## Statistical theory of the boundary friction of atomically flat solid surfaces in the presence of a lubricant layer

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<u>Abstract.</u> A rheological model and a thermodynamic model are proposed for describing the melting of an ultrathin lubricant film between atomically flat solid surfaces. Hysteresis phenomena are considered, allowing for the stress and strain dependence of the lubricant shear modulus. The self-similar regime of lubricant melting is studied taking the additive noncorrelated noise of basic parameters into account. The regions of dry, sliding, and stick-slip friction are determined in the phase diagram. Shear stress time series are obtained by numerically analyzing the Langevin equation and are then subjected to multifractal fluctuation analysis. The dependence of the stationary friction force on the lubricant temperature and on the shear velocity of rubbing surfaces is investigated.

## 1. Introduction

Elucidation of the causes underlying friction between solid bodies is of paramount importance from both the theoretical and the practical standpoints [1-3]. On the one hand, the development of nanotechnologies and creation of new hightech instruments, such as computer memory-storage devices and micromotors, require a deep understanding of friction processes [4–10]. On the other hand, there is still no consistent theory of the phenomenon of friction, despite numerous

A V Khomenko, I A Lyashenko Sumy State University, ul. Rimskogo-Korsakova 2, 40007 Sumy, Ukraine Tel. + 38 (0542) 333 155 E-mail: khom@mss.sumdu.edu.ua, nabla04@ukr.net

Received 16 August 2011, revised 31 May 2012 Uspekhi Fizicheskikh Nauk **182** (10) 1081–1110 (2012) DOI: 10.3367/UFNr.0182.201210f.1081 Translated by Yu V Morozov; edited by A M Semikhatov attempts to create it. Many ongoing physical studies are focused on nanosize systems because their behavior differs from that of classical solid bodies; besides, these studies have important practical implications. For example, friction nodes or atomically flat surfaces separated by thin lubricant layers can be used in position control systems of microdevices for which minimal friction between moving parts is a major design criterion.

In the last 20 years, experimental [1, 2, 4–6, 11–13] and theoretical [1, 14, 15] studies, combined with computer simulations [16–24], have greatly promoted understanding physical mechanisms of sliding friction between rubbing surfaces separated by an ultrathin layer (less than ten atomic diameters) of liquid fluid. It is shown experimentally that the film becomes increasingly thinner as friction continues; first, its physical properties change in a quantitative manner, and then distinctive qualitative changes occur [1, 2, 5, 6, 11], manifested as a non-Newtonian flow mechanism and the substitution of ordinary melting by glass transition (after which the lubricant film nonetheless continues to behave as a fluid). Such behavior, known in tribology as 'mixed lubrication,' is an intermediate friction state characterized by the transition from bulk properties to boundary ones.

In thinner films, the changes are more pronounced because they have a qualitative character. Specifically, such films can undergo first-order phase transitions into solid-like or liquid-like phases [4–6, 17, 18, 24], whose properties (e.g., viscosity) cannot be described in terms applicable to bulk liquids. Thin films exhibit a yield stress that normally characterizes the degree of degradation of a solid body, while their molecular diffusion and relaxation times can be ten orders of magnitude higher than the respective times in bulk fluids or even films that are only slightly thicker.

This review describes the dynamic properties of films in the boundary state, e.g., in the case where transitions occur between different types of dynamic states during sliding. Recent experiments with mica, quartz, metal oxide, and monomolecular surfactant surfaces separated during sliding by various organic fluids and aqueous solutions [4–6, 12, 13] have demonstrated that such phase transitions are the rule rather than the exception. They manifest themselves in the appearance of stick–slip motion similar to the one arising in dry friction between solid surfaces [25], characterized by periodic transitions between two or more dynamic states during stationary sliding.

Our aim in this review is to consider phenomenological models describing the behavior of ultrathin lubricant films in the presence of friction between atomically flat solids with the use of both deterministic and stochastic approaches.

In Section 2, we describe characteristics of the boundary friction state with special reference to the geometry of a tribologic experiment. The main focus in the discussion of various models of the stick–slip behavior is on the concept of phase transitions of molecular layers from solid-like to liquid-like states and vice versa, giving rise to a variety of kinetic friction states, with special reference to the non-Newtonian properties of molecularly thin liquid films. A generalized Stribeck–Hersi diagram is presented to illustrate new tribologic regimes. Theoretical mechanistic and rheologic approaches to the description of the boundary friction state are analyzed. We conclude that the two models allow taking both thermodynamic and shear melting of the lubricant into consideration; for this reason, they are the focus of this review.

In Section 3, we present our nonlinear model of melting of an ultrathin lubricant film based on the concept of a viscoelastic medium possessing heat conductivity and taking hysteresis phenomena and the temperature dependence of viscosity into account. Phase portraits of melting-mode kinetics corresponding to different relations between relaxation times of shear stress, strain, and temperature of the lubricant are presented. Thermal and elastic parameters at which friction decreases are determined.

Stick–slip lubrication dynamics, recently attracting much attention at both the atomic [6, 13, 26, 27] and macroscopic [12, 14, 15] levels, are explained based on the random nature of shear stress and strain components (Section 4). Under real conditions, rubbing bodies have originally rough surfaces exhibiting a system of dimples and projections. The stochastic model of stick–slip friction ensues from the fractal nature of rubbing bodies. It is shown that temperature noise results in self-similar behavior of stress time series (in this case, the time series are multifractal). Therefore, the introduction of random forces (noises) must take account of surface roughness and irregularities that do not appear explicitly in the model.

A thermodynamic model describing a first-order phase transition between structural states of a lubricant in the boundary lubrication state is considered in Section 5. It is shown that the lubricant melts when the temperature increases as well as when a shift of rubbing surfaces after elastic strains (stresses) exceed a critical value. A phase diagram containing dry and fluid friction regions is developed. A mechanical analog of a tribologic system was used to analyze the dependence of the friction force on the lubricant temperature and relative shear rate of rubbing surfaces. The experimentally observable stick–slip friction behavior is described as the main cause of the wear of rubbing parts [28]. Factors responsible for stick-slip friction are elucidated.

# **2.** Phase dynamics of friction of ultrathin lubricant films

## 2.1 Experimental method and a model of phase transitions of the intermittent friction state

Investigations of the properties of ultrathin lubricant layers require special high-tech equipment as dictated by their molecular size. Despite all the difficulties, however, there are experimental units and relevant methods that allow measuring the thickness, viscosity, friction coefficients, shear components of viscous and elastic stresses, and other properties of molecular layers.

A typical setup of a mechanical analog of the tribologic system is shown in Fig. 1. Here, a spring with a stiffness K is connected with a block of mass M placed on a smooth surface and separated from it by an *h*-thick lubricant layer. The free end of the spring is set in motion with a fixed velocity  $V_0$ . The motion of the block creates a friction force F that hampers its movement. In general, the block and spring velocities (V and  $V_0$ , respectively) do not coincide due to the oscillating nature of the force F responsible for the stick–slip motion of the block.



Figure 1. Schematic of a tribologic system.

The simplest method to measure dynamic lubricant parameters in the presence of friction between two solid surfaces is to use two small atomically flat surfaces deposited onto spherical or cylindrical rubbing surfaces. This principle was used in designing a surface force apparatus (SFA) and atomic force microscope [29–33].

The distance between rubbing surfaces is measured in the SFA by optic interference techniques in which white light passes through and normally to the interacting surfaces. The passed beam is focused onto a spectrometer that splits it into components of different wavelengths. The analysis of the resulting spectrum of color bands allows determining the shape of the rubbing surfaces and the distance between them (with an accuracy of 0.1 nm). In addition, the contact area  $A \sim 1 - 10^4 \,\mu\text{m}^2$  can be determined (with an accuracy of  $\pm 5\%$ ). The SFA is also used to measure normal and shear components of the action force and the applied load L (with an accuracy of  $\pm 1\%$  and  $\pm 5\%$ , respectively). The chamber containing the rubbing surfaces is wholly filled with the liquid lubricant of interest, which is not squeezed from between the friction surfaces under the load and thus facilitates conducting the experiment. The above methods were used to carry out experiments with atomically flat mica [5, 6, 12, 13, 34-36], quartz [37], and sapphire [38] surfaces, polymer chains [39], surface active compounds [32, 40], protein compositions [41], and metallic layers deposited on mica surfaces [42] served as lubricants.



Figure 2. Dependence of the total friction force F on time t in an experiment on the friction between two atomically flat surfaces in the surface force apparatus at the onset of motion at the instant t = 0. The lubricant layer consists of (a) chain hexadecane molecules and (b) quasi-spherical molecules of OMCTC [6].

Figure 2a shows typical friction trajectories obtained with an SFA for two sliding mica surfaces separated by a lubricant layer ( $\approx 1$  nm hexadecane). Figure 2b presents the results of an experiment with spherical molecules of octamethylcyclotetrasiloxane (OMCTS) and shows the peaks of the stick-slip behavior representing solid-like and liquid-like film states [4, 18]; these data show first the static friction force  $F_s$  (dry friction) and then the kinetic friction force  $F_k$  (liquid friction). In this experiment, the shear velocity  $V_0$  (see Fig. 1) and temperature are constant. The friction force F monotonically increases at the initial stage, to be replaced thereafter by the stick-slip behavior. As follows from a comparison of Figs 2a and 2b, the ordering of chain molecules takes more time than that of spherical ones, i.e., the latent period of the former is longer [3]. Also worthy of note is the fact that the established stick-slip behavior in the case of spherical molecules is practically stationary (the  $F_s$  and  $F_k$  values do not change with time).

Computer simulation of molecular dynamics has demonstrated that during sliding, a thin intersurface film undergoes a first-order phase transition between solid-like and liquidlike states [17, 18, 43]; this transition may be the cause of the observed stick–slip behavior of the simple isotropic liquid enclosed between two solid crystal surfaces. According to such an interpretation, the stick–slip behavior arises from sharp changes in flow characteristics of the film during transition [18, 24], rather than from their gradual continuous variations. The relevant model shown schematically in Fig. 3a suggests that during the phase transition, the internal friction force varies in a stick–slip behavior is also different, for example, the peaks are sharper, and the behavior may



**Figure 3.** (a) Phase transition model of stick–slip friction in which a liquid film alternately melts and solidifies during the shear between solid crystal surfaces. (b) Time evolution of the friction force *F* at  $T < T_c$  with an increase in the velocity *V* [6].

terminate after a certain critical velocity  $V_c$  is exceeded, yielding to kinetic sliding behavior [44, 45]. We note that the displacement by only two lattice constants during sliding as shown in Fig. 3 is in contrast to a significantly greater one in many practical problems. This type of friction is most frequently observed in experiments with smooth surfaces and liquid lubricants or condensed vapors [4, 6, 16]. Simulation of many properties of ultrathin liquid films [17, 18] undergoing a shift between molecularly flat surfaces has thus far provided the best interpretation of experimental data in the case illustrated by Fig. 2.

The 'shear melting' concept has been introduced to explain the stick–slip motion observed at a constant temperature of the friction surface [15, 22]; Fig. 3 illustrates the stick– slip behavior. An initially solid-like (stick) lubricant is sharply converted into the liquid-like (slip) phase as a result of disordering after a certain critical yield stress  $\sigma$  is exceeded, with the top surface slightly going up. Further motion is accompanied by wall compression under the effect of load *L* (see Fig. 1), and the lubricant sticks again.

As noted above and as is shown in what follows, the two states of a thin intersurface film cannot be identified with bulk liquid or solid phases [6, 13]. The properties of these film states are usually different from those of bulk phases and it is more correct to consider them as solid-like and liquid-like states or, possibly, as dynamic and static 'epitaxial' states, because these states arise only in intersurface films whose properties are determined not only by interactions between lubricant molecules (as in bulk liquids) but also by the constraint and epitaxial interactions between film and surface atoms. Results of molecular dynamics simulation suggest that chain molecules in solid-like and liquid-like (adhesion, sliding) states have a similar structure [24].

To conclude this section, we note that the stick-slip motion behavior with which most of this review deals was observed by Bowden and Tabor more than 50 years ago [2] when they experimented on the influence of the molecular weight of polymers on the friction coefficient. In the stick-slip motion behavior, the sliding rate periodically varies in time, because sliding periods alternate with periods of 'adhesion' between contacting surfaces. However, there is no consistent theory yet to describe all the peculiarities of stick–slip friction. One of the objectives of this review is to describe the stick–slip behavior of boundary friction between two atomically flat solid surfaces separated by a nonpolar lubricant film a few atomic diameters thick, without taking physicochemical processes into account.

## 2.2 Non-Newtonian properties of molecularly thin films and theoretical approaches to their description

Some authors [32] suggest a higher degree of ordering in a lubricant due to wall compression at which the density distribution function becomes more pronounced while the lubricant remains a 'true' liquid. Others have shown in numerical experiments [17, 18] that liquids crystallize in the presence of discrete films as thick as a few molecular layers. It is argued in [6] that both views are well substantiated, because the results of simulating liquids enclosed between smooth structureless surfaces indicate that each molecule is likely to be captured by them without losing the ability to rapidly pass from one surface to the other. Thus, the ordering and the high diffuse mobility normal to the surfaces coexist. There is a probability that even in the presence of long-range order parallel to structural surfaces, the molecules frozen into each of the trapping centers can hop between them (and the surfaces) with rates characteristic of bulk liquids. Evidently, these intersurface films have unique properties, qualitatively and quantitatively distinguishing them from large-volume liquid phases.

As shown in Ref. [11], relaxation times of liquids enclosed between two solid surfaces can increase by many orders of magnitude compared even with relaxation times of liquid films only a few angstroms thicker. Moreover, the processes of molecular reordering in such thin films are correlated at large distances [46]. Recent numerical studies have confirmed the opinion that phase transitions in thin liquid films are associated with coordinated movements of a large number of molecules with different characteristic times [47].

Rather long relaxation times of chain molecules are responsible for 'memory effects'. If rubbing surfaces are stopped for a time  $t_s$  and then set in motion in the same direction and with the same speed, the stationary state needs a time depending on  $t_s$  to set in. In certain cases, however, the stationary state develops instantaneously, as shown in Ref. [6]. For viscous liquids, the frictional flow law holds:

$$\sigma = \eta \dot{\varepsilon} \,, \tag{2.1}$$

where  $\sigma$  is the stress causing the lubricant flow,  $\eta$  is the viscosity, and  $\varepsilon$  is the strain, whose derivative is related to the velocity of motion.

When rubbing surfaces stop ( $\dot{e} = 0$ ), the stresses, in accordance with (2.1), must instantaneously relax to zero. However, such relaxation does not occur because, after a  $t_s$ -long stop, the motion continues with the previously established stick–slip friction behavior [6]. If the stresses had relaxed more quickly, the renewal of the stick–slip behavior after the stop would have been preceded by a monotonic increase in the friction force. The observed feature — inherent only in ultrahigh layers — occurs because the lubricant separating the friction surfaces at rest remains squeezed between them in the same state as before the stop. Therefore, ultrathin lubricant films may exhibit properties of solid bodies at temperatures at which bulk lubricants behave as liquids.

A sharp decrease in the peak friction force F (see Fig. 2) suggests the jump-like character of film melting and accompanying variations of friction. That is why it was noted in Ref. [6] that melting originating at a certain point of the contact zone must rapidly spread over the entire contact zone. This process seems to be analogous to fragile destruction in a solid body, at which a fresh crack rapidly extends over the entire material. Also, this phenomenon resembles a sharp increase in the velocity of grain boundary sliding in non-equilibrium grain boundaries due to the confluence of their liquid-like parts after the critical value of shear stress  $\sigma$  is reached [48].

According to [6], the liquid-like state is characterized by an effective viscosity  $\eta_{\text{eff}}$  that is many orders of magnitude greater than that of a bulk liquid and is non-Newtonian. The non-Newtonian properties of hydrocarbonate films are also manifested in the behavior of the kinetic friction force  $F_k$ independent of sliding velocity. This means that the effective viscosity decreases upon increasing the shear velocity; in other words, a film in the liquid-like state undergoes shear-related thinning [49, 50]. It was shown under different sliding conditions [4, 24] that two surfaces remain separated by several layers of liquid, even if their thickness and structure somewhat change. Moreover, these many-layer liquids undergo dynamic phase transitions of more than one type, manifesting themselves in the existence of various stick–slip motion behaviors in other tribologic systems [4, 11, 16, 21].

In the stick-slip mode, friction forces typically decrease as the temperature increases; stick-slip sliding totally disappears after the temperature exceeds a certain critical value  $T_{\rm c}$ , while the liquid continues to move uniformly in the liquid-like kinetic mode [6]. Static and kinetic friction coefficients also decrease at increased temperatures due to the lowering of the activation energy barrier for molecular jumps at a higher temperature, when the film becomes more liquid-like. On the other hand, such characteristics of stick-slip friction as the ratio of static to kinetic friction force  $(F_s/F_k)$  remain roughly constant [51] or increase only insignificantly with temperature, despite the complete disappearance of the static component of  $F_s$  at a critical temperature  $T_c$ . According to Ref. [6], the abrupt disappearance of static friction at a temperature above  $T_c$  resembles its rapid disappearance after the critical velocity  $V_c$  is exceeded. The higher the sliding velocity, the more liquid-like or 'nonsolid-like' the film becomes and the longer the time  $\tau_0$  needed to solidify again.

