\circ}$ ,  $\varepsilon_{he}^{\circ}$  and  $\varepsilon_{he}$  in equation (15). Firstly, the factor at  $\varepsilon_{he}^{\circ\circ}$ :

$$\begin{aligned} (R_+ - R_-) &= R_+^0 \cdot (1 + 2\gamma x^0 y) - R_-^0 \cdot (1 - 2\gamma x^0 y) = \\ &= R_+^0 \cdot (1 + \nu) - R_-^0 \cdot (1 - \nu) = (R_+^0 - R_-^0) + \nu(R_+^0 + R_-^0). \end{aligned} \tag{16}$$

Secondly, we transform the factor at  $\varepsilon_{he}^{\circ}$  (here and below we will neglect the terms containing  $\nu^2$  and  $\nu \cdot \dot{\nu}$  because of a higher order of smallness):

$$\begin{aligned} (R_+ - R_-)^2 - \alpha(R_+ + R_-) &= (R_+^0 - R_-^0)^2 + 2\left((R_+^0)^2 - (R_-^0)^2\right)\nu - \alpha(R_+^0(1 + \nu) + \\ &+ R_-^0(1 - \nu)) = (R_+^0 - R_-^0)^2 + 2\left((R_+^0)^2 - (R_-^0)^2\right)\nu - (R_+ + R_-) \cdot \dot{\nu}. \end{aligned} \tag{17}$$

Thirdly, we transform the factor at  $\varepsilon_{he}$ :

$$\begin{aligned} (R_+^2 + R_-^2)(R_+ - R_-) + A(R_+ - R_-) + 2A\alpha &= \\ &= \left[ (R_+^0)^2(1 + \nu)^2 + (R_-^0)^2(1 - \nu)^2 \right] \cdot \left[ (R_+^0 - R_-^0) + \nu(R_+^0 + R_-^0) \right] + \\ &+ A \cdot \left[ (R_+^0 - R_-^0) + \nu(R_+^0 + R_-^0) \right] + 2A\dot{\nu} = \\ &= \left[ \left( (R_+^0)^2 + (R_-^0)^2 \right) + 2\left( (R_+^0)^2 - (R_-^0)^2 \right)\nu \right] \cdot \left[ (R_+^0 - R_-^0) + \nu(R_+^0 + R_-^0) \right] + \\ &+ A \cdot \left[ (R_+^0 - R_-^0) + \nu(R_+^0 + R_-^0) \right] + 2A\dot{\nu} = \left( (R_+^0)^2 + (R_-^0)^2 \right) (R_+^0 - R_-^0) + \\ &+ \nu \left[ 2\left( (R_+^0)^2 - (R_-^0)^2 \right) (R_+^0 - R_-^0) + (R_+^0 + R_-^0) \left( (R_+^0)^2 + (R_-^0)^2 \right) \right] + \\ &+ A \cdot \left[ (R_+^0 - R_-^0) + \nu(R_+^0 + R_-^0) \right] + 2A\dot{\nu} = \\ &= \left( (R_+^0)^2 + (R_-^0)^2 \right) (R_+^0 - R_-^0) + \nu(R_+^0 + R_-^0) \left( 3\left( (R_+^0)^2 + (R_-^0)^2 \right) - 4A \right) + \\ &+ A \cdot \left[ (R_+^0 - R_-^0) + \nu(R_+^0 + R_-^0) \right] + 2A\dot{\nu}. \end{aligned} \tag{18}$$

Now let us rewrite the left side of equation (15), substituting the resulting expressions (16), (17), (18) for the factors at  $\varepsilon_{he}^{\circ\circ}$ ,  $\varepsilon_{he}^{\circ}$  and  $\varepsilon_{he}$  and substituting  $\varepsilon_{he} = \varepsilon_{he}^{\circ} + \mu$ . Neglecting the terms containing  $\mu \cdot \nu$  and  $\mu \cdot \dot{\nu}$  as being of a higher order of smallness, we obtain the following on the left side of (15):

