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Localized shear as a quasi-plastic mechanism of momentum transfer in liquids

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Two types of dispersion relations (DRs) in condensed media and associated momentum transfer mechanisms are discussed: gapless DRs corresponding to acoustic modes and DRs with energy (or frequency) gap. Studying the viscoelastic effects in condensed media based on the generalization of the Maxwell-Frenkel approach, the emphasis was placed on the gap states in the momentum transfer mechanisms (GMS), when the k-gap was accompanied by qualitative changes in the DRs type. In this paper, the gap states in the mechanisms of momentum transfer in liquids are associated with the effects of shear elasticity and the formation of collective modes of quasi-plastic shears under conditions of the established type of critical phenomena in the ensembles of shear defects — structural-scaling transitions. It has been found that the formation of collective shear modes, having the character of self-similar solutions of the autosoliton type, accounts for the qualitative change in the dispersion properties corresponding to the quasi-plastic momentum transfer mechanisms operating in the characteristic range of loading times. The results of comparison with the data of original experiments confirm the initiation of quasi-plastic mechanisms of momentum transfer mechanisms operating in dissipative properties.

Keywords: dispersion relations, structural-scaling transitions in defect ensembles, gapped momentum states, hydroluminescence.

1. Introduction

In open dissipative systems the states with "gaps" in the momentum transfer mechanisms (GMS - Gapped Momentum States [1-3]) are traditionally associated with specific dynamic and thermodynamic properties. States with gaped mechanisms of momentum transfer in the k-space can be of crucial importance for properties of such systems as liquids under turbulence, dusty plasma, plastically deformed materials, and biological objects. These states were identified in the study of viscoelastic properties of liquids using the Maxwell-Frenkel approach and were interpreted as the consequence of changes in the dispersion relations (DR). The latter illustrate the transition from the mechanisms determined by the energy (frequency) spectrum to the mechanisms that determine the behavior of the system in the space of wave numbers (k-space) and the corresponding qualitative changes in the momentum transfer mechanism. The fundamental idea of Frenkel's kinetic theory of liquids [4] is that liquid particles, the same as solid oscillate for some time in the vicinity of the equilibrium position and then move by diffusion to the neighboring quasi-equilibrium positions. To describe this process, an average time $\tau_{_{\rm F}}$ between the diffusion jumps was introduced and the existence of shear modes in liquids was predicted at times τ , which are shorter

than the Frenkel time and determined by the characteristic frequency:

$$\omega > \omega_F = \frac{1}{\tau_F},\tag{1}$$

Frenkel qualified the state of a deformable solid as a basic state for describing a liquid as a condensed medium and proposed to determine viscosity using the operator $A = 1 + \tau_r d/dt$.

$$\frac{1}{\eta} \to \frac{1}{\eta} \left(1 + \tau_F \frac{d}{dt} \right), \tag{2}$$

where $\tau_{F} = \eta / G$ is the relaxation time in the Maxwell equation

$$\frac{d\varepsilon}{dt} = \frac{\sigma}{\eta} + \frac{1}{G}\frac{d\sigma}{dt}.$$
(3)

Here ε is the shear strain, σ is the shear stress. The hydrodynamic viscosity is determined after substituting the operator $M = G(1 - A^{-1})$ for the shear modulus *G* into the momentum conservation law, which, in this case, is transformed to the well-known Navier-Stokes equation for the mass velocity ν

$$\eta \frac{\partial^2 \nu}{\partial x^2} = \rho \tau_F \frac{\partial^2 \nu}{\partial t^2} + \rho \frac{\partial \nu}{\partial t}.$$
 (4)

Substituting solution (4) in the form $v = v_0 \exp(i(kx - \omega t))$ leads to the following dispersion relation:

$$G\omega^2 + \omega \frac{i}{\tau_F} - c^2 k^2 = 0, \qquad (5)$$

containing the complex value of the frequency

$$\omega = -\frac{i}{2\tau_F} \pm \sqrt{c^2 k^2 - \frac{1}{4\tau_F^2}},\tag{6}$$

From (6) follows the expression for the real part of ω for $ck>1/(2\tau)$

$$\omega = \sqrt{c^2 k^2 - \frac{1}{4\tau_F^2}},\tag{7}$$

and solution (4) written as

$$v = \exp\left(-2\frac{t}{\tau_F}\right) \exp(i\omega t).$$
(8)

In this case, the "gap" in the *k*-space appears in the part of the spectrum $k > k_{o}$, where

$$k_g = \frac{1}{2c\tau_F}.$$
 (9)