According to [6], the validity of the phase transition model is confirmed by the fact that film molecules reach the fully liquid-like state with maximum disorder when the sliding velocity V and temperature T exceed certain high values. Thus, any further increase in V or T cannot further increase disorder at a given load. Because the solidification time  $\tau_0$ depends on film conditions when sliding ceases, the aforesaid answers the question of why  $\tau_0$  becomes V- and T-independent when their certain values are exceeded. These maximum critical values must be other than  $V_c$  and  $T_c$ , corresponding to the cessation of the stick-slip behavior; they are higher than  $V_{\rm c}$  and  $T_{\rm c}$  and can be regarded as the velocities and temperatures at which the order parameter (or the Debye-Waller factor) of the film being shifted [18] drops to a minimum and no longer changes, despite a further increase in *V* or *T*[6].

Peculiarities of the boundary friction state arising have long been the focus of attention of many researchers. In 1902,



Figure 4. Stribeck–Hersi diagram. Solid line shows how the friction force of the lubricant depends on experimental parameters [7, 52].

the German scientist R H Stribeck experimentally found the dependence of the friction coefficient on the rotational velocity of a bearing in the presence of a lubricant [3, 52]. This dependence was later called the Stribeck–Hersi diagram (Fig. 4). It is widely applied in tribology to represent the known friction states for thin lubricant layers. Figure 4 shows the friction force *F* as a function of the argument  $A\eta_b V/h$ , where *A* is the contact area,  $\eta_b$  is the bulk viscosity of the lubricant, and *h* is the lubricant thickness. The emerging mechanical stresses are defined in a standard way,  $\sigma = F/A$ , and the shear strain rate  $\dot{\epsilon} \equiv V/h$ . For thin lubricant layers, the friction force is given by the classical equation for the Couette flow:

$$F = \frac{AV\eta_{\rm b}}{h} = A\eta_{\rm b}\dot{\varepsilon}, \qquad \sigma = \frac{F}{A} = \frac{V\eta_{\rm b}}{h} = \eta_{\rm b}\dot{\varepsilon}. \tag{2.2}$$

The diagram shows the boundary friction state at a small lubricant thickness. This state in the given situation is of great importance because it is realized at the beginning of bearing movement. As the bearing continues to move, the lubricant spreads out, and the hydrodynamic mode develops. The elastohydrodynamic (EHD) state occurs at the boundary between mixed and hydrodynamic lubrication zones. In conformity with the Stribeck-Hersi diagram, boundary lubrication has a high friction coefficient  $\mu$ ; accordingly, this state is characterized by a high dissipation energy accompanied by heating and, possibly, destruction of moving parts. That is why studies of the boundary friction state have important practical applications. The advent of modern instruments made it possible to directly observe processes in the contact zone [31]. Characteristically, recent experiments confirm many conclusions made by researchers in the 20th century.

As shown above, the boundary friction state is responsible for a variety of effects that are not included in the Stribeck– Hersi diagram, however. For this, a new friction map has been proposed [7], in agreement with experimental data [5, 6, 12, 13, 31–42]. This map is the generalized Stribeck–Hersi diagram<sup>1</sup> (Fig. 5). As the load *L* increases, the Newtonian



Figure 5. New friction map exposing peculiarities of the boundary state [7]. The sliding velocity V and the friction force F are measured in arbitrary units;  $10^{-10}$ ,  $10^{10}$  and 0,  $10^{10}$  indicate wide ranges.

flow of a viscous liquid in the EHD mode passes to the boundary state. The maximum friction force occurs at large L and a small lubricant thickness h (static friction  $F_s$ ). If the velocity also increases, the stick–slip behavior sets in and a further increase in V leads to a constant velocity-independent value of the friction coefficient  $\mu$  and, accordingly, to a constant kinetic friction force  $F_k$ .

Non-Newtonian shear thinning is observed at a smaller load in thicker lubricant films. The friction force has a maximum at a point with the Deborah number (the ratio of the natural relaxation time of the boundary lubricant to the sliding time in the stick–slip mode)  $\mathcal{D}_e \sim 1$  [6, 53].

The main friction characteristic relevant in applications is the friction coefficient  $\mu$  relating the friction force to the load *L* applied to the rubbing surfaces [3]:

$$F = \mu L \,. \tag{2.3}$$

This law dates back to 1508, when it was established by Leonardo da Vinci, but the precise formulation was given by Amontons 200 years later, and the law was therefore called the Amontons law. It follows from (2.3) that the friction force is independent of the contact area. In 1785, Coulomb generalized the Amontons law by including the surface adhesion *C*:

$$F = C + \mu L \,. \tag{2.4}$$

According to Amontons–Coulomb law (2.4), friction due to interaction between surfaces exists even at a zero load L. It is especially true for boundary friction when a thin lubricant layer does not completely prevent interaction between the rubbing surfaces. Moreover, friction can partly destroy boundary layers and thereby create direct contact zones for which the first term in the right-hand side of (2.4) becomes essential.

Empirical relations (2.3) and (2.4) do not describe friction processes or mechanisms. Therefore, many researchers have sought to derive them theoretically to obtain a deeper insight into the nature of boundary friction. At the same time, it has become increasingly obvious in light of new experimental data that the Amontons–Coulomb law is by no means always fulfilled, which suggests its approximate character. For

<sup>&</sup>lt;sup>1</sup> Here, the effective viscosity  $\eta_{\text{eff}} = Fh/AV$  not coincident with the bulk viscosity can be found only experimentally, with  $\eta_{\text{eff}}$  being dependent on the shear strain rate  $\dot{e}$  rather than being a constant.

example, Hardy [54] experimented with two planar surfaces separated by a boundary layer of aromatic compounds and showed that the static friction coefficient  $\mu$  decreases with the load. In other words,  $\mu$  is a function of L in (2.3), at variance with the law being considered. Hardy explains this discrepancy by the fact that low pressure is associated with a multimolecular lubricant layer lacking the fluidity inherent in bulk lubricants. Hence, the flow starts after a threshold internal stress is exceeded. Hardy concludes that increasing pressure makes the lubricant layer thinner, which accounts for a change in  $\mu$ ; he introduces the dependence of the friction coefficient on the lubricant layer h. We here focus on such systems and draw attention to the fact that boundary lubricant layers have a yield stress that characterizes destruction in solid bodies.

In Section 3.4, we describe the case of lubricant melting after stress exceeds a certain critical value found analytically. In Section 5, we discuss a quantitative thermodynamic model in which the lubricant thickness h explicitly enters the dependence for the friction force.

Hardy wrote: "The lubricant state is neither liquid nor solid." Modern authors speak in terms of solid-like and liquid-like states differing from equilibrium thermodynamic phases because the microrelief of rubbing surfaces has an appreciable effect on the lubricant by breaking the state symmetry. In this context, Hardy's suggestions are quite justified and agree with the current views of boundary friction. Interestingly, what Hardy observed was a very easy slip [55]. He noticed that rubbing surfaces lubricated with oxyoleic acid slightly deflected from the horizontal plane, which caused spontaneous movements of the top surface. Such rubbing surfaces were difficult to hold in the desired position. If they were left to themselves, the top plate immediately slipped from the bottom one. Later, this behavior was observed by L V Panova (see [3] and the references therein), who studied surfaces with accurately processed boundary layers of stearic acid. This phenomenon is currently called superlubricity and is attracting the attention of an increasingly greater number of researchers seeking ways to minimize the size of systems and designing nanomechanical devices [9, 56].

Hardy's work was continued by Bear and Bowden in experiments on the pressure dependence of the kinetic friction force [57]. These authors identified a number of lubricants and friction surfaces for which the Amontons law is not necessarily satisfied. Based on these observations, Deryagin developed a molecular theory of friction [58] in which the friction force was expressed as

$$F = \mu(Ap_0 + L), \qquad (2.5)$$

where A is the true contact area and  $p_0$  is the specific molecular interaction force. According to Deryagin, the friction force therefore depends on the surface adhesion  $\mu A p_0$  and normal pressure  $\mu L$ , which confirms Amontons– Coulomb law (2.4). Epifanov suggested the following dependence for the friction force [3, 59]:

$$F = \Theta A , \qquad (2.6)$$

$$\Theta = \Theta_0 + k\sigma_N, \qquad (2.7)$$

where  $\Theta$  is the resistance to shear, A is the area of shear, and k is a coefficient. The resistance to shear is in turn determined by the resistance to shear  $\Theta_0$  at a zero normal stress  $\sigma_N$  and by the value of  $\sigma_N$ . Substituting (2.7) in (2.6) yields a binomial

law of friction in the form

$$F = \Theta_0 A + kL \,. \tag{2.8}$$

In Epifanov's theory, an increase in the friction force with the load L is mediated through an increase in the area A.

The last three formulas are important for the discussion of the thermodynamic theory in Section 5. The friction force is defined by formulas (5.18) and (5.22), where it is proportional to the contact area A and does not depend on the load L, because the consideration is confined to specific friction conditions for two atomically flat surfaces separated by a layer of a simple nonpolar lubricant. In that case, the area A is close to the area of the rough surfaces and is independent of the applied pressure. In addition, irrespective of the area of the surfaces, the distance between them remains constant, i.e., the thickness h of the lubricant is independent of time. Modern techniques facilitate the creation of such conditions. Epifanov concludes that boundary friction forces are totally determined by shear resistance but are unrelated to the adhesive strength [3, 59]. Epifanov does not use the notion of the friction coefficient because it makes no sense. Precisely such a case is considered below [cf. (2.6) and (5.18)].

The Amontons law and its interpretations discussed above take no account of the dependence of the friction force on slip velocity. However, the F(V) dependences have a different form for different lubricants, friction surfaces, sliding conditions, etc. The friction may either increase or decrease with the velocity V and may change nonmonotonically. An important contribution to the studies of the velocity dependence of the friction force was made by Bowden [2, 57] and Akhmatov, Sukhov, et al. (see [3] and the references therein). However, modern views of this phenomenon are based on the work of experimenters such as Israelachvili [31] using more sophisticated techniques for direct observations and measurement. Tolstoy and Kaplan found that the dependence F(V) at small slip velocities shows a maximum [60], which is confirmed by our observations (Fig. 19b in Section 5.3).

Most surfaces in technical applications (e.g., friction in bearings) are rough. Bowden suggested the following equation for the friction of rough metallic surfaces separated by boundary lubrication [2]:

$$F = \left| \alpha \tau_{\mathrm{M}} + (1 - \alpha) \tau_{\mathrm{L}} \right| A , \qquad (2.9)$$

where A is the area carrying the load,  $\alpha$  is the fraction of the total area giving rise to metal bridges as a result of squeezing out or destruction of the lubricant layer,  $\tau_{\rm M}$  is the shear resistance of the bridges, and  $\tau_{\rm L}$  is the shear resistance of the lubricant layer. In the case where the surfaces are atomically flat and are separated by a homogeneous lubricant layer of the same thickness,  $\alpha = 0$ ; hence, the formula

$$F = \tau_{\rm L} A \,, \tag{2.10}$$

coincides with relation (5.18) in Section 5. Bowden concludes that the stress  $\tau_L$  is constant for a given substance, regardless of the layer thickness. Based on logical reasoning and experimental data, Akhmatov generalized [3] Bowden formula (2.9) as

$$F = \left[\alpha \tau_{\mathbf{M}}(V, T) + (1 - \alpha) \tau_{\mathbf{L}}(h, V, T)\right] A.$$
(2.11)

We show in what follows that  $\tau_L$  is a function of the temperature *T*, the relative shear rate *V*, and the lubricant thickness *h*, in excellent agreement with Akhmatov's conclusions.

The results of experiments are explained largely based on the phenomenological concept of the phase transition of a lubricant film from a liquid-like state to a solid-like one and back. However, this phase transition, unlike the usual liquid– solid transition, may be of both first and second orders. The latter case occurs when the symmetry of the two film states is altered considerably by the presence of limiting solid bodies and elastic strain [4, 61].

Stick-slip motion is dealt with in several theoretical studies. One of the first presents the fundamental material equation relating the friction force to velocity, coordinates, and a variable playing the role of the order parameter and reflecting the degree of film melting [14]. In another approach [61], phase transitions in a lubricant film are considered in terms of the Landau theory under the assumption of partial thermodynamic equilibrium of a slowly changing order parameter, whose square is equal to the shear modulus.

The understanding of the anomalous features of boundary friction described in the preceding paragraphs was promoted by studies of rheological film properties [5, 6, 12]. Moreover, theoretical representation of lubricants as viscoelastic media having heat capacity was used to the same effect. For example, the experimentally observed behavior of an ultrahigh-lubricant film is described in [15] in terms of the Ginzburg-Landau equation, in which the order parameter defines shear melting [16]. A dynamic phase diagram is constructed, determining the regions of dry, sliding, and stick-slip friction in the temperature-film thickness coordinates. The phase dynamics in the case of adsorbed molecules are considered in Refs [51, 62], emphasizing the importance of shear melting. The study of time series of the friction force allowed representing the relationship between friction at the macrolevel and the formation/breakdown of molecular bonds [23].

Investigations of the influence of noise on friction is of great theoretical and practical importance, because fluctuations in concrete experimental situations critically affect frictional behavior, e.g., by providing conditions for reducing friction [1, 20, 22, 63]. Specifically, thermal noise present in any experiment can transfer an ultrathin lubricant film from a stable solid-like state to a liquid-like one and thereby transform dry friction to sliding or stick—slip behavior. That is why so much attention has been given in recent years to the effect of noise and unintentional impurities on static and dynamic friction [20, 64, 65]. These studies have shown that periodic surfaces are characterized by a lower friction coefficient in the process of sliding than the nonperiodic ones.

# **3.** Nonlinear model of melting of an ultrathin lubricant film

## 3.1 Basic equations

The basis of the proposed approach is the synergetic concept of phase transition [66–70], which is actually a generalization of the Landau phenomenological theory in which a phase transition is described by an order parameter  $\phi$  in powers of which the free energy F is expanded [71]. The equilibrium value of  $\phi$  is given by the condition

$$\frac{\partial F}{\partial \phi} = h \,, \tag{3.1}$$

where *h* is an external field and *F* is the free energy at h = 0.

The process of relaxation to equilibrium is described by the equation [72]

$$\dot{\phi} = -\frac{1}{\eta} \left( \frac{\partial F}{\partial \phi} - h \right), \tag{3.2}$$

where  $\eta$  is the kinetic coefficient, which can be regarded as a generalized viscosity. If  $\phi$  is close to the equilibrium value  $\phi_0 = 0$ , the linear approximation  $\partial F/\partial \phi \approx \phi/\chi$  is used, where  $\chi \equiv \partial \phi/\partial h = (\partial^2 F/\partial \phi^2)^{-1}$  is the susceptibility. As a result, relaxation equation (3.2) linearizes into

$$\tau \dot{\phi} = -\phi + \chi h \,, \tag{3.3}$$

where

$$\tau = \chi \eta \tag{3.4}$$

is the relaxation time.

Landau and Khalatnikov used expressions (3.3) and (3.4) to study anomalous sound absorption near a second-order transition point [73], where, as they supposed, the susceptibility tends to infinity,  $\chi \to \infty$ , and the viscosity  $\eta$  is practically independent of the temperature *T*. In the theory of Landau and Khalatnikov, the abnormally high value of  $\tau$  corresponds to the freezing process.

For a viscoelastic medium, the shear modulus G plays the role of the inverse susceptibility. In the case of the viscoelastic transition and a displacement-type (martensitic) transition, the modulus G tends to zero near the transition point, and the relaxation time tends to infinity [74, 75]. Some authors (see, e.g., [76]) used the fact that the generalized susceptibility is virtually independent of temperature and the viscosity increases appreciably at the glass-transition temperature. Hence, the abnormally high susceptibility characterizes phase transitions, while the abnormally high kinetic coefficient (viscosity) is associated with freezing.

Under experimental conditions, lubricants are viscous liquids behaving like amorphous solids; they are characterized by high effective viscosity and yield stress [6, 77, 78]. Bearing this in mind and based on the rheologic description of a viscoelastic medium exhibiting the property of heat conductivity, we obtained a system of kinetic equations describing the mutually correlated behavior of shear stresses  $\sigma$  and strains  $\varepsilon$  as well as temperature T, in an ultrathin lubricant film during friction of atomically flat mica surfaces [69, 79]. The main assumption of the proposed approach is that the relaxation equation for  $\sigma$  has a mathematical structure similar to that of Landau–Khalatni-kov equation (3.3):

$$\tau_{\sigma}\dot{\sigma} = -\sigma + G\varepsilon. \tag{3.5}$$

Here, the first term in the right-hand side describes Debye relaxation during the time  $\tau_{\sigma} \equiv \eta_{\sigma}/G$  determined by the effective viscosity  $\eta_{\sigma}$  and the unrelaxed shear modulus  $G \equiv G(\omega)|_{\omega \to \infty}$ . By the substitution  $\varepsilon/\tau_{\sigma} \to d\varepsilon/dt$ , kinetic equation (3.5) is reduced to a Maxwell-type relation for a viscoelastic medium widely used in the boundary friction theory [1, 80]. In the stationary case  $\dot{\sigma} = 0$ , Eqn (3.5) reduces to Hooke's law  $\sigma = G\varepsilon$ .