$$\begin{aligned} (R_+^0 - R_-^0) \mu + (R_+^0 - R_-^0)^2 \dot{\mu} + \left[ \left( (R_+^0)^2 + (R_-^0)^2 \right) (R_+^0 - R_-^0) + A(R_+^0 - R_-^0) \right] \mu + \\ + \varepsilon_{he}^{\circ} \left[ \left( (R_+^0)^2 + (R_-^0)^2 \right) (R_+^0 - R_-^0) + A(R_+^0 - R_-^0) \right] + \\ + \varepsilon_{he}^{\circ} \left[ \nu(R_+^0 + R_-^0) \left( 3\left( (R_+^0)^2 + (R_-^0)^2 \right) - 4A \right) + A(R_+^0 + R_-^0)\nu + 2A\dot{\nu} \right]. \end{aligned} \tag{19}$$

Let us transform the right side of (15):



$$\begin{aligned}
 & \delta m_0(2R_+^2 + A)(R_+ - R_-) - \Delta_0(R_+^2 + R_-^2 + A)(R_+ - R_-) - \\
 & -2\delta m_0\alpha A(R_+ - R_-) + 2A\alpha\Delta_0 = \delta m_0 \left[ 2(R_+^0)^2(1+\nu)^2 + A \right] \cdot \left[ (R_+^0 - R_-^0) + \nu(R_+^0 + R_-^0) \right] - \\
 & -\Delta_0 \left[ (R_+^0)^2(1+\nu)^2 + (R_-^0)^2(1-\nu)^2 + A \right] \cdot \left[ (R_+^0 - R_-^0) + \nu(R_+^0 + R_-^0) \right] - \\
 & -2\delta m_0 A \overset{\circ}{\nu}(R_+^0 - R_-^0) + 2A\Delta_0 \overset{\circ}{\nu} = \\
 & = \delta m_0 \left[ 2(R_+^0)^2 + A \right] \cdot (R_+^0 - R_-^0) - \Delta_0 \left[ (R_+^0)^2 + (R_-^0)^2 + A \right] (R_+^0 - R_-^0) + \\
 & + \delta m_0 \left\{ 4(R_+^0)^2(R_+^0 - R_-^0) + (2(R_+^0)^2 + A)(R_+^0 + R_-^0) \right\} \nu - \\
 & -\Delta_0 \left\{ (2(R_+^0)^2 - 2(R_-^0)^2)(R_+^0 - R_-^0) + ((R_+^0)^2 + (R_-^0)^2 + A)(R_+^0 + R_-^0) \right\} \nu + \\
 & + \Delta_0 \cdot 2A\overset{\circ}{\nu} - 2\delta m_0 A(R_+^0 - R_-^0) \overset{\circ}{\nu}.
 \end{aligned} \tag{20}$$

Let us consider equation (15), which contains only the dynamic part (we equate the left (19) and the right (20) parts, leaving only the terms with factors in the form  $\mu$  and  $\nu$ , as well as their derivatives). In addition, we take into account that the terms with the multiplier  $\Delta_0$  (initial deformation) will also disappear in the dynamics (this is the damping part). Then we obtain:

$$\begin{aligned}
 & (R_+^0 - R_-^0) \overset{\circ\circ}{\mu} + (R_+^0 - R_-^0)^2 \overset{\circ}{\mu} + (R_+^0 - R_-^0) \left[ (R_+^0)^2 + (R_-^0)^2 + A \right] \mu + \\
 & + \varepsilon_{he}^0 \left[ (R_+^0 + R_-^0) \left( 3 \left( (R_+^0)^2 + (R_-^0)^2 \right) - 4A \right) \nu + A(R_+^0 + R_-^0) \nu + 2A\overset{\circ}{\nu} \right] = \\
 & = \delta m_0 \left\{ 4(R_+^0)^2(R_+^0 - R_-^0) + (2(R_+^0)^2 + A)(R_+^0 + R_-^0) \right\} \nu - 2\delta m_0 A(R_+^0 - R_-^0) \overset{\circ}{\nu}.
 \end{aligned} \tag{21}$$

### Study of low-amplitude longitudinal vibrations for highly oriented polymer threads

Let us consider low-amplitude oscillations in the following system: a load of mass  $m$  is suspended on a thread from the polymer sample under study. Initial thread length is denoted as  $L$ ,  $S_0$  stands for the cross-sectional area,  $\sigma_0$  stress value at equilibrium is designated as  $\sigma_0$  and  $\sigma$  during vibrations,  $E_0$  is elastic modulus,  $Z$  is vertical coordinate  $Z$  (Fig. 5).

