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Bridging the Gap Between the Atomic-Scale and Macroscopic Modeling of Friction

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Abstract A short survey of a modern view on the problem of friction from the physical viewpoint is presented. An atomically thin lubricant film confined between two substrates in moving contact has been studied with the help of molecular dynamics (MD) based on Langevin equations with coordinate- and velocity-dependent damping coefficient. Depending on model parameters, the system may exhibit either the liquid sliding regime, when the lubricant film melts during sliding (the “melting-freezing” mechanism of stick-slip motion), the “layer-over-layer” sliding regime, when the film keeps a layered structure at sliding, or the solid sliding regime, which may provide an extremely low friction (“superlubricity”). Atomic-scale MD simulations of friction, however, lead to a “viscosity” of the thin film, as well as to the critical velocity of the transition from stick-slip to smooth sliding, which differ by many orders of magnitude from the values observed in macroscopic experiments. This contradiction can be resolved with the help of the earthquakelike (EQ) model with a continuous distribution of static thresholds. The evolution of the EQ model is reduced to a master equation which can be solved analytically. This approach describes stick-slip and smooth sliding regimes of tribological systems within a framework which separates the calculation of the friction force from the atomic-scale studies of contact properties.

keywords Boundary lubrication · Nanotribology · Viscosity · Stick-slip

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1 Introduction

Science about friction, or tribology (from Greek *tribos*, translated as rubbing) is very important both from scientific and practical points of view. We remind that one has to distinguish between the static and kinetic friction. The static friction force f_s is the force that has to be applied to initiate sliding. Namely thanks to static friction, we are able to walk and cars to move, as well as mechanical constructions connected by bolts and nuts are stable. The kinetic friction force f_k is the force needed to maintain the smooth sliding with a given velocity v . Therefore, the energy $f_k v$ is pumped into the system per every time unit, and is converted into heat, finally leading to heating of atmosphere. According to estimation [1], the losses of energy because of friction achieve more than 6% of the gross national income in economically developed countries. Therefore, even a small reduction of friction promises an enormous economical effect. In typical situations $f_k < f_s$, e.g., $f_k \sim 0.5 f_s$, and the ratio of the friction force to the loading force f_l , $\mu_{s,k} = f_{s,k}/f_l$, known as the friction coefficient, is typically of the order of magnitude of $\mu \sim 0.1$.

Because of importance of friction, its study began more than four centuries ago [2]. The first known study of friction belongs to Leonardo da Vinci (1452–1519) who found that the friction coefficient does not depend on the area of contact. Later Guillaume Amontons (1663–1705) showed that the friction is directly proportional to the load. Leonhard Euler (1707–1783) noted that one has to distinguish between the static friction studied by Vinci, and the kinetic friction explored by Amontons. Finally, Charles Coulomb (1736–1806) discovered that the kinetic friction does not depend on the sliding speed.

These laws, getting the name of the Amontons laws, remained purely empirical up to the middle of the last

century, when Bowder and Tabor [3] made the first attempt of their explanation from the physical viewpoint. They noted that the contacting surfaces are rough typically. Therefore, the real contact is attained only on asperities. An estimation [1] shows that the real area of contact A_{real} makes only $\sim 10^{-5}$ of the geometrical (visible) area A_{vis} . The contacts themselves are under the extremal condition of enormous pressure—forces in the contacts are close to the limit of plasticity of the materials that form the contact. This explains the Amontons laws: with the increase of the load f_1 , the real contact area grows either due to the increase of the number of contacts (in the regime of elastic response of the system), or because of the plastic deformation of contacts [4]. As a result, the ratio $\mu = f_{s,k}/f_1$ remains approximately constant.

Later, more careful experiments showed that the Amontons laws are valid approximately only [1, 5–9]. First, friction does depend on the sliding velocity. Second, it depends on a prehistory of the contact, i.e., friction is different for a “newborn” contact and for the contact which already undergone some sliding. A new era in the study of friction began only two decades ago thanks to the development of new experimental methods (first of all, the “tip-based technologies” coming from surface physics—the scanning tunnel microscope (STM) [10] and its subsequent improvements—the atomic force microscope (AFM) [11] and the friction force microscope (FFM) [12]), and also due to the great progress in computer power allowing the molecular dynamics (MD) simulation of real tribosystems.

In this brief survey we present a modern view on the problem of friction from the physical viewpoint. First, in Sect. 2 we mention two most popular simple models explored in tribology, the Tomlinson model and the Frenkel-Kontorova model. Then, in Sect. 3 we shortly discuss the study of kinetic friction by the molecular dynamics method. We consider the regime of boundary lubrication only, when the surfaces are separated by a very thin, of few monomolecular layers, lubricant film. Note that such a film is almost always present: it may be either a specially chosen lubricant, grease (oil), dust, wear debris produced by sliding, water, or a thin layer of hydrocarbon adsorbed from air, etc.—all these are called the “third bodies” in tribology. Moreover, even if the lubricant film is thick, at the moments of the onset of motion or at its stop, the lubricant is squeezed out from the contact area, and the system turns into the regime of boundary lubrication. Atomic-scale MD simulations of friction, however, lead to the following problems: the “viscosity” of the thin film, as well as the critical velocity of the transition from stick-slip to smooth sliding, differ by many orders of magnitude from typical values observed in macroscopic experiments. This contradiction can be resolved with the help of the earthquakelike (EQ) model

with a continuous distribution of static thresholds discussed in Sect. 4. The evolution of the EQ model is reduced to a master equation which can be solved analytically. This approach describes the stick-slip and smooth sliding regimes of tribological systems within a framework which separates the calculation of the friction force from the atomic-scale studies of contact properties. Finally, Sect. 5 concludes the paper with the discussion of other important questions related to tribology.