The relaxation behavior of a viscoelastic lubricant in the friction process is also described by the Kelvin–Voight equation [77, 78]

$$\dot{\varepsilon} = -\frac{\varepsilon}{\tau_{\varepsilon}} + \frac{\sigma}{\eta} \,, \tag{3.6}$$

where  $\tau_{\varepsilon}$  is the strain relaxation time and  $\eta$  is shear viscosity. The second term in the right-hand side describes the flow of a viscous liquid under the effect of the shear stress component. In the stationary case  $\dot{\varepsilon} = 0$ , the expression  $\sigma = G_{\varepsilon}\varepsilon$  resembling Hooke's law is obtained, where  $G_{\varepsilon} \equiv \eta/\tau_{\varepsilon} \equiv G(\omega)|_{\omega \to 0}$  is the relaxed shear modulus. Because (3.5) does not formally reduce to Kelvin–Voight equation (3.6) [77, 78, 80], we suppose that the effective  $\eta_{\sigma} \equiv \tau_{\sigma}G$  and real  $\eta$  viscosities do not coincide. Moreover, we accept the simplest approximations for temperature dependences  $G_{\varepsilon}(T)$ , G(T),  $\eta_{\sigma}(T) = \text{const:}$ 

$$\eta = \frac{\eta_0}{T/T_c - 1} \,, \tag{3.7}$$

where  $\eta_0$  is the characteristic value of the viscosity  $\eta$  at  $T = 2T_c$  due to the very weak temperature dependence of  $G_{\varepsilon}$ , G, and  $\eta_{\sigma}$ , while the real viscosity  $\eta$  tends to infinity as the temperature decreases to the critical value  $T_c$  [70, 76, 81]. Taken together, Eqns (3.5)–(3.7) make up a new rheological model, because they reduce to a second-order differential equation for the stress  $\sigma$  and strain  $\varepsilon$ . We emphasize that the rheologic properties of lubricant films are amenable to experimental study, which allows constructing the phase diagram [5, 6].

The synergetic concept [66–70] implies the necessity of a kinetic equation for the temperature to supplement the system of equations (3.5), (3.6) containing the order parameter  $\sigma$ , the conjugate field  $\varepsilon$ , and the driving parameter T. This equation can be derived from the basic relations of elasticity theory [80]. We start from the expression relating the time derivative of the entropy S and the internal energy U to the equilibrium elastic stress  $\sigma_{el}$ :

$$T\frac{\mathrm{d}S}{\mathrm{d}t} = \frac{\mathrm{d}U}{\mathrm{d}t} - \sigma_{\mathrm{el}}\frac{\mathrm{d}\varepsilon}{\mathrm{d}t}$$
(3.8)

(a change in the amount of heat  $\delta Q = T \delta S$  in equilibrium). In the nonequilibrium case of nonuniform heating of the medium, this relation takes the form

$$-\operatorname{div} \mathbf{q} = \frac{\mathrm{d}U}{\mathrm{d}t} - \sigma \,\frac{\mathrm{d}\varepsilon}{\mathrm{d}t}\,,\tag{3.9}$$

where the heat flux is given by the Onsager relation

$$\mathbf{q} = -\kappa \nabla T \tag{3.10}$$

with  $\kappa$  being the heat conductivity coefficient and the total stress  $\sigma = \sigma_{el} + \sigma_v$  including the viscous component  $\sigma_v$ . Subtracting (3.9) from (3.8), using the relations

$$\frac{\mathrm{d}S}{\mathrm{d}t} = \frac{\partial S}{\partial U} \left( \frac{\partial U}{\partial T} \right)_{\varepsilon} \frac{\mathrm{d}T}{\mathrm{d}t} + \frac{\partial S}{\partial U} \frac{\partial U}{\partial \varepsilon} \frac{\mathrm{d}\varepsilon}{\mathrm{d}t} + \left( \frac{\partial S}{\partial \varepsilon} \right)_{U} \frac{\mathrm{d}\varepsilon}{\mathrm{d}t} = \frac{\rho c_{v}}{T} \frac{\mathrm{d}T}{\mathrm{d}t} + \frac{1}{T} \frac{\partial U}{\partial \varepsilon} \frac{\mathrm{d}\varepsilon}{\mathrm{d}t} - \frac{\sigma_{\mathrm{el}}}{T} \frac{\mathrm{d}\varepsilon}{\mathrm{d}t} , \qquad (3.11)$$

and assuming that the lubricant layer and the atomically flat mica surfaces have different temperatures T and  $T_e$ , we obtain

$$\rho c_v \dot{T} = \frac{\kappa}{h^2} (T_e - T) + \sigma_v \dot{\varepsilon} + T \frac{\partial \sigma_{el}}{\partial T} \dot{\varepsilon}. \qquad (3.12)$$

Here,  $\rho$  is the lubricant density,  $c_v$  is the specific heat capacity, and h is the lubricant thickness; also, the equalities  $(\kappa/h^2)(T_e - T) \approx -\text{div} \mathbf{q}$  and  $\partial U/\partial \varepsilon = \sigma_{el} - T \partial \sigma_{el}/\partial T$  are used. The first term in the right-hand side of (3.12) describes heat transfer from the lubricant layer to the friction surfaces. The second term accounts for the dissipative heating of the viscous liquid flowing under the effect of stresses [82, 83]. The third term is a heat source governed by the reversible mechanocaloric effect, for which  $T(\partial \sigma_{el}/\partial T)\dot{k} \approx \sigma_{el}\dot{k}$  in the linear approximation. As a result, the heat conductivity equation becomes

$$\rho c_v \dot{T} = \frac{\kappa}{h^2} (T_e - T) + \sigma \dot{\varepsilon} \,. \tag{3.13}$$

We introduce measurement units

$$\sigma_{\rm s} = \left(\frac{\rho c_v \eta_0 T_{\rm c}}{\tau_T}\right)^{1/2}, \quad \varepsilon_{\rm s} = \frac{\sigma_{\rm s}}{G_0} \equiv \tau_{\varepsilon} \left(\frac{\rho c_v T_{\rm c}}{\eta_0 \tau_T}\right)^{1/2}, \quad T_{\rm c} \qquad (3.14)$$

for the respective variables  $\sigma$ ,  $\varepsilon$ , and T ( $G_0 = \eta_0/\tau_{\varepsilon}$ , and  $\tau_T \equiv \rho h^2 c_v/\kappa$  is the heat transfer time). Then the substitution of  $\dot{\varepsilon}$  expressed from (3.6) in (3.13) takes the basic equations (3.5), (3.6), and (3.13) to the form

$$\tau_{\sigma}\dot{\sigma} = -\sigma + g\varepsilon, \qquad (3.15)$$

$$\tau_{\varepsilon}\dot{\varepsilon} = -\varepsilon + (T-1)\sigma, \qquad (3.16)$$

$$\tau_T \dot{T} = (T_e - T) - \sigma \varepsilon + \sigma^2, \qquad (3.17)$$

with the constant  $g = G/G_0$ . Equations (3.15)–(3.17) formally coincide with the synergetic Lorentz system [66–68] used to describe phase thermodynamic and kinetic transformations. Positive feedback between stresses  $\sigma$  and temperature T, on the one hand, and the strain  $\varepsilon$  in (3.16) on the other is responsible for the melting that causes an increase in  $\varepsilon$  related to the instability of the solid-like phase. At the same time, the negative feedback between stresses  $\sigma$ , the strain  $\varepsilon$ , and the temperature T in (3.17) reflects the Le Chatelier principle for the problem being considered, because it leads to a decrease in T resulting from the self-organization process.

The aforementioned positive feedback of  $\sigma$  and T on  $\varepsilon$ means that the solid-liquid transition of lubricants is due to both heating and stresses created by solid surfaces in the presence of friction [69]. This agrees with the consideration of the solid-phase instability in the framework of disorderinduced shear dynamic melting in the absence of thermal fluctuations [15]. The thermal mean root square deviation of molecules (atoms) is given by

$$\langle u^2 \rangle = \frac{T}{Ga} \,, \tag{3.18}$$

where a is the lattice constant or the intermolecular distance. The mean displacement caused by the shear is found from the relation

$$\langle u^2 \rangle = \frac{\sigma^2 a^2}{G^2} \,. \tag{3.19}$$

Thus, the proposed approach suggests that the solid– liquid transition of a lubricant is a result of thermodynamic and shear melting. We suppose that the lubricant film becomes more liquid-like and the friction force decreases with increasing temperature owing to a reduction in the molecular jump activation energy.

## 3.2 Transition conditions

As shown in experiment, the stress relaxation time  $\tau_{\sigma} \sim 10^{-10}$  s for organic lubricants at normal pressure [5, 6]

increases by a few orders of magnitude and reaches macroscopic values as the pressure increases. Shear strain takes a microscopic time  $\tau_{\varepsilon} \approx a/c \approx 10^{-12}$  s to relax, where  $a \approx 1$  nm; the speed of sound is  $c \approx 10^3$  m s<sup>-1</sup>. Because an ultrathin lubricant film contains less than ten molecular layers, the temperature relaxes to  $T_e$  in a time  $\tau_T \ll \tau_{\sigma}$ . This permits analyzing system (3.15)–(3.17) in the framework of the adiabatic approximation, in which the characteristic time scales satisfy the inequalities

$$\tau_{\varepsilon} \ll \tau_{\sigma}, \quad \tau_T \ll \tau_{\sigma}.$$
 (3.20)

These inequalities suggest that both the strain  $\varepsilon(t)$  and the temperature T(t) follow the stress variations  $\sigma(t)$ . Based on the definition of the thermal conductivity  $\chi \equiv \kappa/c_v$ , the effective kinematic viscosity  $v_{\sigma} \equiv \eta_{\sigma}/\rho$ , and the speed of sound  $c \equiv (G/\rho)^{1/2}$ , the second condition in (3.20) can be conveniently written in the form

$$h \ll L \,, \tag{3.21}$$

according to which the lubricant thickness must not be greater than

$$L = \sqrt{\frac{\chi v_{\sigma}}{\rho c^2}} \,. \tag{3.22}$$

Then the left-hand sides of Eqns (3.16) and (3.17) can be equated to zero and the Landau–Khalatnikov equation can be obtained from (3.15) [72, 73]:

$$\tau_{\sigma}\dot{\sigma} = -\frac{\partial V}{\partial\sigma}\,,\tag{3.23}$$

where the synergetic potential has the form

$$V = \frac{1}{2}(1-g)\sigma^2 + g\left(1-\frac{T_e}{2}\right)\ln(1+\sigma^2).$$
 (3.24)

In the stationary state  $\dot{\sigma} = 0$ , potential (3.24) has a minimal value. At a temperature  $T_e$  below the critical value

$$T_{\rm c0} = 1 + g^{-1}, \qquad g \equiv \frac{G}{G_0} < 1, \qquad G_0 \equiv \frac{\eta_0}{\tau_{\varepsilon}}, \qquad (3.25)$$

this minimum corresponds to  $\sigma = 0$ ; in this case, there is no melting and a solid-like state of the lubricant is realized. In the opposite case  $T_e > T_{c0}$ , the stationary stress value is nonzero:

$$\sigma_0 = \left[\frac{gT_e - (g+1)}{1-g}\right]^{1/2},$$
(3.26)

and increases as the square root of  $T_{e}$ , leading to the melting of the lubricant and its transition to a liquid-like state. The corresponding stationary strain and temperature values are

$$\varepsilon_0 = \frac{\sigma_0}{g}, \qquad T_0 = 1 + g^{-1}.$$
 (3.27)

We note that the stationary temperature value  $T_0$ , on the one hand, coincides with the value in (3.25) but, on the other hand, differs from  $T_e$  because the first equality in (3.27) is realized instead of the relation  $\sigma_0 = \varepsilon_0$  in the stationary state. This means ( $T_{c0}$  being the minimal temperature at which solid-liquid transition begins) that the negative feedback of shear stresses  $\sigma$  and strain  $\varepsilon$  on the temperature T [see the third term in the right-hand side of Eqn (3.17)] decreases the lubricant temperature, and hence the self-organization process is maintained only in the limit. In the stationary state, the viscosity during melting becomes  $\eta_m = \eta_0 g$ .

The solid-like state of a lubricant corresponds to shear stresses  $\sigma = 0$ , because Eqn (3.15) falls out of consideration. Equation (3.16), containing viscous stresses, reduces to the Debye law describing fast strain relaxation within a microscopic time  $\tau_{\varepsilon} \sim 10^{-12}$  s. Thermal conductivity equation (3.17) takes the form of the simplest expression for temperature relaxation that contains no terms describing dissipative heating or the mechanocaloric effect of a viscous medium.

According to (3.16), the appearance of viscous stresses  $\sigma_v$  gives rise to a plastic flow of a liquid-like lubricant with the speed  $V = h \partial \varepsilon / \partial t$  [69]. Specifically, in the case of a surface force apparatus [31, 84, 85], the effective strain amplitude  $\varepsilon = x_{\text{max}}/h$  in Eqns (3.15)–(3.17) is defined as the ratio of the strain (deflection) amplitude  $x_{\text{max}}$  to the thickness *h*. The effective shear rate

$$\dot{\varepsilon} = \varepsilon \omega = \frac{V}{h} = \frac{\varepsilon}{\tau_{\sigma}} \tag{3.28}$$

is the product of the strain  $\varepsilon$  and the fluctuation frequency  $\omega$ . In our problem,  $\varepsilon$  must be understood as the sum of elastic and viscous components of shear strain.

It was shown in Refs [61, 84] that the plastic flow of a lubricant layer occurs in the presence of elastic stresses, while the effects of shear stresses account for the reduced shear modulus of the lubricant [86]. According to [7], the enhancement of viscous strains

$$\sigma_{\rm v} = \frac{F_{\rm v}}{A} \tag{3.29}$$

in the boundary friction regime leads to an increase in the viscous friction force

$$F_{\rm v} = \frac{\eta_{\rm eff} V A}{h} \,, \tag{3.30}$$

where  $\eta_{\text{eff}}$  is the effective viscosity and A is the contact area.

A comparison of (3.29) and (3.30) gives the expression for the velocity in terms of stresses:

$$V = \frac{\sigma_{\rm v} h}{\eta_{\rm eff}} \,. \tag{3.31}$$

Therefore, enhanced stresses increase the relative displacement rate of the surfaces and cause the lubricant to melt. This inference is consistent with Maxwell-type relations between stresses and strains:  $\partial \sigma / \partial t = -\sigma / \tau_{\sigma} + G \partial \varepsilon / \partial t$ .