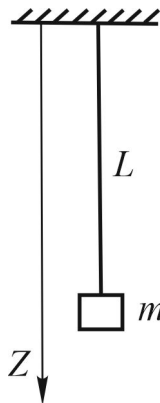


Fig. 5. Load of mass on a suspension (polymer thread) of initial length  $L$

In equilibrium position:  $mg = \sigma_0 S_0$ .

In oscillation mode:  $mg - \sigma_0 S_0 - \delta\sigma S_0 = m\ddot{Z}$ ;  $Z = L \cdot \theta$ ,

where  $\theta$  is the relative deformation.

The value of elastic deformation:

$$x = \frac{\sigma}{E_0} = \frac{\sigma_0}{E_0} + \frac{\delta\sigma}{E_0}; \quad x = x^0 + y.$$

Then for the oscillation mode we obtain the following equation:

$$mL\ddot{\theta} + yE_0S_0 = 0.$$

Dividing the latter by  $mL$ , we bring it to the form:

$$\ddot{\theta} + \Omega_0^2 \cdot y = 0,$$

where the following designation is introduced

$$\Omega_0^2 = \frac{E_0S}{mL}. \tag{22}$$

Thus, a differential equation of harmonic oscillations is obtained, where  $\Omega_0$  is their frequency.

Here, just as before, we will introduce dimensionless variables, ensuring the universality of the curves characterizing various polymer samples. For this purpose, we use the notation:

$$\tilde{\Omega}_0 = \Omega_0 \cdot \tau_\delta. \tag{23}$$

Here, as before,  $\tau = t/\tau_\delta$ , where  $\tau_\delta$  is the relaxation time. Then the vibration equation takes the following form:

$$\ddot{\theta} + \tilde{\Omega}_0^2 \cdot y = 0. \tag{24}$$

Now, just as in Section 3, we apply the following notation:

$$\theta = \mu + y; \quad \nu = 2\gamma x^0 y = Ky; \quad \dot{\nu} = K \dot{y}.$$

Then equation (24) takes the following form:

$$\ddot{\mu} + \ddot{y} + \tilde{\Omega}_0^2 \cdot y = 0$$

Let us substitute and obtain:

$$\ddot{\mu} + \frac{1}{K} \ddot{\nu} + \frac{\tilde{\Omega}_0^2}{K} \cdot \nu = 0. \tag{25}$$

### Analysis of the solution for low-amplitude longitudinal vibrations

Next, we will consider the obtained differential equations: the dynamic part of constitutive equation (21) and equation of small-amplitude oscillations (25). We will look for solutions of these equations in the following form:

$$\mu = \mu_0 \cdot \exp(\lambda \tau),$$

$$\nu = \nu_0 \cdot \exp(\lambda \tau).$$

Then equation (21) after substituting these expressions and simplification takes the form:

$$\begin{aligned} &\lambda^2(R_+^0 - R_-^0)\mu_0 + \lambda(R_+^0 - R_-^0)^2\mu_0 + (R_+^0 - R_-^0)\left[(R_+^0)^2 + (R_-^0)^2 + A\right]\mu_0 + \\ &+ \varepsilon_{he}^0 \left[ (R_+^0 + R_-^0) \left( 3 \left( (R_+^0)^2 + (R_-^0)^2 \right) - 4A \right) + (R_+^0 + R_-^0)A + 2A\lambda \right] \nu_0 = \dots \\ &= M_0 \nu_0 \left\{ 4(R_+^0)^2(R_+^0 - R_-^0) + (2(R_+^0)^2 + A)(R_+^0 + R_-^0) - 2A\lambda(R_+^0 - R_-^0) \right\} \end{aligned}$$

Here and further below the notation  $M_0 = \delta m_0$  is used.