The "gap" in a certain range of the wave numbers k corresponds to a change in the momentum transfer mechanism during the formation of shear modes. The value of k_a is interpreted as an "order parameter", which determines the "hardness" of the transition. A jump in k-space can be associated with a finite length $c\tau_{F}$ of the shear wave propagation in a liquid, which is interpreted as an "elastic" shear commonly observed in a solid $d_{el} = c\tau_{F}$. This definition of a jump in the wave numbers reflects the fact that in liquids there are regions of dynamic behavior of characteristic size $c\tau_{\rm F}$, which experience shear deformation typical of solids. The identification of the jump region in the space of wave numbers, beginning with k_{o} , allows us to introduce an ensemble of dynamic regions of characteristic size $c\tau_{r}$, where shear deformation with a wavelength λ is realized. The range of the shear wave propagation length *d* is defined as $d \approx \lambda \omega \tau_{\mu}$, where $\omega \tau_{E} > 1$. According to $d/\lambda \approx \omega \tau_{E}$, the propagation length d significantly exceeds the wavelength λ in the mode $\omega \tau_{r} > 1$.

It is crucially important that a qualitative change in the dynamics occurs in k-space of wave numbers. This is the consequence of differences in the local nature of relaxation processes (the jump-like movement of particles considered by Frenkel) and the extensional nature of the wave. In liquids, the propagation of shear waves does not imply that the behavior of the entire ensemble of particles will be similar to the behavior in a solid. This is because of the particle displacement to a distance d, where a wave front of a new dissipative nature arises. The observed dynamics corresponds to Frenkel's definition of a liquid as a condensed state that retains the properties of a solid and, as a result, provides the realization of the momentum transfer mechanisms according to scenarios characteristic of a solid with the dynamics of the corresponding structural (thermodynamic) variables. This conclusion was confirmed in [5] by measuring the relaxation spectrum under conditions of shear flow of liquids under the applied shear harmonic perturbations with a frequency of 10⁵ Hz, at which shear elasticity occurred.

The presence of large relaxation times is explained in [6] by the ability of the oriented groups of molecules to perform coordinated displacements and the presence of localized slips between groups of molecules. These localized slips are of a mesoscopic nature (similar to that of dislocations) and represent the change in the symmetry properties of the medium in terms of localization of the distortion tensor [7]. The nature and mechanism of localized shear modes are the subjects of intense research.

2. Collective shear modes in condensed media

Statistical thermodynamics and kinetics of condensed matter with microshears, which were developed in [8], made it possible to establish a special type of critical behavior (structural scaling transitions) accompanied by the formation of collective (autosoliton) shear modes. The established form of the nonequilibrium free energy F of a medium with microshears has the form of a generalized Ginzburg-Landau expansion of the variable characterizing the shear deformation, which determines the average value of the ensemble of microshears and is caused by their interaction:

$$F = \frac{1}{2} A \left(1 - \frac{\delta}{\delta_{c}} \right) p^{2} - \frac{1}{4} B p^{4} + \frac{1}{6} C \left(1 - \frac{\delta}{\delta_{c}} \right) p^{6} - D \sigma p + \frac{1}{2} \chi \left(\nabla_{l} p \right)^{2}, \quad (10)$$

where *A*, *B*, *C*, *D* and χ are the expansion parameters; δ is the structural parameter of the "susceptibility" of the medium to microshears, which depends on the nature of the intermolecular interaction and temperature; σ is the shear component of the stress tensor. Two critical values of the "susceptibility" parameter δ_* , δ_c play the role similar to that of critical temperatures in the Ginzburg-Landau theory of phase transitions [9]. The kinetic equation for the shear parameter is written as

$$\frac{dp}{dt} = -\Gamma_{p} \left(A \left(1 - \frac{\delta}{\delta_{\star}} \right) p - Bp^{3} + C \left(1 - \frac{\delta}{\delta_{c}} \right) p^{5} - D\sigma - \frac{\partial}{\partial x_{l}} \left(\chi \frac{\partial p}{\partial x_{l}} \right) \right), \quad (11)$$

where Γ_p is the kinetic coefficient. Transitions through the critical points δ_* , δ_c are accompanied by the formation of collective modes of shear ensembles, which are of the nature of self-similar solutions. In the range of the structural susceptibility parameter δ_* , δ_c , the self-similar solution has the form of an autosoliton mode, the wavelength and propagation velocity of which are determined by the dynamics of the "orientational" transition in the ensemble of shears ("spinodal decomposition" in the region of the thermodynamic potential metastability — nonequilibrium free energy) [10–12]:

$$p(x-Vt) = \frac{1}{2} \Delta p \Big[1 - \tanh(\zeta) \Big], \quad \zeta = \frac{1}{L_s} (x-Vt),$$
$$L_s = \frac{4}{\Delta p} \Big(2\frac{\chi}{C} \Big)^{1/2}, \quad V_s = \frac{1}{2} \Gamma_p (\chi C)^{1/2} \Delta p.$$
(12)

Solution (12) describes the autowave dynamics as a result of "spinodal decomposition" of the metastability region during the formation of orientationally ordered ensemble of microshears at the wavelength L_s and the wave front velocity V_s . The dependence of the defect dynamics to the self-similar solution (12) reflects a global change in the symmetry in a condensed medium. It was shown in [6] that the existence of the scale L_s is indicative of the formation of one-dimensional topological object (considered in "string theory") with a qualitatively new dynamics, which plays the role of the degree of freedom, triggering a new momentum transfer mechanism.