## **3.3** Hysteresis phenomena in the stress dependence of the shear modulus

**3.3.1 Jump-like melting.** The Maxwell equation implies the use of an idealized Hankey model. For the strain dependence of stresses, this model is represented by the Hooke law  $\sigma = G\varepsilon$  for  $\varepsilon < \varepsilon_m$  and a constant  $\sigma_m = G\varepsilon_m$  at  $\varepsilon \ge \varepsilon_m$  [ $\sigma_m$  and  $\varepsilon_m$  are the maximum values of elastic shear stresses and strains;  $\sigma > \sigma_m$  leads to a viscous flow with the strain rate  $\dot{\varepsilon} = (\sigma - \sigma_m)/\eta$ ]. In fact, the simplest curve  $\sigma(\varepsilon)$  has two parts: the steep Hookean section specified by the shear modulus *G* is followed by a gently sloping portion corresponding to plastic deformation, whose slope is determined by the strengthening factor  $\Theta < G$ . Evidently, such a picture

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means that the lubricant shear modulus entering Eqn (3.5) depends on the stress value [61, 86]. We use the simplest approximation

$$G(\sigma) = \Theta + \frac{G - \Theta}{1 + (\sigma/\sigma_{\rm p})^{\beta}}$$
(3.32)

(where  $\sigma_p$  is the characteristic value of shear stress) describing the aforementioned transition of the elastic strain state to the plastic one. This transition is taken into account by substituting the constant coefficient g in (3.15) by dependence (3.32) in the dimensionless form  $g(\sigma) = G(\sigma)/G_0$ :

$$g(\sigma) = g_{\theta} \left[ 1 + \frac{\theta^{-1} - 1}{1 + (\sigma/\alpha)^{\beta}} \right], \qquad (3.33)$$

where  $g_{\theta} = \Theta/G_0 < 1$ ,  $\alpha = \sigma_p/\sigma_s < 1$ , and  $\theta = \Theta/G < 1$  is the parameter determining the ratio of the slopes of the plastic and Hookean sections of the strain curve. An expression of type (3.33), proposed for the first time by Haken [66] to describe the solid laser radiation mode, was used in earlier studies [68] for representing first-order phase transition kinetics. However, that expression contained the square of the ratio  $\sigma/\sigma_p$  ( $\beta = 2$ ), and the resulting dependence of the synergetic potential on the order parameter  $V(\sigma)$  was an even function. The description of structural phase transitions in a liquid includes third-order invariants responsible for parity breaking [71]. For this reason, the linear term  $\sigma/\sigma_p$  ( $\beta = 1$ ) is used instead of the quadratic one  $(\sigma/\sigma_p)^2$  ( $\beta = 2$ ) in approximation (3.33). We note that dependence (3.40) given below is no longer even.<sup>2</sup>

A more comprehensive analytic study was carried out for  $\beta = 2$ , and we present this case in more detail (the positive- $\sigma$  region in Fig. 6). In the framework of adiabatic approximation (3.20), the system of Lorentz equations (3.15)–(3.17) in which the dependence  $g(\sigma)$  must be used instead of g reduces to Landau–Khalatnikov equation (3.23), as before. But the coefficient  $g = G/G_0$  in synergetic potential (3.24) is replaced by the parameter  $g_{\theta} = \Theta/G_0 < 1$  and a term proportional to  $\theta^{-1} - 1$  appears:

$$V = \frac{1}{2}(1 - g_{\theta})\sigma^{2} + g_{\theta}\left(1 - \frac{T_{e}}{2}\right)\ln(1 + \sigma^{2})$$
  
$$-\frac{1}{2}g_{\theta}\alpha^{2}(\theta^{-1} - 1)(\alpha^{2} - 1)^{-1}$$
  
$$\times \left[(T_{e} - 2)\ln(1 + \sigma^{2}) + (\alpha^{2} - T_{e} + 1)\ln\left(1 + \frac{\sigma^{2}}{\alpha^{2}}\right)\right].$$
  
(3.34)

The dependence  $V(\sigma)$  in (3.34) increases monotonically at small  $T_e$  and has a minimum at  $\sigma = 0$  corresponding to the stationary solid-like state (curve *I*). Figure 6 shows the appearance of a plateau (curve *2*) at

$$T_{\rm c}^{0} = 1 + g_{\theta}^{-1} + \frac{\alpha^{2}}{\theta g_{\theta}} (\theta + g_{\theta} - 2) + \frac{2\alpha}{\theta g_{\theta}} D^{1/2}, \qquad (3.35)$$

$$D = (\theta - 1)(\alpha^{2} - \theta)(g_{\theta} - 1), \qquad (3.36)$$

and at  $T_{\rm e} > T_{\rm c}^0$  it passes to a minimum corresponding to a stress  $\sigma_0 \neq 0$  and to a maximum  $\sigma^{\rm m}$  separating the minima at



**Figure 6.** The synergetic potential in (3.40) plotted versus stress for the first-order phase transition at  $g_{\theta} = 0.6$ ,  $\theta = 0.1$ , and  $\alpha = 0.75$  at temperatures  $T_{\rm e} < T_{\rm c}^0$  (curve 1),  $T_{\rm e} = T_{\rm c}^0$  (curve 2),  $T_{\rm c}^0 < T_{\rm c} < T_{\rm c0}$  (curve 3), and  $T_{\rm e} \ge T_{\rm c0}$  (curve 4).



**Figure 7.** Temperature  $T_e$  dependence of stationary stress values at  $g_{\theta} = 0.6$ ,  $\theta = 0.1$ , and  $\alpha = 0.75$  (solid curve corresponds to the stable value of  $\sigma_0$ , dashed line denotes an unstable  $\sigma^{\rm m}$  value).

the points  $\sigma = 0$  and  $\sigma = \sigma_0$  (curve 3). As the temperature  $T_e$  increases further, the minimum of the 'ordered' phase corresponding to the liquid-like state  $\sigma = \sigma_0$  becomes deeper, while the height of the interphase barrier decreases; it disappears at the critical value  $T_{c0} = 1 + \theta/g_{\theta}$  in (3.25). The stationary values of shear stresses in the liquid-like state are (see Figs 6 and 7)

$$\sigma_0^{\rm m} = \left\{ \frac{1}{2} \, \alpha^2 (g_\theta - 1)^{-1} \\ \times \left[ 1 + \alpha^{-2} - g_\theta \alpha^{-2} (T_{\rm e} - 1) - g_\theta \theta^{-1} \pm D_0^{1/2} \right] \right\}^{1/2}, \quad (3.37)$$

$$D_0 = \left[ g_\theta (T_{\rm e} - 1) \alpha^{-2} + g_\theta \theta^{-1} - 1 - \alpha^{-2} \right]^2$$

$$-4\alpha^{-2}(g_{\theta}-1)[g_{\theta}\theta^{-1}(T_{e}-1)-1], \qquad (3.38)$$

where the upper and lower signs are respectively for the maximum and minimum of  $V(\sigma)$ . At  $T_e \ge T_{c0}$ , the depen-

<sup>&</sup>lt;sup>2</sup> Odd values of  $\beta$  correspond to the experiment in which the block slips in opposite directions that are not equivalent from the energy standpoint.

A characteristic feature of our approach is the energy barrier inherent in the first-order synergistic transition, i.e., the melting of a crystalline lubricant. It is observed only in the presence of a shear modulus defect. This situation is more complicated than usual phase transitions: indeed, the stationary temperature of the system  $T_0$  standardly reduces to a value  $T_e$  determined by the thermostat. In our case,  $T_0$ reduces to the critical value  $T_{c0}$  of the second-order synergetic transition (melting of an amorphous lubricant) that occurs in the absence of a modulus defect (see Section 3.2). When the modulus defect is taken into account, the temperature

$$T_0 = T_e + (2 - T_e) \frac{\sigma_0^2}{1 + \sigma_0^2}, \qquad (3.39)$$

specified by the position of the minimum of dependence (3.34), (3.40) is realized. According to (3.37)–(3.39), the stationary system temperature  $T_0$  gradually increases from  $T_{\rm m}$  at  $T_{\rm e} = T_{\rm c}^0$ . The stationary temperature  $T_0$  increases linearly from 0 to  $T_{\rm c0}$  as the temperature  $T_{\rm e}$  increases within the same range. At  $T_{\rm e} = T_{\rm c0}$ , the value of  $T_0$  increases smoothly after a jump upward. The stationary temperature  $T_0$  decreases after a decrease in  $T_{\rm e}$ . As soon as the point  $T_{\rm c}^0$  is reached [see (3.35)], the value of  $T_0$  falls abruptly from  $T_{\rm m}$  to  $T_{\rm c}^0$ . At  $T_{\rm e} < T_{\rm c}^0$ , the stationary temperature  $T_0$  becomes equal to  $T_{\rm c}$  again.

In the case  $\beta = 1$ , the synergetic potential is an odd function of  $\sigma$  given by the expression (see Fig. 6)

$$V = \frac{1}{2}(1 - g_{\theta})\sigma^{2} + g_{\theta}\left(1 - \frac{T_{e}}{2}\right)\ln(1 + \sigma^{2}) - g_{\theta}(\theta^{-1} - 1)\alpha$$

$$\times \left\{\sigma - \alpha \ln\left|1 + \frac{\sigma}{\alpha}\right| + (T_{e} - 2)\left[\frac{\alpha}{1 + \alpha^{2}}\ln\left|1 + \frac{\sigma}{\alpha}\right|\right]$$

$$+ \frac{\alpha}{2(1 + \alpha^{2})}\ln(1 + \sigma^{2}) + (1 + \alpha^{2})^{-1}\arctan\sigma\right]\right\}. \quad (3.40)$$

The dependence  $V(\sigma)$  at  $\sigma > 0$  is analogous to the one in the case  $\beta = 2$ . The negative semiaxis at  $T_e \ge T_{c0}$  shows a very small minimum of  $V(\sigma)$ , accompanied by an infinite increase in V at  $\sigma = -\sigma_p$ . Hence, negative values of  $\sigma$  and  $\varepsilon$  hardly occur. A cubic equation must be solved to determine the stationary states of the system; the resulting cumbersome expressions are not presented here.

### 3.3.2 Kinetics of a second-order phase transition.

1. The case  $\tau_{\varepsilon} \ll \tau_{\sigma}, \tau_{T}$ . Section 3.2 reports a qualitative analysis of the system of equations (3.15)–(3.17) in the adiabatic approximation when characteristic relaxation times satisfy the conditions  $\tau_{\varepsilon}, \tau_{T} \ll \tau_{\sigma}$ . In this section, we discuss the cases where one of the relaxation times is the minimum one [68, 87]. Specifically, setting  $\tau_{\varepsilon}\dot{\varepsilon} = 0$  with  $\tau_{\varepsilon} \ll \tau_{\sigma}, \tau_{T}$  in (3.16) gives the relation

$$\varepsilon = (T-1)\sigma. \tag{3.41}$$

Substituting Eqn (3.41) in Eqns (3.15) and (3.17) and using the scale  $\tau_{\sigma}$  to measure time, we obtain the system of equations

$$\dot{\sigma} = -\sigma \left[ 1 - g(T - 1) \right], \tag{3.42}$$

$$\dot{T} = \tau^{-1} [T_e - T - \sigma^2 (T - 2)]$$
 (3.43)

with the parameter  $\tau \equiv \tau_T/\tau_\sigma$ . The phase portrait is characterized by the presence of two singular points,  $D(T_e, 0)$  and  $O(1 + g^{-1}, \{[T_e - (g^{-1} + 1)](g^{-1} - 1)^{-1}\}^{1/2})$ . Because it is impossible to obtain the exact solution of the system of equations (3.42), (3.43) analytically, the phase plane method [88] is used to find phase portraits of the system. Their exact form is deduced by numerical integration using the fourthorder Runge–Kutta method.

The analysis of Lyapunov's parameters

$$\lambda_{\rm D} = \frac{1}{2} \left[ g(T_{\rm e} - 1) - 1 - \tau^{-1} \right] \\ \times \left( 1 \pm \sqrt{1 + 4\tau^{-1} \left[ g(T_{\rm e} - 1) - 1 \right] \left[ g(T_{\rm e} - 1) - 1 - \tau^{-1} \right]^{-2}} \right)$$
(3.44)

indicates that point D is a stable node in the precritical region  $T_e \leq T_{c0}$ . At such  $T_e$  values, the singular point O is not realized, and the system evolves into the stationary solid-like state corresponding to point D.

An increase in the parameter  $\tau = \tau_T/\tau_\sigma$  leads to the winding of trajectories around point D. In other words, relatively small variations of temperature and rapid changes in stresses are associated with the tendency toward the development of the stick-slip friction behavior. In the post-critical region  $T_e > T_{c0}$ , point D is a saddle (Fig. 8).

Lyapunov's parameters of the singular point O are given by

$$\lambda_{\rm O} = \frac{T_{\rm e} - 2g^{-1}}{2\tau(g^{-1} - 1)} \left[ 1 \pm \sqrt{1 - 8\tau \frac{g(T_{\rm e} - 1 - g^{-1})(g^{-1} - 1)^2}{(T_{\rm e} - 2g^{-1})^2}} \right].$$
(3.45)

It follows that point O is a stable node when the parameter  $\tau$  is bounded from above by the quantity

$$\tau_{\rm c} = \frac{(T_{\rm e} - 2g^{-1})^2}{8g(T_{\rm e} - 1 - g^{-1})(g^{-1} - 1)^2} , \qquad (3.46)$$

and is a focus at  $\tau > \tau_c$ .

Thus, stick–slip friction behavior (Fig. 8c) develops in the postcritical region  $T_e > T_{c0}$  for  $\tau_\sigma \ll \tau_T$ ; it is characterized by the frequency

$$\omega = \frac{2g^{-1} - T_{\rm e}}{2\tau_T(g^{-1} - 1)} \left[ 8\tau \frac{g(T_{\rm e} - 1 - g^{-1})(g^{-1} - 1)^2}{(T_{\rm e} - 2g^{-1})^2} - 1 \right]^{1/2}$$
(3.47)

and the attenuation factor

$$\alpha = \frac{1}{2} \tau_T^{-1} (g^{-1} - 1)^{-1} (T_e - 2).$$
(3.48)

The frequency  $\omega$  decreases while  $\alpha$  and  $\tau_c$  increase as the temperature increases in the interval  $T_e > T_{c0}$ . In other words, the excitation of a system leads to the disappearance of damping oscillations that represent the stick–slip friction behavior. However, Fig. 8 shows that the stick–slip motion is promoted by an increase in the parameter  $\tau = \tau_T / \tau_\sigma \ge 1$ .

The opposite limit  $\tau_T \ll \tau_{\sigma}$  corresponds to the adiabatic approximation, representing the standard picture of a phase transition (lubricant melting). It can be seen from Fig. 8a that a decrease in the parameter  $\tau \rightarrow 0$  permits distinguishing an MOD segment on the phase portrait of the system to which all trajectories rapidly converge with time [68, 87]. The MOD



Figure 8. Phase portraits of a liquid-like phase ( $T_e = 3.3$ , g = 0.5): (a)  $\tau_{\varepsilon} \ll \tau_{\sigma} = 10^2 \tau_T$ , (b)  $\tau_{\varepsilon} \ll \tau_{\sigma} = \tau_T$ , (c)  $\tau_{\varepsilon} \ll \tau_T = 10^2 \tau_{\sigma}$ . Dashed and dotted lines respectively indicate points at which phase trajectories have vertical and horizontal tangents.

segment, whose position depends on the external conditions (e.g., the  $T_e$  value) is called the 'mainstream' in [89]. Studies of time-dependent paths covered by a configurative point show that it rapidly arrives at the MOD segment and slowly moves over it. In other words, the motion primarily determines the system kinetics, which accounts for the universal kinetic picture of phase transitions [89].

2. The case  $\tau_{\sigma} \ll \tau_{\varepsilon}, \tau_{T}$ . Setting  $\tau_{\sigma}\dot{\sigma} = 0$  in (3.15) yields the relation

$$\sigma = g\varepsilon. \tag{3.49}$$

Its substitution in (3.16) and (3.17) gives the system of equations

$$\dot{\varepsilon} = -\varepsilon \left[ 1 - g(T - 1) \right], \tag{3.50}$$

$$\dot{T} = \tau^{-1} [T_{\rm e} - T - \epsilon^2 g(1 - g)],$$
(3.51)

where time is measured in units of  $\tau_{\varepsilon}$  and  $\tau \equiv \tau_T/\tau_{\varepsilon}$ . The phase portrait is characterized by the presence of singular points  $D(T_e, 0)$  and  $O(1 + g^{-1}, \{[T_e - (g^{-1} + 1)]g^{-1}(1 - g)^{-1}\}^{1/2})$ . Lyapunov's parameters are given by

$$\lambda_{\rm D} = \frac{1}{2} \left[ g(T_{\rm e} - 1) - \tau^{-1} - 1 \right] \\ \times \left\{ 1 \pm \sqrt{1 + 4\tau^{-1} \frac{g(T_{\rm e} - 1 - g^{-1})}{\left[\tau^{-1} - g(T_{\rm e} - 1 - g^{-1})\right]^2}} \right\}, \quad (3.52)$$

$$\lambda_{\rm O} = -\frac{1}{2} \, \tau^{-1} \Big[ 1 \pm \sqrt{1 - 8\tau (T_{\rm e} - 1 - g^{-1})} \, \Big] \,. \tag{3.53}$$

Similarly to case 1, point D is a stable node at  $T_e \leq T_{c0}$ and a saddle at  $T_e > T_{c0}$ . Point O is realized only in the liquidlike region  $T_e > T_{c0}$ , where it is a stable node at small  $\tau$  and a stable focus at  $\tau$  above the critical value

$$\tau_{\rm c} = \frac{1}{8} (T_{\rm e} - 1 - g^{-1})^{-1} \,. \tag{3.54}$$

The corresponding expressions for the frequency and decrement of oscillation damping are

$$\omega = \frac{1}{2} \tau_T^{-1} \left[ 8\tau (T_e - 1 - g^{-1}) - 1 \right]^{1/2}, \qquad (3.55)$$

$$\alpha = \left(2\tau_T\right)^{-1}.\tag{3.56}$$

The analysis conducted and the phase portraits obtained indicate that similarly to case 2, the system with a large parameter  $\tau$  is characterized by damping oscillations, i.e., stick-slip motion (Fig. 8c); a decrease in  $\tau$  to  $\tau \ll 1$  induces the dissipative relaxation mode (Fig. 8a). The universality of the kinetic behavior manifests itself similarly in the adiabatic limit  $\tau \rightarrow 0$  due to the presence of the MOD segment identified in Fig. 8a, at which the system slowly evolves to the stationary point O [68, 87].