To obtain a more compact representation, we use temporary notation for the coefficients at  $\mu_0$  and  $\nu_0$ . Then the last equation takes the form:

$$\left[ (R_+^0 - R_-^0)\lambda^2 + (R_+^0 - R_-^0)^2\lambda + G \right] \mu_0 + L\nu_0 + N\lambda\nu_0 = 0, \quad (26)$$

where  $G = (R_+^0 - R_-^0) \left[ (R_+^0)^2 + (R_-^0)^2 + A \right]$

$$\begin{aligned} L &= \varepsilon_{he}^0 \left[ (R_+^0 + R_-^0) \left( 3 \left( (R_+^0)^2 + (R_-^0)^2 \right) - 4A \right) + (R_+^0 + R_-^0)A \right] - \\ &- M_0 \left[ 4(R_+^0)^2(R_+^0 - R_-^0) + (2(R_+^0)^2 + A)(R_+^0 + R_-^0) \right] = \\ &= \varepsilon_{he}^0 \left[ 3(R_+^0 + R_-^0) \left[ (R_+^0)^2 + (R_-^0)^2 - A \right] \right] - M_0 \left[ R_+^0 \left[ 6(R_+^0)^2 + (R_-^0)^2 - A \right] \right] \end{aligned}$$

$$N = 2A\varepsilon_{he}^0 + 2A(R_+^0 - R_-^0)M_0.$$

Now consider equation (25), which after substituting solutions and simplification takes the form:

$$K\lambda^2\mu_0 + \left( \lambda^2 + \tilde{\Omega}_0^2 \right) \cdot \nu_0 = 0. \quad (27)$$

Now we, therefore, have a system of algebraic equations (26) and (27) with respect to  $\lambda$ .

We will take into account that in practice we are dealing with polymeric materials for which:

$$R_+^0 = \exp\left[\gamma(x^0)^2\right] \gg 1; \quad R_-^0 = A \cdot \exp\left[-\gamma(x^0)^2\right] \ll 1; \quad A < 1.$$

Then the coefficients  $L$ ,  $G$  and  $N$  in the equation take the form:

$$\begin{aligned} L &= \varepsilon_{he}^0 \left[ 3(R_+^0 + R_-^0) \left[ (R_+^0)^2 + (R_-^0)^2 - A \right] \right] - M_0 \left[ R_+^0 \left[ 6(R_+^0)^2 + (R_-^0)^2 - A \right] \right] = \\ &= \varepsilon_{he}^0 \cdot 3(R_+^0)^3 - M_0 \cdot 6(R_+^0)^3 = 3(R_+^0)^3(\varepsilon_{he}^0 - 2M_0) \end{aligned}$$

$$G = (R_+^0 - R_-^0) \left[ (R_+^0)^2 + (R_-^0)^2 + A \right] \approx (R_+^0)^3$$

$$N = 2A\varepsilon_{he}^0 + 2A(R_+^0 - R_-^0)M_0 = 2A\varepsilon_{he}^0 + 2AM_0R_+^0.$$

Let us rewrite equation (26):

$$\left[ R_+^0\lambda^2 + (R_+^0)^2\lambda + (R_+^0)^3 \right] \mu_0 + 3(R_+^0)^3(\varepsilon_{he}^0 - 2M_0)\nu_0 + 2A(\varepsilon_{he}^0 + M_0R_+^0)\lambda\nu_0 = 0$$

Let us divide everything into  $(R_+^0)^3$ :

$$\left[ \frac{\lambda^2}{(R_+^0)^2} + \frac{\lambda}{R_+^0} + 1 \right] \mu_0 + 3(\varepsilon_{he}^0 - 2M_0)\nu_0 + 2A\nu_0 \frac{\lambda}{(R_+^0)^2} \left( M_0 + \frac{\varepsilon_{he}^0}{R_+^0} \right) = 0.$$

Let us introduce the notation:  $\eta = \frac{\lambda}{(R_+^0)^2}$ . Then we obtain:

$$\left[ (R_+^0)^2\eta^2 + R_+^0\eta + 1 \right] \mu_0 + 3(\varepsilon_{he}^0 - 2M_0)\nu_0 + 2A\nu_0\eta \left( M_0 + \frac{\varepsilon_{he}^0}{R_+^0} \right) = 0. \quad (28)$$