The relation determining the influence of shear effects on the "viscous" properties of the medium, was obtained in [8,13]

$$\sigma = \eta \, e^{\nu} + \zeta \, \dot{p}, \tag{13}$$

where $e^{\nu} = (e - \dot{\sigma}/G - \dot{p})$ is the "viscous" component of strain rates; η is the dynamic viscosity of the liquid under normal conditions; ζ is the "viscosity" coefficient of the quasi-plastic shear.

Following (13), the effective viscosity of the medium with consideration for the shear effects at times $t \sim \tau_F \gg \eta / G$, can be represented as

$$\eta_{im} = \frac{\sigma}{e} = \eta - (\eta - \zeta) \frac{p}{\dot{\epsilon}}.$$
 (14)

When the strain rates in the interval $\delta_c < \delta < \delta_*$ are "subordinated" to the spectrum of autosoliton collective shear modes (12) at $\dot{\epsilon} \rightarrow \dot{p}$, viscosity tends to the asymptotic value $\eta \rightarrow \eta_a$ (degeneration of the diffusion mechanism of momentum transfer). The established asymptotics, $\eta_a \rightarrow \zeta$ ($\eta_a \approx 10^3 \text{ Pa} \cdot \text{s}$) is apparently for strain rates $\dot{\epsilon} \approx 10^4 - 10^6 \text{ s}^{-1}$ and is the result of the formation of collective shear modes on the wave number scales $k_a \sim 1/L_s$ in [2,3,13]

3. Experimental study

Two types of experiments were carried out to study the mechanisms of momentum transfer and dissipation due to the formation of localized shear modes responsible for the quasi-plastic relaxation mechanism at times significantly exceeding the diffusion times in liquids. The quasi-hydrodynamic mechanism of liquid flows was studied in [14,15] when estimating the relaxation times at the shock wave front. According to these studies, the relaxation times $\tau > 10^{-5}$ s found for water and mercury are 6 orders of magnitude higher than the diffusion times τ_D estimated from the Einstein relation [4]

$$\tau_D = \Delta^2 / 6D_{\rm sd} \sim 10^{-11} \,\rm s, \tag{15}$$

where Δ is the distance between particles, $D_{\rm sd}$ is the self-diffusion coefficient.

The experiment performed for this study was based on the registration of wave fronts by the Doppler interferometry method [16], which allowed us to study the relaxation mechanisms during the propagation of perturbations of the finite amplitude P_0 in a condensed medium under impulse loading. The possibility of the quasi-plastic mechanism of momentum transfer, having the universal power-law nature for a wide class of materials [17-21] in the range of strain rates $\dot{\epsilon} > 10^5 \,\text{s}^{-1}$, was studied for liquids (distilled water, transformer oil) under shock-wave loading using the method of electrical wire explosion and the method of an explosive generator [22, 23]). The wave profiles shown in Fig. 1a, of the free surface velocity were obtained at different distances from the site of the electrical wire explosion. They demonstrate the universal power-law quasi-plastic behavior $\dot{\epsilon}_{\star} \sim P_0^{3.2}$ (Fig. 1b) on the wave front scale L_s and the dependence of the propagation velocity V_s , on the amplitude of the wave pulse. The decrease in the wave front width L_c with an increase in the pulse amplitude P_0 is consistent with the interpretation of k_{a} as the "hardness" parameter of the transition [1]. The range of scales L_c corresponding to the power law can be interpreted in terms of the self-similar (autosoliton) solution (12) and is associated with the range of wave numbers $k \sim L_s^{-1}$, which determines the collective shear mode in the quasi-plastic mechanism of momentum transfer.

The universal power-law nature of the dependence of the strain rate on the pulse amplitude on the wavefront scale L_s reflects the "subordination" of the relaxation properties of the liquid to the autosoliton collective shear deformation mode in the range of wave numbers determined by the pulse



Fig. 1. Wave profiles of free surface velocity in distilled water at different distances from the point of initiation of an electric explosion wire: 1 - 8 mm, 2 - 11 mm, 3 - 18 mm, 4 - 25 mm (a); dependence of the strain rate $\dot{\epsilon}_*$ on the wave front on the pulse amplitude P_0 (b).

amplitude. The scales of wave fronts are estimated using the rise times of the wave pulse $t_L: L_s = t_L (K/\rho)^{1/2}$, where K is the bulk modulus of elasticity of the liquid. The dependences of the scales of wave fronts L_s on the wave pulse amplitude are shown for distilled water and transformer oil in Fig. 2. It should be noted that there is a sharp decrease in the scales L_s with an increase in the wave front velocity (pressure amplitude) for distilled water and relative constancy of the scales for transformer oil. This reflects the difference in widths of the wave number "gaps", where the quasi-plastic mechanisms of momentum transfer are realized.