3. The case  $\tau_T \ll \tau_{\sigma}, \tau_{\varepsilon}$ . Setting  $\tau_T \dot{T} = 0$  in (3.17) yields the relation

$$T = T_{\rm e} - \sigma \varepsilon + \sigma^2 \,, \tag{3.57}$$

and Eqns (3.15) and (3.16) take the form

$$\dot{\sigma} = -\sigma + g\varepsilon, \qquad (3.58)$$

$$\dot{\varepsilon} = \tau^{-1} \left[ -\varepsilon + (T_{\rm e} - 1)\sigma - \sigma^2(\varepsilon + \sigma) \right], \qquad (3.59)$$

where time is measured in units of  $\tau_{\sigma}$  and  $\tau \equiv \tau_{\varepsilon}/\tau_{\sigma}$ . The phase portrait has singular points D(0,0) and

$$O\Big(\big\{\big[T_{e} - (g^{-1} + 1)\big]g^{-1}(1 - g)^{-1}\big\}^{1/2}, \\ \big\{\big[T_{e} - (g^{-1} + 1)\big](g^{-1} - 1)^{-1}\big\}^{1/2}\Big)\Big\}$$

(see Fig. 8), with the latter realized only in the liquid-like region  $T_{\rm e} > T_{\rm c0}$ . The corresponding Lyapunov parameters are

$$\lambda_{\rm D} = -\frac{1}{2}(1+\tau^{-1}) \times \left[1 \pm \sqrt{1+4\tau^{-1}g(1+\tau^{-1})^{-2}(T_{\rm e}-1-g^{-1})}\right], \quad (3.60)$$

$$\lambda_{\rm O} = -\frac{1}{2} \left( 1 + \tau^{-1} \frac{T_{\rm e} - 2}{g^{-1} - 1} \right) \\ \times \left[ 1 \pm \sqrt{1 - 8\tau \frac{(g^{-1} - 1)[T_{\rm e} - g^{-1} - g(T_{\rm e} - 1)]]}{[\tau(g^{-1} - 1) + T_{\rm e} - 2]^2}} \right]. \quad (3.61)$$

Point D is a stable node at  $T_e \leq T_{c0}$  but transforms into a saddle point upon transition to the post-critical region  $T_e > T_{c0}$ . Point O, which characterizes the liquid-like phase



Figure 9. Phase portraits for the first-order phase transition ( $T_e = 0.9$ ,  $g_\theta = 0.6$ ,  $\theta = 0.1$ ,  $\alpha = 0.75$ ): (a)  $\tau_{\varepsilon} \ll \tau_{\sigma} = 10^2 \tau_T$ , (b)  $\tau_{\varepsilon} \ll \tau_{\sigma} = \tau_T$ , (c)  $\tau_{\varepsilon} \ll \tau_T = 10^2 \tau_{\sigma}$ . Dashed and dotted lines respectively indicate points at which phase trajectories have vertical and horizontal tangents.

at the values of  $\tau$  in the range  $(\tau_{-}, \tau_{+})$ , where

$$\begin{aligned} \pi_{\pm} &= (g^{-1} - 1)^{-1} \left[ 3T_{\rm e} + 2 - 4g^{-1} - 4g(T_{\rm e} - 1) \right] \\ &\pm (g^{-1} - 1)^{-1} \left\{ 8 \left[ g^{-1} + g(T_{\rm e} - 1) - T_{\rm e} \right] \right. \\ &\times \left[ 2g^{-1} + 2g(T_{\rm e} - 1) - T_{\rm e} - 2 \right] \right\}^{1/2}, \end{aligned}$$
(3.62)

is a stable focus or a stable node at the  $\tau$  values outside the interval  $(\tau_{-}, \tau_{+})$ . The characteristic frequency

$$\omega = \frac{1}{2} \tau_{\varepsilon}^{-1} (g^{-1} - 1)^{-1} \Big\{ 8\tau (g^{-1} - 1) \big[ T_{e} - g^{-1} - g(T_{e} - 1) \big] \\ - \big[ \tau (g^{-1} - 1) + T_{e} - 2 \big]^{2} \Big\}^{1/2}$$
(3.63)

and the attenuation factor

$$\alpha = \frac{1}{2} \tau_{\varepsilon}^{-1} (g^{-1} - 1)^{-1} [\tau (g^{-1} - 1) + T_{\rm e} - 2]$$
(3.64)

are comparable at all  $T_e$  and  $\tau$  values. This is why the stick– slip friction behavior is virtually absent here, unlike in cases 1 and 2 considered above.

Phase portraits suggest that the universality of the kinetic behavior of the system is manifested at both  $\tau_{\varepsilon} \ll \tau_{\sigma}$  and  $\tau_{\varepsilon} \gg \tau_{\sigma}$ . In the former case, a universal segment is reached owing to the rapid change in the strain  $\varepsilon(t)$  while stresses  $\sigma(t)$ remain virtually unaltered (Fig. 8a); in the latter case, the opposite picture is realized, i.e.,  $\sigma(t)$  changes very rapidly and  $\varepsilon(t)$  is almost unaltered. In the intermediate region  $\tau_{\varepsilon} \sim \tau_{\sigma}$ , universality manifests itself only at small initial values of the strain  $\varepsilon(0)$  and stresses  $\sigma(0)$ :  $\varepsilon(0) \ll \varepsilon_0, \sigma(0) \ll \sigma_0$ . Unlike cases 1 and 2, the universal part of phase trajectories has a downward, rather than upward, character corresponding to the deformation curve [61, 90–92].

## 3.3.3 Kinetics of a first-order phase transition.

1. The case  $\tau_{\varepsilon} \ll \tau_{\sigma}, \tau_{T}$ . As in the case of a second-order phase transition, we consider different limit relations for relaxation times  $\tau_{\sigma}, \tau_{\varepsilon}$ , and  $\tau_{T}$  [68, 87] using dependence (3.33) at  $\beta = 2$  instead of the constant g in the original Eqn (3.15). The standard analytic analysis of stability of singular points is not performed for a first-order phase transition because the resulting expressions are too cumbersome. The phase portraits are developed for surface temperatures in the interval  $(T_{c}^{0}, T_{c0})$  where the first-order phase transition is realized. The phase portraits for  $T_{e} \ge T_{c0}$  are analogous to those considered in Section 3.3.2.

Setting  $\tau_{\varepsilon}\dot{\varepsilon} = 0$  in (3.16) yields the relation  $\varepsilon = (T-1)\sigma$ . Taking it into consideration in Eqns (3.15), (3.17) and using the scale  $\tau_{\sigma}$  for measuring time leads to system (3.43) and

$$\dot{\sigma} = -\sigma \left[ 1 - g_{\theta} \left( 1 + \frac{\theta^{-1} - 1}{1 + (\sigma/\alpha)^2} \right) (T - 1) \right].$$
(3.65)

The phase portrait of system (3.65), (3.43) has three singular points,  $D(T_e, 0)$ ,  $S(T^m, \sigma^m)$ , and  $O(T_0, \sigma_0)$ , where

$$T_0^{\rm m} = T_{\rm e} - \sigma_0^{\rm m} \varepsilon_0^{\rm m} + (\sigma_0^{\rm m})^2 , \qquad (3.66)$$

$$\varepsilon_0^{\rm m} = \left(1 + (\sigma_0^{\rm m})^2\right)^{-1} \left(T_{\rm e} + (\sigma_0^{\rm m})^2 - 1\right) \sigma_0^{\rm m}, \qquad (3.67)$$

and the characteristic values of  $\sigma_0^{\rm m}$  are given by (3.37) and (3.38).

In the temperature interval of interest, S is a saddle point, O is a stable node or focus, and D is a stable node. It can be seen from Fig. 9 how the phase portrait changes upon increasing the relaxation time ratio  $\tau = \tau_T/\tau_{\sigma}$ . A comparison of Figs 8 and 9 shows that trajectories in the vicinity of point O behave virtually the same as in the case of a secondorder phase transition, i.e., they rapidly converge to the universal OS segment in the adiabatic limit  $\tau_T \ll \tau_{\sigma}$  (Fig. 9a), whereas in the opposite limit  $\tau_T \gg \tau_{\sigma}$ , the damping oscillation mode representing stick–slip motion develops. The main difference between the two cases is in the appearance of a separatrix in the region of small stress values, reflecting the existence of a barrier in the dependence  $V(\sigma)$ .

2. The case  $\tau_{\sigma} \ll \tau_{\varepsilon}, \tau_T$ . Substituting expression (3.33) for g in the original Eqn (3.15), where we set  $\tau_{\sigma}\dot{\sigma} = 0$ , leads to a cubic equation whose solution can be conveniently written in the form

$$\sigma = \frac{1}{3} \left( g_{\theta} \varepsilon + \sigma_+^0 + \sigma_-^0 \right). \tag{3.68}$$

Here, the functions

$$\sigma_{\pm}^{0} = \left[\frac{1}{2}(2g_{\theta}^{3}\varepsilon^{3} + 27g_{\theta}\varepsilon\alpha^{2}\theta^{-1} - 9g_{\theta}\varepsilon\alpha^{2}) \pm Q^{1/2}\right]^{1/3}, \quad (3.69)$$
$$Q = (3\alpha^{2} - g_{\theta}^{2}\varepsilon^{2})^{3} + \frac{1}{4}(9g_{\theta}\varepsilon\alpha^{2} - 27g_{\theta}\varepsilon\alpha^{2}\theta^{-1} - 2g_{\theta}^{3}\varepsilon^{3})^{2}$$
$$(3.70)$$

are introduced.

Substitution of relation (3.68) in (3.16) and (3.17) gives the system of equations [cf. (3.50), (3.51)]

$$\dot{\varepsilon} = -\varepsilon + \frac{1}{3}(T-1)(g_{\theta}\varepsilon + \sigma_{+}^{0} + \sigma_{-}^{0}), \qquad (3.71)$$

$$\tau \dot{T} = T_{\rm e} - T - \frac{1}{3} (g_{\theta} \varepsilon + \sigma_+^0 + \sigma_-^0) \left[ \varepsilon - \frac{1}{3} (g_{\theta} \varepsilon + \sigma_+^0 + \sigma_-^0) \right].$$
(3.72)

The phase portrait of system (3.71), (3.72) has three singular points (see Fig. 9):  $D(T_e, 0)$ ,  $S(T^m, \varepsilon^m)$ , and  $O(T_0, \varepsilon_0)$ , where characteristic values of  $T_0^m$  and  $\varepsilon_0^m$  are given by (3.66) and (3.67). As in case 1, point D is a stable node, S is a saddle point, and O is an attracting node or a focus. It is equally clear that the universal character of kinetic behavior of the system is fairly apparent at  $\tau_T \ll \tau_{\varepsilon}$  (Fig. 9a); in the opposite limit  $\tau_T \gg \tau_{\varepsilon}$ , the damping oscillation mode representing stick–slip friction is observed (Fig. 9c). As in case 1, comparison of the phase portrait of system (3.71), (3.72) with that for the second-order transition (see Fig. 8) reveals the appearance of a separatrix in the region of small T and  $\varepsilon$  values corresponding to the energy barrier that separates liquid-like and solid-like phases.

3. The case  $\tau_T \ll \tau_{\sigma}, \tau_{\varepsilon}$ . Setting  $\tau_T \dot{T} = 0$  in (3.17) yields relation (3.57). Its substitution in (3.16) gives Eqn (3.59), and Eqn (3.15) takes the form [cf. (3.58)]

$$\dot{\sigma} = -\sigma + g_{\theta} \left[ 1 + \frac{\theta^{-1} - 1}{1 + (\sigma/\alpha)^2} \right] \varepsilon.$$
(3.73)

The phase portrait of system (3.59), (3.73) has three singular points (see Fig. 9): D(0,0),  $S(\varepsilon^m, \sigma^m)$ , and  $O(\varepsilon_0, \sigma_0)$ , where characteristic values of  $\sigma_0^m$  and  $\varepsilon_0^m$  are given by (3.37) and (3.67). As in cases 1 and 2, point D is a stable node, S is a saddle point, and O is an attracting node or a focus.

Comparison with the phase portrait for the second-order transition shows that the only complication, as before, is the appearance of a separatrix in the region of small  $\varepsilon$  or  $\sigma$  values. As in the case of the second-order phase transition, the universality of the kinetic behavior of the system at both  $\tau_{\varepsilon} \ll \tau_{\sigma}$  and  $\tau_{\varepsilon} \gg \tau_{\sigma}$  is preserved for the first-order transition.

The analysis of the case  $\beta = 1$  shows that the synergetic potential is the odd function of  $\sigma$  given by expression (3.40). Phase portraits for all relations of relaxation times are completely analogous to those at  $\beta = 2$ .

### 3.4 Hysteresis phenomena

### in the strain dependence of the shear modulus

As noted above, the replacement  $\varepsilon/\tau_{\sigma} \rightarrow \partial \varepsilon/\partial t$  reduces Eqn (3.15) with  $g(\sigma) = G/G_0 \equiv \text{const}$  to a Maxwell-type relation for the description of a viscoelastic medium. The Maxwell equation implies the use of the idealized Genki model. The shear modulus of a lubricant is known to depend on stress [61, 86]. In Section 3.3, this dependence was taken into account by using the simplest approximation (3.32) describing the transition of the elastic deformation mode to the plastic one. This transition occurs at characteristic values of shear stresses  $\sigma_p$  and strain  $\varepsilon_p$ .

Dependence (3.32) describes hysteresis during melting of a thin lubricant only in  $T_e - \sigma$  coordinates (see Fig. 7). The deformation curve  $\sigma(\varepsilon)$  is monotonic and permits representing only a continuous transition. However, experimental data

unambiguously suggest that the melting of a molecularly thin lubricant film is a jump-like process [5, 6], even if it is underlain by the second-order phase transition mechanism. As shown below [90–92], this feature can be described by replacing the dependence  $g(\sigma)$  in (3.15) with the dependence  $g(\varepsilon) \equiv G(\varepsilon)/G_0$ , where

$$G(\varepsilon) = \Theta + \frac{G - \Theta}{1 + (\varepsilon/\varepsilon_{\rm p})^{\beta}}.$$
(3.74)

Here, the value of the parameter  $\beta > 0$  determining the character of the potential also plays a key role.

We consider a stationary state in which  $\dot{\sigma} = 0$  in (3.15) and the stresses  $\sigma$  in the lubricant do not change. According to (3.31), this case corresponds to sliding at a constant speed. We then arrive at an equation resembling the Hooke law:

$$\sigma = g(\varepsilon)\varepsilon, \qquad g(\varepsilon) = g_{\theta} \left( 1 + \frac{\theta^{-1} - 1}{1 + (\varepsilon/\alpha)^{\beta}} \right), \tag{3.75}$$

where the parameter  $\theta = \Theta/G < 1$  and the coefficients  $g_{\theta} = \Theta/G_0 < 1$  and  $\alpha = \varepsilon_p/\varepsilon_s$  are introduced. Formula (3.75) in the case of a surface force apparatus [84] describes the dependence  $\sigma = \sigma_{el} + \sigma_v$  of the overall internal shear stress in the lubricating material on the deflection amplitude  $x_{max}$ , where  $\sigma_{el}$  and  $\sigma_v = \eta_{eff} V/h$  are the elastic and viscous components. This means that two situations can be realized, with the curve  $\sigma(\varepsilon)$  monotonically increasing at small  $\beta$  and becoming nonmonotonic at

$$\beta > \frac{1+\sqrt{\theta}}{1-\sqrt{\theta}} \,. \tag{3.76}$$

The former case corresponds to the continuous melting of the lubricant; the latter case is illustrated in detail in Fig. 10. As stresses increase further up to point A, the lubricant melts and transition to point B occurs. An even further increase in stresses causes the deformation to grow monotonically, while the lubricant remains liquid-like. If the stresses then decrease, the lubricant preserves its liquid-like structure down to point C, and thereafter undergoes jump-like solidification as it passes to point D. It remains solid-like if stresses continue to decrease. Such transitions are repre-



**Figure 10.** Dependence of the stationary value of shear stresses  $\sigma_0$  and the strain  $\varepsilon_0$  in (3.75) at  $\theta = 0.2$ ,  $g_{\theta} = 0.1$ ,  $\alpha = 0.3$ , and  $\beta = 3.0$  showing a hysteretic behavior. The inset presents the same dependence at  $\beta = 1.0$ .

sented in Ref. [93] as first-order phase transitions, but between states that are not true thermodynamic phases. The notion of shear melting has been introduced to explain these transitions [15]. Such a hysteretic behavior has been observed in experiment [31, 84, 85].

The use of (3.75) yields the abscissas of transition points A and C:

$$\varepsilon_{A,C} = 2^{-1/\beta} \alpha \left[ b(\beta - 1) - 2 \mp b \sqrt{(1 - \beta)^2 - \frac{4\beta}{b}} \right]^{1/\beta}, \quad (3.77)$$
$$b = \theta^{-1} - 1,$$

where point A has the minus sign and point C the plus sign. It follows from (3.77) that the jump during melting increases as  $\alpha$  increases; as  $\beta$  increases,  $\varepsilon_A - \varepsilon_C \rightarrow 0$ . Thus, melting and solidification at large  $\beta$  (small  $\alpha$ ) proceed at virtually the same strain value ( $\varepsilon_A \approx \varepsilon_C$ ), but at different stresses  $\sigma$ . As before (see Section 3.2), we assume the shear stress  $\sigma$  to be the order parameter: the lubricant is liquid-like at  $\sigma > \sigma_A$  and solid-like at  $\sigma < \sigma_C$ . In the intermediate region  $\sigma_C < \sigma < \sigma_A$ , the lubricant state is unstable because it can exist in both phases.