Let us consider equation (27). Let us divide it by  $(R_+^0)^4$  and also use the notation  $\eta = \frac{\lambda}{(R_+^0)^2}$ . Then we obtain:

$$K\eta^2\mu_0 + \left( \eta^2 + \frac{\tilde{\Omega}_0^2}{(R_+^0)^4} \right) \cdot v_0 = 0. \tag{29}$$

Divide equation (28) by equation (29) and obtain:

$$\frac{(R_+^0)^2\eta^2 + R_+^0\eta + 1}{K\eta^2} = \frac{3(\varepsilon_{he}^0 - 2M_0) + 2A\eta \left( M_0 + \frac{\varepsilon_{he}^0}{R_+^0} \right)}{\eta^2 + \frac{\tilde{\Omega}_0^2}{(R_+^0)^4}}. \tag{30}$$

Let us introduce the notation:

$$3(\varepsilon_{he}^0 - 2M_0) = \psi; \quad 2A \left( M_0 + \frac{\varepsilon_{he}^0}{R_+^0} \right) = \varphi; \quad \frac{\tilde{\Omega}_0^2}{(R_+^0)^4} = \pi. \tag{31}$$

Then equation (30) takes the form:

$$\left( (R_+^0)^2\eta^2 + R_+^0\eta + 1 \right) \cdot (\eta^2 + \pi) = K\eta^2 (\psi + \varphi\eta).$$

After that, applying algebraic transformations, we obtain:

$$(R_+^0)^2\eta^4 + (R_+^0 - K\varphi)\eta^3 + (1 + \pi(R_+^0)^2 - K\psi)\eta^2 + R_+^0\pi\eta + \pi = 0.$$

Dividing everything by  $(R_+^0)^2$ , we obtain:

$$\eta^4 + \frac{1}{(R_+^0)^2} (R_+^0 - K\varphi)\eta^3 + \frac{1}{(R_+^0)^2} (1 + \pi(R_+^0)^2 - K\psi)\eta^2 + \frac{\pi}{R_+^0}\eta + \frac{\pi}{(R_+^0)^2} = 0. \tag{32}$$

At this stage, the equation has been obtained, which should have four roots basing on algebraic reasons. We want to find out the conditions for beats occurring in the system under consideration. Beats occur when the system and an external influence have close oscillation frequencies. In this case, the imaginary parts of the roots of the equation are also close in value (we will assume they are equal to simplify the transformations). Then we assume that it is possible to represent equation (32) as the product of two polynomials of the following form:

$$(\eta^2 + 2\beta\eta + \omega^2)(\eta^2 + 2\alpha\eta + \omega^2) = 0.$$

Opening the brackets and performing algebraic transformations, we obtain:

$$\eta^4 + 2(\alpha + \beta)\eta^3 + (2\omega^2 + 4\alpha\beta)\eta^2 + 2\omega^2(\alpha + \beta)\eta + \omega^4 = 0. \tag{33}$$

Comparing the corresponding coefficients of the same powers in equations (32) and (33), we obtain a number of relations. For coefficients of free term, we obtain:

$$\omega^4 = \frac{\pi}{(R_+^0)^2}. \tag{34}$$

Also, it should be noted that when substituting the previously introduced notation (31) for  $\pi = \frac{\tilde{\Omega}_0^2}{(R_+^0)^4}$  into (34), we obtain the expression for  $\omega^2$ :

$$\omega^2 = \frac{\tilde{\Omega}_0}{(R_+^0)^3}. \tag{35}$$

Next, let us equate the coefficients at the first degree of  $\eta$ :

$$2\omega^2(\alpha + \beta) = \frac{\pi}{R_+^0}. \tag{36}$$

Let us divide (34) by (36) and express the sum of the attenuation coefficients  $(\alpha + \beta)$ :

$$\alpha + \beta = \frac{1}{2}\omega^2 R_+^0 = \frac{1}{2} \cdot \frac{\tilde{\Omega}_0}{(R_+^0)^3} \cdot R_+^0 = \frac{1}{2} \cdot \frac{\tilde{\Omega}_0}{(R_+^0)^2}. \tag{37}$$