The second class of experiments involves recording of qualitative changes in the dissipative properties of liquids in intense shear flows at the values of strain rate in the boundary layers $\dot{\epsilon} > 10^5 - 10^7 \text{ s}^{-1}$, which are close to the values at the front of the wave pulse generated by the electrical wire explosion, and high enough to realize a quasi-plastic momentum transfer mechanism [22]. An intense shear flow (hydraulic oil, dynamic viscosity $\eta = 0.04 \text{ Pa} \cdot \text{s}$) was generated in a round channel with a diameter of 0.6 mm in the pressure gradient range $\nabla P \approx 0.7 - 5$ GPa/m (Fig. 3 a). Figure 3 b shows the mean frequency of events *f* recorded with the use of a



Fig. 2. Dependences of the wave front scales L_s on the amplitude of the wave pulse P_a for distilled water (\bigcirc) and transformer oil (×).

а

photomultiplier versus the pressure gradient in the channel. This dependence demonstrates a sharp increase in the intensity of hydroluminescence at the threshold values of the strain rate of ~ 10^5 – 10^6 1/s, corresponding to the Reynolds number of ~ $2.7 \cdot 10^3$ and leading to qualitative changes in the dissipation mechanisms [24, 25].

A sharp increase in hydroluminescence can be explained by changes in the mechanism of momentum transfer and dissipation due to the appearance of collective quasi-plastic shear modes.

The threshold character of qualitative changes in dispersion and, as a consequence, dissipative properties, observed in the range of wave numbers $k \sim L_s^{-1}$, is similar to the effects of anomalous energy absorption (dissipation) in the vicinity of phase transition points [26, 27] during the formation of collective shear (autosoliton) modes in the critical region of the "spinodal metastability decomposition" [13], and is caused by a sharp increase in relaxation times.

4. Discussion of results

Reynolds appeared to be the first who noticed that the non-Newtonian behavior of simple fluids in shear flow can be associated with a nonequilibrium viscoelastic response. This is found to be in agreement with the data demonstrating the asymptotic behavior of viscosity [14,15] at strain rates $\dot{\epsilon} > 10^5 s^{-1}$ and shear elasticity in liquids at frequencies $v \sim 10^5$ Hz [6]. Following Frenkel, the above studies lead to the conclusion that in liquids, as condensed media, it is possible to realize a spectral range corresponding to the relaxation times $\tau \sim 10^{-5}$ s of the coordinated microshear of molecular groups under the action of elastic shear stresses [4]. The collective effects of such interaction are similar to the scenarios of the formation of localized shear modes during plastic deformation [7]. It is also found that the dynamics of these modes is determined by self-similar solutions of the autosoliton nature. The self-similar solution relates the scale of the wave front L_{a} to the velocity of its propagation and the value of the shear strain jump in the "orientational transition" in the ensemble of microshears [10]. The self-similar character of the formation of localizled





shear regions in liquids is confirmed by shock wave loading experiments on liquids showing a universal power law dependence of the strain rate at the wave front on the pressure amplitude at characteristic loading times $\tau \sim 10^{-5}$ s. The spectrum of the wave front scales L_s corresponding to the universal power law dependence determines the range of wave numbers $k \sim L_c^{-1}$ at the boundary of which the transition from the diffusion mechanism of momentum transfer to the mechanism of localized quasi-plastic shear takes place. The "critical" nature of such transition, which manifests itself in the formation of collective shear modes, is accompanied by a sharp change in the dissipative properties of the liquid, which appear as "hydroluminescence" effect as soon as the strain rates in the shear flow reach the threshold values $\dot{\epsilon} \sim 10^5 \, \text{s}^{-1}$. The development of instability leading to global instability of the flow [25] can be related to the appearance of quasi-plastic collective shear modes at the Reynolds numbers $\approx 2.7 \cdot 10^3$. "Subordination" of strain rates to the spectrum of collective shear modes on the wave number scale $k_a \sim L_c^{-1}$ determines the asymptotic value of the quasi-plastic shear viscosity $\eta \approx 10^3$ Pa·s, which was found for strain rates $\dot{\varepsilon} \approx 10^4 - 10^6 \mathrm{s}^{-1}$ [14].

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