In adiabatic approximation (3.20), with  $\tau_{\varepsilon}\dot{\varepsilon} \approx 0$  and  $\tau_T \dot{T} \approx 0$ , we obtain Landau–Khalatnikov equation (3.23) with the synergetic potential

$$V = \frac{\sigma^2}{2} - g_{\theta} \int_0^{\sigma} \left[ \sigma' - \frac{(2 - T_e)\sigma'}{1 + {\sigma'}^2} \right] \\ \times \left\{ 1 + \frac{\theta^{-1} - 1}{1 + \left[ \sigma'/\alpha - \sigma'(2 - T_e)/(\alpha + \alpha {\sigma'}^2) \right]^{\beta}} \right\} d\sigma'.$$
(3.78)

Figure 11 shows the dependence of stationary shear stresses  $\sigma_0$  on the temperature  $T_e$  of friction surfaces. When  $T_e$  is below the critical value  $T_{c0}$ , there are no stresses in the lubricant ( $\sigma = 0$ ) and it is solid-like. As the temperature increases to  $T_{cA}$ , corresponding to point A, the stress increases but corresponds to the Hookean elastic section of the dependence shown in Fig. 10; in other words, the lubricant remains solid-like. After the temperature becomes even higher in the  $T_{cA} < T_e < T_{cB}$  interval ( $T_{cB}$  corresponds to point B), the stress persists, and the structure of the lubricant does not change. At temperatures above  $T_{cB}$ , the plastic section of the dependence shown in Fig. 10 is realized. If the



**Figure 11.** Plot of the stationary value of shear stresses  $\sigma_0$  versus the friction surface temperature  $T_e$  at the parameters in Fig. 10.

temperature of the friction surfaces decreases, the lubricant remains liquid-like while temperatures are still higher than  $T_{\rm cC}$ ; the stress persists at  $T_{\rm cD} < T_{\rm c} < T_{\rm cC}$ , while at lower temperatures the lubricant acquires a solid-like structure. The expression for the critical temperature  $T_{\rm c0}$  is derived from the condition  $\partial V/\partial \sigma = 0$ , where V is synergetic potential (3.78):  $T_{\rm c0} = 1 + \theta/g_{\theta} \equiv 1 + G_0/G$  (3.25).

## 4. Effect of fluctuations on the melting of an ultrathin lubricant film

## 4.1 Dynamic phase diagram

Any system experiences fluctuations, which are described mathematically by noises. The noises are introduced in solving problems with many irregularities, defects, external impacts, etc., when all relevant factors cannot be objectively described. Numerous influences can be qualitatively represented as noise. Taking noises into account allows an initially idealized model to be brought closer to experiment. We show below that the presence of fluctuations in a system promotes the development of experimentally observed stick–slip friction. The aim of this section is to elucidate causes behind stick–slip friction.

The stick-slip behavior in which the friction force F periodically varies with time is shown in Fig. 2b. Random variations of F are detected by the molecular dynamics method [20] and confirmed in experiment for surfaces coated with surfactants under different conditions [94]. Such irregular behavior was also observed in experiments [4] in which a 0.8 nm thick tetradecane film (two monolayers) was used as the lubricant and the shear rate was close to the critical value.

Evidently, this behavior is due to fluctuations that have an appreciable effect in view of the system smallness. Temperature fluctuations can be introduced only for ultrathin lubricant films composed of a limited number of particles [1, 20, 22, 63]. According to [15], the total mean square displacement of molecules is the sum of expressions (3.18) and (3.19), assuming that thermal fluctuations and stresses are independent. Therefore, strain fluctuations related to stress oscillations and thermal fluctuations should be considered separately.

In accordance with the method described in Refs [95–98], we introduce stochastic terms in the form of additive noncorrelated noises  $I_{\sigma}^{1/2}\xi_1$ ,  $I_{\varepsilon}^{1/2}\xi_2$ , and  $I_T^{1/2}\xi_3$  into the right-hand sides of Eqns (3.15)–(3.17) (here, intensities  $I_{\sigma}$ ,  $I_{\varepsilon}$ , and  $I_T$  are measured in the units of  $\sigma_s^2$ ,  $\varepsilon_s^2 \tau_{\varepsilon}^{-2}$ , and  $(T_c \kappa/h)^2$ ). The function  $\xi(t)$  is a  $\delta$ -correlated stochastic source (white noise) [99, 100], whose moments are given by<sup>3</sup>

$$\langle \xi_i(t) \rangle = 0, \quad \langle \xi_i(t) \, \xi_j(t') \rangle = 2\delta_{ij}\delta(t-t').$$
 (4.1)

The distribution of  $\xi(t)$  by values is Gaussian [101],

$$P(\xi) = \frac{1}{\sqrt{2\pi}\mu} \exp\left(-\frac{\xi^2}{2\mu^2}\right),\tag{4.2}$$

where

$$\mu^2 \equiv \left< \xi^2(t) \right> = 2\delta(0) \tag{4.3}$$

<sup>3</sup> The factor 2 in (4.1) is chosen to simplify the Fokker–Planck equation (FPE).

is the second moment of the source, diverging as  $1/\tau$  at  $\tau \to 0$ , where  $\tau$  is the width of the delta-function, which is always nonzero in real physical systems.

The discussion in Section 3 shows that an external impact may trigger self-organization, resulting in an 'ordered' state (melting) of a nonequilibrium system (lubricant). It is known from the theory of phase transitions that such a state is formed in a critical mode, i.e., the ordered phase develops as a self-similar structure without a characteristic scale [102]. The formal property of self-similarity is expressed as the homogeneity of the distribution function P(x) in the amplitude x of the hydrodynamic mode responsible for ordering:

$$P\left(\frac{x}{x_{\rm c}}\right) = x_{\rm c}^{\alpha} P(x) \,. \tag{4.4}$$

According to (4.4), a change in the resolution scale  $x_c$  of a random quantity x results in a multiplicative change in the probability P of its realization, characterized by an exponent  $\alpha$ . Introducing the rescaled variable  $y \equiv x/x_c$  and the distribution function  $\mathcal{P}(y) \equiv y^{\alpha} P(y)$ , we rewrite (4.4) as

$$P(x) = x^{-\alpha} \mathcal{P}(y) \,. \tag{4.5}$$

This implies the main property of self-similar systems: the power-series form of the P(x) distribution in the limit of large and small values of the stochastic variable x when the function  $\mathcal{P}(y)$  can be assumed constant. Specifically, the self-organized criticality mode can be realized in such a system [103, 104].

This fact can be taken into account by replacing the order parameter  $\sigma$  in all terms in Eqns (3.15)–(3.17) with  $\sigma^a$  with an exponent 0 < a < 1. Then the basic equations with stochastic additions take the form [96, 105–107]

$$\tau_{\sigma}\dot{\sigma} = -\sigma^{a} + g\varepsilon + \sqrt{I_{\sigma}}\,\xi_{1}(t)\,,\tag{4.6}$$

$$\tau_{\varepsilon}\dot{\varepsilon} = -\varepsilon + (T-1)\sigma^{a} + \sqrt{I_{\varepsilon}}\,\xi_{2}(t)\,,\tag{4.7}$$

$$\tau_T \dot{T} = (T_e - T) - \sigma^a \varepsilon + \sigma^{2a} + \sqrt{I_T} \xi_3(t), \qquad (4.8)$$

where dimensionless variables are used. The physical meaning of such a substitution is that the self-similarity requirement is fulfilled under the assumptions of a nonlinear stress relaxation and a fractional character of positive and negative feedback. The adiabaticity condition  $\tau_{\varepsilon}, \tau_T \ll \tau_{\sigma}$  for time measured in units of  $\tau_{\sigma}$  leads directly to the Langevin equation

$$\dot{\sigma} = f_a(\sigma) + \sqrt{I_a(\sigma)}\,\xi(t)\,,\tag{4.9}$$

where the force  $f_a(\sigma)$  and the noise intensity  $I_a(\sigma)$  are defined as

$$f_a(\sigma) \equiv -\sigma^a + g\sigma^a \left[ 1 - (2 - T_e) d_a(\sigma) \right],$$

$$I_a(\sigma) \equiv I_\sigma + g^2 (I_\varepsilon + I_T \sigma^{2a}) d_a^2(\sigma), \quad d_a(\sigma) \equiv (1 + \sigma^{2a})^{-1}.$$
(4.10)

The Langevin equation is a stochastic differential equation (SDE) because it contains a random force  $\sqrt{I_a(\sigma)}\xi(t)$ . Therefore, each its solution is individual and only statistical characteristics can be considered. For this reason, the solution probability distribution  $P_a(\sigma)$  is considered below in terms of the stress magnitude.

Multiplying (4.9) by dt yields the differential Langevin relation

$$d\sigma = f_a(\sigma) dt + \sqrt{I_a(\sigma)} dW(t), \qquad (4.11)$$

where  $dW(t) = W(t + dt) - W(t) \equiv \xi(t) dt$  is the Wiener process with the properties [100]

$$\langle \mathrm{d}W(t) \rangle = 0, \quad \langle (\mathrm{d}W(t))^2 \rangle = 2 \,\mathrm{d}t.$$
 (4.12)

In the Stratonovich approach, there is a correlation between  $\sigma(t)$  and dW, suggesting memory effects [108] in lubricant films. In other words, such an approach corresponds to Eqn (4.11) with real noise approximated by the Gaussian white noise. With (4.12), the corresponding FPE becomes

$$\frac{\partial P_a(\sigma, t)}{\partial t} = -\frac{\partial}{\partial \sigma} \left[ f_a(\sigma) P_a(\sigma, t) \right] \\ + \frac{\partial}{\partial \sigma} \left[ \sqrt{I_a(\sigma)} \frac{\partial}{\partial \sigma} \sqrt{I_a(\sigma)} P_a(\sigma, t) \right].$$
(4.13)

In the course of time, the distribution of solutions of (4.11) becomes stationary, and its explicit form can be found from (4.13) at  $\partial P_a(\sigma, t)/\partial t = 0$ :

$$P_a(\sigma) = \mathcal{Z}^{-1} \exp\left[-U_a(\sigma)\right]. \tag{4.14}$$

Distribution (4.14) is determined by the normalization constant Z and the effective potential

$$U_a(\sigma) = \frac{1}{2} \ln I_a(\sigma) - \int_0^\sigma \frac{f_a(\sigma')}{I_a(\sigma')} \, \mathrm{d}\sigma'. \tag{4.15}$$

The extremum points of (4.14) are found from the condition  $dU_a/d\sigma \equiv dI_a/d\sigma - 2f_a = 0$ . This equation is written explicitly as

$$\frac{T_{\rm e}-2}{1+\sigma^{2a}} + \frac{ag\sigma^{a-1}}{(1+\sigma^{2a})^3} \left[ 2I_{\varepsilon} - I_T(1-\sigma^{2a}) \right] = \frac{1-g}{g} \,. \quad (4.16)$$

Hence, the abscissas of the extrema of  $P_a(\sigma)$  are independent of the noise intensity  $I_{\sigma}$ . Equation (4.16) differs from that in Ref. [105] by the absence of the coefficient 2 in the second term. In other words, after a two-fold enhancement of noise intensities, the results of the analysis based on the study of the distribution of extrema (construction of phase diagrams, interpretation of stationary states) using the Stratonovich calculus coincide with the results obtained in the framework of the Ito calculus [105]. However, potential (4.15) is not reduced to the one obtained in [105] by simple renormalization of noise intensities, because the difference resides in only the first term (the presence of the coefficient 1/2). Hence, there is a difference in the time dependences of stresses. Because the aim of this review is, inter alia, elucidation of the time evolution of stresses, we use the Stratonovich calculus. Previously, in Ref. [105], we analyzed only stationary states based on the Ito approach.

At a fixed temperature  $T_e$ , the phase diagram has the form shown in Fig. 12, where the lines indicate stability loss ranges of the system. The straight line

$$I_T = 2I_{\varepsilon} \tag{4.17}$$

emanating from the origin, as follows from (4.16), marks the boundary of the zero stationary solution  $\sigma_0 = 0$ . The maximum of  $P_a(\sigma)$  at  $\sigma = 0$  is always present above this line, but absent below it. The diagram shows four regions corresponding to different friction states.



**Figure 12.** Phase diagram at g = 0.9,  $T_e = 1.5$ , and a = 0.75 with sliding (SF), dry (DF), stick–slip (SS), and liquid metastable and stable (MSF + SF) friction.

Figure 13 illustrates the unnormalized probability distribution (4.14) corresponding to points 1–3 in Fig. 12. Point 1 lies in the dry friction (DF) region of the phase diagram; therefore, there is a single maximum of the distribution function at  $\sigma = 0$ . The two-phase region (SS) of the diagram is characterized by the coexistence of the  $P_a(\sigma)$ distribution maxima at zero and nonzero stress values (point 2). Point 3 is in the region with a single probability distribution maximum at  $\sigma_0 \neq 0$  corresponding to the liquid friction mode or sliding. The inset in Fig. 13 shows the form of the distribution in the MSF + SF region. Here, two maxima of  $P_a(\sigma)$  at  $\sigma_0 \neq 0$  coexist, corresponding to the stick–slip behavior at which transitions between metastable and stable liquid friction occur.

The dependences  $P_a(\sigma)$  in Fig. 13 are shown in the double logarithmic scale. It can be seen that the distribution for the DF and SS curves takes a power-like form. Such behavior corresponds to the values  $\sigma \ll 1$  and  $I_{\sigma}, I_{\varepsilon} \ll I_T$ , at which (4.14) reduces to

$$P_a(\sigma) = \sigma^{-a} \mathcal{P}(\sigma), \qquad (4.18)$$

where

$$\mathcal{P}(\sigma) = \mathcal{Z}^{-1} g^{-1} I_T^{-1/2} (1 + \sigma^{2a}) \\ \times \exp\left\{-I_T^{-1} g^{-2} \int_0^{\sigma} \frac{1 - g \left[1 - (2 - T_e) \left(1 + (\sigma')^{2a}\right)^{-1}\right]}{\left(1 + (\sigma')^{2a}\right)^{-2} (\sigma')^a} \, \mathrm{d}\sigma'\right\}.$$

$$(4.19)$$

Self-similar systems are known to be characterized by a homogeneous distribution function [102]. Distribution (4.18) is homogeneous if function (4.19) is constant. At small stress values, the pre-exponential factor  $1 + \sigma^{2a} \rightarrow 1$ . The integral in (4.19) makes a minor contribution at  $\sigma < 0.8$  but begins to rapidly increase after  $\sigma$  exceeds a certain value [106]. In conformity with the structure of expressions (4.18) and (4.19), this integral makes the leading contribution to the resulting distribution (4.18), which becomes exponentially decreasing. The power-like distribution, characteristic of the self-similar behavior, is realized in a narrow stress range



**Figure 13.** Distribution function (4.14) at  $I_{\sigma} = 10^{-10}$  in the states shown by points 1–3 in Fig. 12:  $1 - I_{\varepsilon} = 0$ ,  $I_T = 4$  (DF);  $2 - I_{\varepsilon} = 0$ ,  $I_T = 15$  (SS);  $3 - I_{\varepsilon} = 2$ ,  $I_T = 2$  (SF). The inset shows the dependence  $P_a(\sigma)$  at  $I_{\varepsilon} = 1.968$  and  $I_T = 3.5$  (MSF + SF).

 $\sigma < 0.8$ , as shown in Fig. 13. The self-similarity properties are lost after the critical stress value is surpassed.

## 4.2 Stress time series

Equation (4.11) is numerically solved by the Euler method, although using a different iteration procedure than in Ref. [109], because (4.11) is a Stratonovich SDE. To enable the use of the conventional iteration procedure, it is necessary to pass from the Stratonovich SDE to the equivalent Ito SDE. With (4.12), Eqn (4.11) corresponds to the Ito SDE [100, 110]

$$d\sigma = \left[ f_a(\sigma) + \sqrt{I_a(\sigma)} \frac{\partial}{\partial \sigma} \sqrt{I_a(\sigma)} \right] dt + \sqrt{I_a(\sigma)} dW(t).$$
(4.20)

Bearing in mind the definition of the discrete analog of the random force differential  $dW(t) \equiv \sqrt{\Delta t} W_i$  and Eqn (4.10), we obtain an iteration procedure for solving Eqn (4.20):

$$\sigma_{i+1} = \sigma_i + \left( f_a(\sigma_i) + \frac{ag^2 \sigma_i^{2a-1} \left[ I_T (1 - \sigma_i^{2a}) - 2I_{\varepsilon} \right]}{(1 + \sigma_i^{2a})^3} \right) \Delta t$$
$$+ \sqrt{I_a(\sigma_i) \Delta t} W_i.$$
(4.21)

The equation is solved in the time interval  $t \in [0, T]$ . For a specified number of iterations N (the number of time series points), the time increment is defined as  $\Delta t = T/N$ . The force  $W_i$  has the properties [cf. (4.12)]

$$\langle W_i \rangle = 0, \quad \langle W_i W_{i'} \rangle = 0, \quad \langle W_i^2 \rangle \to 2.$$
 (4.22)

A random force with the properties of white noise is adequately represented by the Box–Muller model [111]:

$$W_i = \sqrt{\mu^2} \sqrt{-2\ln r_1} \cos(2\pi r_2), \quad r_n \in (0,1], \quad (4.23)$$

where, according to (4.22), the dispersion is  $\mu^2 = 2$  and  $W_i$  is an absolutely random number with properties (4.22). Pseudorandom numbers  $r_1$  and  $r_2$  are characterized by a uniform distribution.