Let us equate the coefficients at  $\eta^2$ :

$$2\omega^2 + 4\alpha\beta = \frac{1}{(R_+^0)^2} (1 + \pi(R_+^0)^2 - K\psi).$$

Now using algebraic transformations and substituting expressions for  $\pi$  (31) and  $\omega^2$  (35) and then for  $K = 2\gamma x^0$  and  $\psi$  (31), we obtain:

$$\begin{aligned} 4\alpha\beta &= \frac{1}{(R_+^0)^2} \left( 1 - \frac{2\tilde{\Omega}_0}{R_+^0} + \frac{\tilde{\Omega}_0^2}{(R_+^0)^2} \right) - \frac{K\psi}{(R_+^0)^2} = \frac{1}{(R_+^0)^2} \left( 1 - \frac{\tilde{\Omega}_0}{R_+^0} \right)^2 - \frac{K\psi}{(R_+^0)^2} = \\ &= \frac{1}{(R_+^0)^2} \left( 1 - \frac{\tilde{\Omega}_0}{R_+^0} \right)^2 - \frac{6\gamma x^0 (\varepsilon_{he}^0 - 2M_0)}{(R_+^0)^2}. \end{aligned}$$

Then multiplication of coefficients  $\alpha$  and  $\beta$  is equal to

$$\alpha\beta = \frac{1}{4(R_+^0)^2} \left( 1 - \frac{\tilde{\Omega}_0}{R_+^0} \right)^2 - \frac{6\gamma x^0 (\varepsilon_{he}^0 - 2M_0)}{4(R_+^0)^2}. \tag{38}$$

Now we have expressions for sum (37) and product (38) of  $\alpha$  and  $\beta$ . The coefficients  $\alpha$  and  $\beta$  can be easily determined using this system. In (Romanova 1990), a program was proposed for calculating these values.

Let us equate the coefficients at  $\eta^3$ :

$$\frac{1}{(R_+^0)^2} (R_+^0 - K\varphi) = 2(\alpha + \beta).$$

Taking into account the expression  $(\alpha + \beta)$  (37) we obtain:

$$\frac{1}{(R_+^0)^2} (R_+^0 - K\varphi) = 2(\alpha + \beta) = \frac{\tilde{\Omega}_0}{(R_+^0)^2}$$

Then after dividing by  $(R_+^0)^2$  we obtain:

$$\tilde{\Omega}_0 = R_+^0 - K\varphi.$$

Taking into account (31) and  $K = 2\gamma x^0$  we have:

$$\tilde{\Omega}_0 = R_+^0 - K\varphi = R_+^0 - 2\gamma x^0 \cdot 2A \left( M_0 + \frac{\varepsilon_{he}^0}{R_+^0} \right) = R_+^0 - 4A\gamma x^0 \left( M_0 + \frac{\varepsilon_{he}^0}{R_+^0} \right). \tag{39}$$

It can be shown graphically that this equation may have one solution (corresponding to one type of beating), two solutions in some cases (corresponding to two possible loads at which beating occurs), and may also have no solutions (no beating occurs).

## Physical explanation for the presence of beats in a mechanical system “with one degree of freedom”

On the one hand, elastic oscillations occur in the system, on the other hand, according to the barrier model, oscillations of the occupation numbers of energy levels take place. Thus, another vibration mode arises, associated with highly elastic deformation, since highly elastic deformation is determined by the occupation numbers of the corresponding states, which obeys to vibrational relaxation. It means that the system tends to be “Boltzmann distributed”, while under the influence of an external mode this process occurs in an oscillatory mode. At close frequencies the beat effect is observed.

### Conclusions

- 1) The difference between highly oriented polymer materials and low molecular weight compounds is that they have highly elastic deformation associated with the nature of the supramolecular structure. The accepted model is the barrier theory, i. e. the presence of stable states depending on the spatial orientation of clusters of macromolecules. During the transition from one state to another a deformation quantum is released or absorbed.
- 2) Under the influence of an external load, the occupation numbers  $m_N$  begin to change periodically with a certain natural frequency.
- 3) When the frequencies of the external load and the natural frequency coincide, the beating effect is observed.
- 4) Thus, the hidden degree of freedom (see rheological model) is the occupation number of energy states.

### Conflict of Interest

The authors declare that there is no conflict of interest, either existing or potential.

### Author Contributions

All the authors discussed the final work and took part in writing the article.

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