**Figure 14.** The stress time series  $|\sigma|(t)$  obtained by solving Eqn (4.20) numerically with the use of (4.21) at  $N = 10^4$ , t = 50, and dt = 0.005. DF, SS, and SF states correspond to points 1–3 on the phase diagram in Fig. 12; the MSF + SF state corresponds to the values of the parameters listed in the caption to Fig. 13.

Effective potential (4.15) has minima at positive and negative values of the stress  $\sigma$ . Transitions of the system between the states corresponding to these minima occur as a result of fluctuations inherent in the numerical solution of Eqn (4.20). Because we consider only the unidirectional movement of the upper displaced surface, the negative region  $\sigma < 0$  is considered nonphysical, which allows analyzing the behavior of  $|\sigma|(t)$  in what follows. Typical realizations of  $|\sigma|(t)$  for the states of interest are shown in Fig. 14. In the DF state, extended sections can be seen at which stresses are close to zero. In the stick–slip mode, regular transitions between zero and nonzero stresses  $\sigma$  occur. Realizations of SF and MSF + SF states are visually similar, which requires an additional probability density analysis to determine the friction state (see Fig. 13).

## 4.3 Multifractal fluctuation analysis of self-similar time series

Multifractal fluctuation analysis allows numerically calculating all the main multifractal characteristics [112] describing self-similar systems. This analysis is performed by the method first proposed in [113]. Recently, it has been widely applied to time series analysis in meteorology [114], medicine [115], economics [116], etc.

For a self-similar time series, the method essentially consists in numerical calculation of the fluctuation function  $F_q(s)$  related by a scaling expression to the scale parameter s [113]:

$$F_q \sim s^{h(q)} \,, \tag{4.24}$$

where h(q) is the generalized Hurst exponent depending on the parameter q, which, in turn, can take any real values (we note that h(q) at q = 2 corresponds to the classical Hurst exponent H[117]). Thus, the function h(q) is calculated as the slope of the dependence  $F_q(s)$ , which is linear in logarithmic coordinates. The function h(q) is related to another classical multifractal scaling parameter  $\tau(q)$  [112, 113]:

$$\tau(q) = qh(q) - 1.$$
(4.25)

The self-similarity properties of time series can be described using the multifractal spectrum function  $f(\alpha)$  related to  $\tau(q)$ by the Legendre transformations [112]:

$$\alpha = \tau'(q), \quad f(\alpha) = q\alpha - \tau(q), \quad (4.26)$$

where  $\alpha$  is the Hölder exponent and the prime denotes differentiation with respect to *q*. From (4.25), we obtain a direct relation between  $f(\alpha)$  and h(q):

$$\alpha = h(q) + qh'(q), \quad f(\alpha) = q(\alpha - h(q)) + 1.$$
 (4.27)

The form of these dependences characterizes the properties of the time series. For example, h(q) = const and, accordingly, the linear increase of  $\tau(q)$  corresponds to a monofractal series. The decrease of h(q) with increasing q, together with a linear increase of  $\tau(q)$  is inherent in multifractal series characterized by the fractal dimension spectrum at different q. There is a single value of Hölder's exponent  $\alpha$ , and the dependence  $f(\alpha)$ has a narrow peak. The spectrum of  $f(\alpha)$  values is realized in the case of a multifractal series.

In general, two causes of multifractality for time series are distinguished: a wide probability distribution function for the values of elements of the series and time correlations between terms in the series. In the former case, the multifractality is not eliminated by 'mixing' the series, i.e., randomly rearranging its terms. In the latter case, mixing leads to the disappearance of correlations. Such a series transforms into a monofractal one because the grounds for multifractality are gone. For a series characterized by both causes of multifractality, its mixed series has weaker multifractality than the initial one [113]. Hence, the analysis of mixed series by the method described in [113] allows elucidating the cause of multifractality, e.g., the presence of time correlations.



**Figure 15.** Multifractal characteristics of h(q),  $\tau(q)$ , and  $f(\alpha)$  corresponding to parameters of the series shown in Fig. 14. The group of curves *I* corresponds to the series obtained directly by the iteration procedure (4.21), group 2 corresponds to the same 'mixed' series.

We apply this method to the analysis of the stress time series  $|\sigma|(t)$  presented in Fig. 14 [106]. Dependences  $F_q(s)$  in double logarithmic coordinates at several q values for a time series corresponding to the DF state have a linear form virtually over the entire range of s. It is characteristic of all the series analyzed in this review and permits us unambiguously and with a high accuracy to calculate the Hurst exponent h(q) from scaling relation (4.24). The region 50 < s < 500 in which the linear form of  $F_q(s)$  is most clearly expressed is chosen for the calculation of multifractal characteristics.

We calculate the dependences h(q),  $\tau(q)$ , and  $f(\alpha)$  for the series shown in Fig. 14 with the parameters  $N = 10^5$ ,  $t = 10^3$ , and dt = 0.01. The dependences in Fig. 15 suggest that the series corresponding to the DF state has the strongest multifractality, and the series corresponding to the stick-slip mode has a weaker multifractality. The series corresponding to MSF+SF and SF states are characterized by minor changes in the parameter h depending on q, which corresponds to a monofractal behavior. The strong multifractality for the DF state is attributable to the fact that the distribution function at small stresses in this state has a power-like form, as in self-similar systems. Multifractality is weaker in the stick-slip mode because the probability distribution corresponding to the solution of the Langevin equation also has a nonzero maximum, making it possible to pass, in the course of time, into a state defined by this maximum, which corresponds to lubricant melting. The system loses the selfsimilarity and multifractality properties under such a transition, and the stationary liquid friction mode sets in. However, the presence of two maxima of  $P_a(\sigma)$  that correspond to the stick-slip mode leads in the course of time to the reverse transition into a solid-like structure, and the system exhibits the self-similar behavior again. In MSF + SF and SF states, the system lacks multifractality, because the power-like distribution function is not realized.

It can be inferred from the above description that multifractality is due to the power-like form of the distribution function. We clarify whether the fact that  $P_a(\sigma)$  has the power-like form is a sufficient condition for multifractality. For this, we rearrange all the series and find all the fractal characteristics of the system again [106]. We first consider the stress oscillation spectrum obtained using the fast Fourier transform (FFT) algorithm [111] in the analysis of the time series for the stick–slip behavior shown in Fig. 14. The corresponding time series was obtained by iteration procedure (4.21) with  $N = 2 \times 10^5$ ,  $t = 10^3$ , and dt = 0.005. The signal strength in the spectrum decreases with increasing the frequency as  $S_p(f) \propto 1/f^{1.8}$ . This suggests the presence of correlations in the system [in the absence of correlations, i.e., in the presence of white noise alone,  $S_p(f) = \text{const}$ ]. The  $S_p(f)$  spectra have such a form for all the states being considered.

Hence, the system of interest based on Eqn (4.9) converts white noise present in virtually all physical experiments into color noise with a nonzero correlation time. After mixing, the stress oscillation spectrum is described by  $S_p(f) \propto 1/f^{0.017}$ , i.e., the exponent is virtually frequency independent. This spectrum corresponds to white noise, which means that correlations disappear after mixing. However, the form of the distribution function remains unaltered, because the terms are neither inserted into the series nor removed from it. The group of curves 2 in Fig. 15 corresponds to the analysis of series that correspond to curves 1, but after these series are mixed. Evidently, h(q) represents the same straight line h = 0.5 for the parameters of all series, the dependence  $f(\alpha)$ has a narrow peak, and  $\tau(q)$  is a straight line with a constant slope at all its sections. These features correspond to a monofractal system, with the value h = 0.5 being typical of series with no correlations. We conclude that multifractality in the system under consideration is due to two factors: a power-like distribution function and the presence of correlations.

# 5. Thermodynamic representation of boundary friction

#### 5.1 Free energy

The free energy density for a lubricant has the form [61, 118, 119]

$$f = \alpha (T - T_{\rm c}) \varphi^2 + \frac{a}{2} \varphi^2 \varepsilon_{\rm el}^2 - \frac{b}{2} \varphi^4 + \frac{c}{3} \varphi^6 + \frac{g}{2} (\nabla \varphi)^2, \quad (5.1)$$

where T is the lubricant temperature,  $T_c$  is the critical temperature,  $\varepsilon_{el}$  is the shear component of elastic strain,  $\alpha$ , a, b, c, and g are positive constants, and  $\varphi$  is the order parameter (the amplitude of the periodic part of the microscopic medium density function) [61]. The parameter  $\varphi$  is equal to zero in the liquid-like phase and differs from zero in the solid-like one. In what follows, a homogeneous case is considered, which means setting g = 0 everywhere.



**Figure 16.** Dependence of the free energy density f in (5.1) on the order parameter  $\varphi$  (dimensionless quantity) at  $\alpha = 0.7$  J K<sup>-1</sup> m<sup>-3</sup>,  $T_c = 290$  K, b = 285 J m<sup>-3</sup>, and c = 1600 J m<sup>-3</sup>. The curves (from bottom up) correspond to the temperatures T = 288, 302, 310 K and to shear strain  $\varepsilon_{el} = 0$ .

In accordance with (5.1), we define elastic stresses as  $\sigma_{el} = \partial f / \partial \varepsilon_{el}$ :

$$\sigma_{\rm el} = a\varphi^2 \varepsilon_{\rm el} \,. \tag{5.2}$$

Then the shear modulus is expressed as

$$u = a\varphi^2. \tag{5.3}$$

The analysis of potential (5.1) allows distinguishing the following situations. If the condition

$$\frac{a}{2}\varepsilon_{\rm el}^2 + \alpha(T - T_{\rm c}) \leqslant 0 \tag{5.4}$$

is satisfied, two symmetric nonzero minima of the potential separated by a single zero maximum (lower curve in Fig. 16) are realized, which corresponds to a solid-like lubricant. In the range of intermediate values

$$0 < \frac{a}{2} \varepsilon_{\rm el}^2 + \alpha (T - T_{\rm c}) < \frac{b^2}{4c} , \qquad (5.5)$$

the zero potential maximum transforms into a minimum accompanied by two additional symmetric maxima separating the central minimum from two symmetric nonzero minima (the middle curve in Fig. 16). In his situation, the lubricant can have both a solid-like and a liquid-like structure, depending on the initial conditions. For

$$\frac{a}{2}\varepsilon_{\rm el}^2 + \alpha(T - T_{\rm c}) \geqslant \frac{b^2}{4c}, \qquad (5.6)$$

a single zero minimum of  $f(\varphi)$  is realized (the upper curve in Fig. 16), which corresponds [in agreement with (5.3)] to the zero value of the shear modulus  $\mu$  and the liquid-like structure of the lubricant. The abscissas of the extrema of potential (5.1) are given by

$$\varphi_{\mp}^2 = \frac{b}{2c} \mp \sqrt{\left(\frac{b}{2c}\right)^2 - \left[\frac{a}{2c}\,\varepsilon_{\rm el}^2 + \frac{\alpha(T-T_{\rm c})}{c}\right]} \,, \tag{5.7}$$

where the minus sign corresponds to the symmetric maxima of the potential and the plus sign corresponds to its symmetric minima. According to (5.4)–(5.6), the melting of a lubricant is a result of both an increase in the temperature T and a mechanical impact accompanied by an increase in the shear component of elastic deformation  $\varepsilon_{el}$ . Thus, the model takes thermodynamic and shear melting into account.

## 5.2 Stationary states and phase diagrams

We let V denote the relative shear rate of rubbing surfaces separated by an ultrathin lubricant layer of thickness h. The relation between the shear rate and the elastic strain developing in the lubricant layer is fixed using the Debye approximation relating the elastic strain component  $\varepsilon_{el}$  to the plastic one [61]:

$$\dot{\varepsilon}_{\rm pl} = \frac{\varepsilon_{\rm el}}{\tau_{\varepsilon}} \,, \tag{5.8}$$

where  $\tau_{\varepsilon}$  is the Maxwell relaxation time of internal stresses. The overall deformation in the layer determined by the sum of the elastic and plastic components [61, 120],

$$\varepsilon = \varepsilon_{\rm el} + \varepsilon_{\rm pl} \,, \tag{5.9}$$

determines the velocity of the top block V[7]:

$$V = h\dot{\varepsilon} = h(\dot{\varepsilon}_{\rm el} + \dot{\varepsilon}_{\rm pl}). \tag{5.10}$$

The expression for the elastic component of shear strain follows from the last three relations as [121, 122]

$$\tau_{\varepsilon} \dot{\varepsilon}_{\rm el} = -\varepsilon_{\rm el} + \frac{V \tau_{\varepsilon}}{h} \,. \tag{5.11}$$

It follows from (5.11) that the stationary value of elastic strain is reached at a constant shear rate V:

$$\varepsilon_{\rm el}^0 = \frac{V\tau_\varepsilon}{h} \,. \tag{5.12}$$

In accordance with the minimum energy principle, a system tends toward a state corresponding to a minimum of the potential  $f(\varphi)$  (see Fig. 16). In this case, a stationary value of the order parameter is reached, given by Eqn (5.7) with the plus sign (because the minus sign corresponds to unstable states). Figure 17 presents stationary values of the squared order parameter computed from expression (5.7) in which the stationary elastic strain  $\varepsilon_{el}$  is defined by formula (5.12). Solid and dashed sections of the curves respectively correspond to stable and unstable stationary states.

A lubricant is solid-like at the zero shear rate (shear stresses and strains are zero) and a low temperature T, because a nonzero value of  $\varphi$  is realized; also, the shear modulus  $\mu$  is nonzero according to (5.3) (Fig. 17a, solid section of curve I). In this case, the lower curve in Fig. 16 depicts the potential. As the temperature T exceeds the critical value

$$T_{\rm c0} = T_{\rm c} - \frac{a}{2\alpha} \left(\frac{\tau_{\varepsilon} V}{h}\right)^2 + \frac{b^2}{4\alpha c}, \qquad (5.13)$$

the square of the order parameter changes jumpwise from  $\varphi_A^2 = 0.5b/c$  to zero, and the lubricant transits to the liquidlike state (the upper curve in Fig. 16). If this transition is followed by a decrease in the temperature *T*, the lubricant



**Figure 17.** Dependence of the stationary value of the squared order parameter  $\varphi^2$  (dimensionless quantity) in (5.7) on the lubricant temperature *T* and the shear rate *V* with the parameters in Fig. 16 and  $a = 4 \times 10^{12}$  Pa,  $h = 10^{-9}$  m, and  $\tau_{\varepsilon} = 10^{-8}$  s. Curves *I*-4 correspond to (a) the fixed values of the shear rate V = 0, 650, 900, 1020 nm s<sup>-1</sup>, (b) fixed temperatures T = 0, 170, 270, 300 K.

solidifies at a lower temperature:

$$T_{\rm c}^0 = T_{\rm c} - \frac{a}{2\alpha} \left(\frac{\tau_{\varepsilon} V}{h}\right)^2.$$
 (5.14)

Also, a jump of the squared order parameter from zero to  $\varphi_{\rm B}^2 = b/c$  occurs. In the intermediate region  $T_c^0 < T < T_{c0}$ , the potential has the shape shown by the middle curve in Fig. 16. It follows that the dependence  $\varphi^2(T)$  has a hysteretic character and corresponds to first-order phase transitions. It follows from Fig. 17a that the lubricant melts at a lower temperature when the shear rate increases. Curve 4 corresponds to the case where the lubricant, once melted, cannot solidify again after the temperature is decreased.<sup>4</sup> The lubricant always remains liquid-like, regardless of the temperature, if the shear rate exceeds a critical value ( $\mu = 0$ ).<sup>5</sup>

As follows from Fig. 17b, when the shear rate exceeds the critical value

$$V_{\rm c0} = \frac{h}{\tau_{\varepsilon}} \sqrt{\frac{2\alpha(T_{\rm c} - T)}{a} + \frac{b^2}{2ac}},\tag{5.15}$$

the lubricant melts; when the shear rate decreases below

$$V_{\rm c}^0 = \frac{h}{\tau_{\varepsilon}} \sqrt{\frac{2\alpha(T_{\rm c} - T)}{a}}, \qquad (5.16)$$

it solidifies.

Figure 18 shows the temperature *T* dependence of the lubricant critical melting rate  $V_{c0}$  in (5.15) and the critical solidification rate  $V_c^0$  in (5.16). The lubricant is liquid-like above the  $V_{c0}$  curve, and the SF state is realized. In the  $V < V_c^0$  region, the lubricant has a solid-like structure. In the region between the curves in Fig. 18, the potential  $f(\varphi)$  has the form depicted by the middle curve in Fig. 16; therefore, the lubricant state in this region is indefinite and depends on the initial conditions. Thus, Fig. 18 presents a phase diagram with two stationary friction states.



**Figure 18.** Phase diagram with the regions of sliding (SF) and dry (DF) friction with the parameters in Fig. 17.

### 5.3 Friction force

The overall stress in a lubricant layer is the sum of viscous  $\sigma_v$  and elastic  $\sigma_{el}$  contributions:

$$\sigma = \sigma_{\rm el} + \sigma_{\rm v} \,. \tag{5.17}$$

The friction force F is the total stresses times the contact area A of rubbing surfaces,

$$F = \sigma A \,. \tag{5.18}$$

We determine viscous stresses in the lubricant layer from the formula [7]

$$\sigma_{\rm v} = \frac{\eta_{\rm eff} V}{h} \,, \tag{5.19}$$

where  $\eta_{\text{eff}}$  is the effective viscosity of the lubricating material. The boundary lubricant is a non-Newtonian liquid. Such

<sup>&</sup>lt;sup>4</sup> The corresponding critical rate is easy to find from (5.14) at  $T_c^0 = 0$  or from (5.16) at T = 0.

<sup>&</sup>lt;sup>5</sup> The corresponding critical rate is found from (5.13) at  $T_{c0} = 0$  or from (5.15) at T = 0.



**Figure 19.** Dependence of the friction force *F* in (5.22) on the temperature *T* of the friction surfaces and the shear rate *V* with the parameters in Fig. 17 and  $\gamma = -2/3$ ,  $A = 3 \times 10^{-9}$  m<sup>2</sup>, and  $k = 4 \times 10^5$  Pa s<sup>1/3</sup>: (a) curves 2–4 correspond to constant values of the shear rate V = 650, 900, 1020 nm s<sup>-1</sup>; (b) curves *I*–4 correspond to fixed values of temperature T = 0, 170, 270, 300 K.

liquids are characterized by intricate dependences  $\eta(\dot{\epsilon})$ . For example, the viscosity of polymer solutions and melts usually decreases (pseudoplastic liquids) as the strain rate  $\dot{\epsilon}$  increases. By contrast, the viscosity of solid particle suspensions increases with increasing  $\dot{\epsilon}$  (dilatant liquids). For a qualitative analysis, we therefore use the simple approximation [7]

$$\eta_{\rm eff} = k(\dot{\varepsilon})^{\gamma} \tag{5.20}$$

to take both situations into account. The proportionality coefficient *k* [Pa s<sup> $\gamma$ +1</sup>] is thus introduced. According to (5.20),  $\gamma < 0$  for pseodoplastic liquids,  $\gamma > 0$  for dilatant ones, and  $\gamma = 0$  for Newtonian liquids, because viscosity is here independent of the shear rate [see (5.20)].

With (5.10) and (5.20), we rewrite the expression for viscous stresses (5.19) as

$$\sigma_{\rm v} = k \left(\frac{V}{h}\right)^{\gamma+1}.\tag{5.21}$$

Substituting (5.17) and (5.21) in (5.18) yields the final expression for the friction force [118, 121]

$$F = \left[\sigma_{\rm el} + k\,{\rm sgn}\,(V)\left(\frac{|V|}{h}\right)^{\gamma+1}\right]A\,,\tag{5.22}$$

where elastic stresses  $\sigma_{el}$  are defined by relation (5.2).<sup>6</sup>

Dependence (5.22) is shown in Fig. 19. All the curves in Fig. 19a correspond to the parameters of the curves in Fig. 17a. Curve I is absent in Fig. 19a because it was drawn in Fig. 17a at a zero shear rate and the friction force at rest vanishes [see (5.12), (5.22)]. The parameters of all curves in Fig. 19b correspond to the parameters of the curves in Fig. 17b.

It can be seen from Fig. 19a that the friction force decreases as the temperature increases at a fixed shear rate due to a decrease in the shear modulus. When the lubricant melts  $(T > T_{c0})$ , the friction force does not depend on temperature, because the shear modulus is then equal to zero in the framework of the model being considered. The

dependences exhibit hysteresis because shear modulus (5.3) changes jumpwise during the phase transition. With the parameters of curve 4, the melted lubricant does not solidify again after the subsequent decrease in temperature (see the caption to Fig. 17a); due to this, the friction curve remains constant after melting at any T.

Figure 19b demonstrates a somewhat different behavior of the friction force. Here, in accordance with (5.22), the total friction force first increases with increasing the shear rate as a result of increase in elastic strain component (5.12). But the shear modulus decreases as the rate increases, which results in a decrease in the elastic component of F. After the critical velocity existing in this case is exceeded, the lubricant remains solid-like, but the total friction force begins to decrease. As the shear rate increases further,  $V > V_{c0}$  [see (5.15)], the lubricant melts and elastic strain (5.2) vanishes, together with the first term in (5.22), which results in a sharp decrease in the overall friction force. If V is increased again, F also increases due to the growth of the viscous component [the second term in (5.22)]. The lubricant solidifies with a sharp increase in the friction force F at a different shear rate  $V = V_c^0$ in (5.16). We note that the dependences of the friction force after melting coincide for all curves in Fig. 19b, unlike those in Fig. 19a, because the viscous component of F depends only on the shear rate, not on temperature. Figure 19b does not show the melting transition for curve 4 to avoid cluttering the figure. Curve 4 differs from the others in that its solid section (stable friction force F before melting) and dashed section (unstable F) form a closed line. The friction force after melting is always described by the dependence shown with the dashed-dotted curve (stable F value after melting), because the lubricant cannot solidify if the velocity Vdecreases. The results shown in Fig. 19b are in good qualitative agreement with the friction map (see Fig. 5) for the boundary state proposed in Ref. [7] in summarizing experimental data.

## 5.4 Melting kinetics

The dynamic characteristics of any tribologic system are determined by its properties as a whole. Specifically, in the hysteresis region considered above, the stick–slip motion behavior can be realized in the friction process [6, 23].

<sup>&</sup>lt;sup>6</sup> Equation (5.22) contains the sign function sgn (x) and the absolute shear rate |V| because V can take negative values.

$$M\ddot{X} = K\Delta X - F, \qquad (5.23)$$

where  $\Delta X$  is the spring extension, defined as

$$\Delta X = \int_0^t V_0 \, \mathrm{d}t' - X, \qquad (5.24)$$

where *t* is the time of motion of the free spring end. If  $V_0$  does not change with time, expression (5.24) becomes

$$\Delta X = V_0 t - X. \tag{5.25}$$

To describe the lubricant behavior, we write the Landau– Khalatnikov kinetic relaxation equation [72, 73] as

$$\dot{\varphi} = -\delta \,\frac{\partial f}{\partial \varphi}\,,\tag{5.26}$$

where  $\delta$  is the kinetic coefficient characterizing the inertial properties of the system. Substituting energy (5.1) in (5.26) yields the equation in the explicit form

$$\dot{\varphi} = -\delta \left[ 2\alpha (T - T_{\rm c})\varphi + a\varphi\varepsilon_{\rm el}^2 - 2b\varphi^3 + 2c\varphi^5 \right].$$
 (5.27)

Time evolution of the system is calculated by solving kinetic Eqns (5.11), (5.23), and (5.27) and determining the spring extension  $\Delta X$  from (5.24), the friction force *F* from (5.22), and the elastic stresses  $\sigma_{el}$  from (5.2). The relation  $\dot{X} = V$  must be taken into account in solving the above equations. With a small strain relaxation time  $\tau_{\varepsilon}$ , Eqns (5.23) and (5.27) are solved together, and the current strain is found from (5.12).

The solution of these equations is shown in Fig. 20. At the selected temperature and the friction surfaces at rest, the lubricant is solid-like. The right end of the spring begins to move with a velocity  $V_0$  at the instant t = 0. Initially, both constituents of friction force (5.22) increase monotonically due to the increasing speed of the rubbing block V. Elastic stresses  $\sigma_{\rm el}$  and the spring extension  $\Delta X$  also increase. When the velocity exceeds the critical value  $V > V_{c0}$ , the lubricant melts and elastic stresses  $\sigma_{el}$  vanish. But the force F does not undergo jump-like changes because the sliding rate V of the top block and the viscous component of the friction force sharply increase. As the motion speed increases, the block rapidly moves over a long distance, as is evidenced by the increased slope of X(t) after melting. Because the block moves with a speed V that is much higher than the speed of the spring  $V_0$ , its extension  $\Delta X$  decreases. The value of  $V_0$  remains constant up to the first dotted line in Fig. 20; gradually, the stationary state of the liquid-like lubricant sets in with the unaltered values of the friction force F, the block speed V, the spring extension  $\Delta X$ , and zero elastic stresses  $\sigma_{el}$ . Also, the dependence X(t) becomes linear. After the first dotted line, the speed  $V_0$  is assumed to be zero (the right end of the spring stops). The rubbing block starts to decelerate slowly because the lubricant is liquid-like (the time instant between dotted lines in Fig. 20). After the second dotted line, the speed becomes  $V < V_c^0$  and the lubricant rapidly solidifies and undergoes elastic stress, resulting in a nonzero elastic component of the friction force; this change compensates the jump-like decrease in the viscous component of the

**Figure 20.** Dependence of the total friction force *F*, the shear rate of the upper rubbing surface *V*, its coordinate *X*, the moving spring extension  $\Delta X$ , and the elastic shear stresses  $\sigma_{el}$  on time *t* with the parameters in Figs 17 and 19 and M = 0.4 kg, K = 2500 N m<sup>-1</sup>,  $\delta = 100$  J<sup>-1</sup> m<sup>3</sup> s<sup>-1</sup>, and T = 260 K. The speed  $V_0$  is 600 nm s<sup>-1</sup> before the first dotted line (t < 30 s) and  $V_0 = 0$  nm s<sup>-1</sup> after this line (t > 30 s).

friction force F due to the decreased speed of motion V. For this reason, the friction force does not undergo jump-like changes again. Lubricant solidification is followed by slow relaxation of the quantities shown in Fig. 20, because the motion of the top block is now accompanied by a decrease in the spring extension  $\Delta X$  and the elastic force maintaining the motion.

### 5.5 Intermittent state

Experimental studies have shown unambiguously that the intermittent (stick-slip) behavior of motion, in which the relative velocity of rubbing surfaces varies periodically with time, frequently appears in the framework of the boundary friction state [6, 31, 84, 85, 123], which is illustrated in Fig. 21 showing the same dependences as in Fig. 20 but at different parameters. It follows from Fig. 21 that the lubricant periodically melts and solidifies, which accounts for the oscillating motion. Figure 22 presents the same dependences as merced the same dependences and the same dependences for the same dependences.





 $350 \text{ nm s}^{-1}$ ; the other parameters are the same as in Fig. 20.

dences, but the phase transition region is shown in more detail. In Figs 21 and 22, the spring extension  $\Delta X$  monotonically increases at the onset of motion as in Fig. 20; this results in an increase in the shear rate of the top block V, leading to the enhancement of elastic stresses  $\sigma_{el}$  and the friction force F. Meanwhile, the block covers a distance X, which grows further with time. It is clear from Fig. 22 that the lubricant melts and stresses relax to zero when the condition  $V > V_{c0}$  is satisfied, which results in a decrease in the total friction force and a decrease in the shear rate V. Because the rubbing block moves much faster than the right end of the spring, the extension  $\Delta X$  also decreases, as does the elastic force that drives the motion. As the speed V increases in the melted state, the friction force also increases. When the force F and speed V reach maximum values, the extension of the spring becomes so weak that V and F begin to decrease. However, the condition  $V > V_0$  continues to hold for some time after this; therefore,  $\Delta X$  decreases further, until the instant when the condition  $V < V_{c0}$  is satisfied and the lubricant solidifies. Solidification corresponds to a peakshaped increase in the friction force F (see the inset in



Figure 22. Magnified parts of the dependences shown in Fig. 21. Insets show magnified regions enclosed by dashed lines. The dashed line is absent in the dependence V(t), because it is poorly discernible at the scale used.

Fig. 22a), because other elastic stresses appear in the lubricant (see the inset in Fig. 22d). The dependence V(t)shown in the inset in Fig. 22b suggests that V decreases after solidification. Such a sharp decrease in the speed is due to the inability of the elastic force  $K\Delta X$  to maintain the high-speed motion, because  $\Delta X$  falls significantly during sliding. The lubricant now being solid-like and the speed V low, the spring deforms again until the speed reaches the critical value at which the lubricant melts. This process is periodic in time. The friction force in a liquid-like lubricant must be small if the solidification condition  $V < V_{c0}$  is to be fulfilled after melting; it is necessary to ensure that the rubbing block moves over a large distance and spring extension weakens appreciably for the sliding time. Therefore, smaller values of the coefficient k and the surface contact area A are chosen in Figs 21 and 22 than in Fig. 20.

Figure 23 illustrates the time dependence of the friction force at fixed temperature and increased speed V. According to Fig. 23a, the frequency of stick–slip peaks increases with as the speed increases because the critical elastic stresses at which the lubricant melts are reached faster at a higher speed. As a



**Figure 23.** Time *t* dependence of the friction force with the parameters in Fig. 21, speed values  $V_{01} = 400$  nm s<sup>-1</sup>,  $V_{02} = 700$  nm s<sup>-1</sup>,  $V_{03} = 1000$  nm s<sup>-1</sup>,  $V_{04} = 1050$  nm s<sup>-1</sup>, and temperatures *T* (a) 260 K, (b) 300 K, (c) 310 K.



**Figure 24.** Time *t* dependence of the friction force with the parameters in Fig. 21, the temperatures  $T_1 = 230$  K,  $T_2 = 250$  K,  $T_3 = 266$  K,  $T_4 = 280$  K and the velocities (a)  $V_0 = 400$  nm s<sup>-1</sup>, (b)  $V_0 = 1100$  nm s<sup>-1</sup>.

result, melting occurs earlier and the system performs more melting–solidification transitions per unit time. Figure 23b reflects a higher temperature of the lubricating material. Here, the lubricant is liquid-like for all selected speeds, but it has a solid-like stricture at rest; therefore, the dependence has a peak (shown in the inset) at the onset of motion at  $V_0 = V_{01}$ . This magnified portion is analogous to that in Fig. 22a, the sole difference being the absence of the peak corresponding to solidification, because the lubricant remains liquid-like after melting. Figure 23c corresponds to the temperature at which the lubricant is liquid-like even at rest; therefore, there is no peak corresponding to the melting.

Figure 24 depicts a situation with an increased lubricant temperature T. The dependence in Fig. 24a matches the speed  $V_0$  at which the temperature  $T = T_1$  corresponds to the solidlike lubricant structure (at this temperature, the lubricant does not melt as it moves and stick-slip friction is not present). As the temperature increases further, the stick-slip behavior develops, the amplitude of friction force fluctuations decreases, and the frequency of phase transitions increases. In Figure 24b, drawn for a higher speed  $V_0$ , the lubricant is liquid-like at  $T = T_3$  and  $T = T_4$ , which results in the sliding mode characterized by constant values of the kinetic friction force and the shear stress of the rubbing block. In the present model, the temperature T after melting has no effect on friction force (5.22); therefore, F remains the same at these temperatures.

## 6. Conclusion

The foregoing considerations enabled us to develop models that can be used to describe the effects observed in the friction of atomically flat solid surfaces in the presence of an ultrathin lubricant layer. The results extend our knowledge of friction physics of nanoscale objects that do not obey standard laws, such as the Amontons law.

The melting of ultrathin lubricant films in the presence of friction between atomically flat surfaces is represented as the result of the action of spontaneous shear stresses induced by external supercritical heating. The critical temperature of friction surfaces at which melting occurs increases linearly with the characteristic value of shear viscosity and decreases with increasing the lubricant shear modulus. It is shown that the stick–slip friction behavior is realized if the lubricant temperature relaxation time is much longer than the relaxation time of shear stresses and strain. Hysteresis is described for the dependences of stationary stresses on stationary strain and the temperature of friction surfaces.

The additive noncorrelated noise of shear stresses and strain is taken into consideration in conjunction with the lubricant film temperature. The phase diagram is constructed in which the intensities of lubricant strain and temperature fluctuations define regions of liquid, intermittent, and dry friction. Numerical analysis of the Langevin equation for different regions of the diagram has enabled the construction of stress time series that help explain friction experiments with stick–slip motion. The conditions under which these series become multifractal are elucidated.

A thermodynamic model of the melting of an ultrathin lubricant layer has been developed. Melting and solidification of lubricating materials are represented as first-order phase transitions. When rubbing surfaces slide with a constant velocity, the lubricant melts after the temperature or elastic shear stresses exceed their critical values. Because a first-order phase transition is considered, the lubricant solidifies at lower values of these quantities. The relevant phase diagram is constructed in the shear-rate–lubricant temperature coordinates. The system kinetics is considered in the framework of the mechanical analog of a simple tribologic system. Three cases are feasible depending on the values of the above parameters: (1) the lubricant is always solid-like during friction, (2) the lubricant is liquid-like, (3) periodic meltingsolidification transitions occur in the lubricant, resulting in intermittent motion. The transition region for stick-slip friction is considered in detail at small times, and the cause behind such behavior is explained. The data obtained are in qualitative agreement with the known experimental findings.

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