2 Simple Models of Friction

In physics, a very large role is played by simple models, which, on the one hand, correctly describe the basic aspects of the problem and, on the other hand, allow either the exact solution or at least a well grounded one with a predicted accuracy. In tribology, there are two such models—the Tomlinson model [13] (see Fig. 1, left panel) and the Frenkel-Kontorova (FK) model [14], schematically shown in Fig. 1 (right panel). Later, a number of generalized and combined models were proposed as well (e.g., see [15]). However, already the simplest model—a single atom placed into the external periodic potential—allows us to understand some important aspects of friction.

If we apply a constant force f to the atom, it will remain at rest (in a local minimum of the potential $V(x) = \sin x - fx$) whilst $f < f_s = 1$; thus, the force f_s is an analog of the static friction force. At $f > f_s$ the atom begins to slide over the potential relief. However, if we now reduce the force, the smooth sliding will survive up to the force $f = f_b \approx \eta\sqrt{M}$ (here η is the damping coefficient and M is the atomic mass), as the atom can overcome the maxima of the potential relief due to its inertia. The force f_b is the analog of the kinetic friction force. The important result is that the minimal speed v_b , at which the atom can slide due to inertia, is of the “atomic-scale” order, $v_b \sim 50 \text{ \AA/ns} = 5 \text{ m/s}$.

In the tribosystem, the periodic potential corresponds, e.g., to the surface potential of the lower (immobile) substrate, while the “atom”, to the moving top substrate. As $v_b \propto M^{-1/2}$, one may speculatively suppose that in a macroscopically large system, when $M \rightarrow \infty$, we will obtain $v_b \rightarrow 0$. This assumption, however, is wrong [16, 17]. Upon the reduction of the driving force, first the lowest atomic

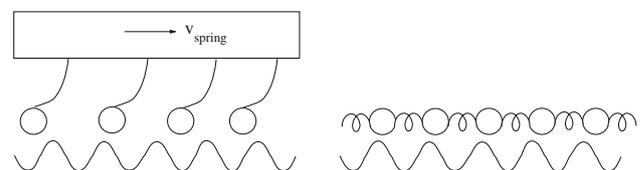


Fig. 1 The Tomlinson model (*left*) and the Frenkel-Kontorova model (*right*)

layer of the top substrate (i.e., the layer nearest to the contact) is stopped, and this occurs at a speed of the atomic-scale order. At this moment, a stopping wave is created, and then the second, third, etc. atomic layers of the top substrate stop successively one after another.

If we now push the atom not directly, but through a spring (which describes, e.g., the elasticity of the top substrate), the end of the spring moving with the velocity v , we come to the Tomlinson model. At $v > v_b$ the system will demonstrate smooth sliding, and at lower speeds $v < v_b$, the so-called stick-slip motion, well known as door creak, bowing a violin, etc. Namely, with the onset of motion the spring stretches and the driving force grows, until it reaches the static threshold f_s . At this moment the system begins to move with the increasing speed, until it catches the end of the spring; thus, the spring is weakened again, and the driving force falls down. As a result, the system slows down up to the complete stop, and the whole process repeats itself. In the stick-slip regime, the friction force does not depend on the driving velocity; however, if the system temperature is nonzero, there is a weak (logarithmic) dependence $f(v)$ because of thermally activated jumps of lubricant atoms [1] (another reason for the velocity dependence comes from the aging of contacts, see Sect. 4).

The second important model widely used in tribology is the Frenkel-Kontorova model. First it was proposed for description of dislocations in solids, and then it was widely used in surface physics for description of commensurate and incommensurate structures of films adsorbed on a surface [18]. Generally, a great progress in understanding of friction problems, attained lately, first of all is governed by the progress in surface physics.

The FK model describes a chain of interacting atoms (e.g., adsorbed or lubricant atoms), placed in the external periodic potential created by the surface atoms of the substrate.

The use of the FK model was so successful because in the continuum limit (valid for strong interaction between the atoms) its equations of motion reduce to the exactly integrable sine-Gordon (SG) equation. The solutions of the SG equation, besides the linear waves (phonons), include the topological solitons (so-called “kinks”) and dynamical solitons (“breathers”). The kink describes a spatially localized compression of the chain (or its extension in the case of the antikink), and is characterized by extremely high mobility. Namely kinks are responsible for the fast mass transport along the chain, i.e., for the mobility of the adsorbed or lubricant film. In two-dimensional (2D) or three-dimensional (3D) systems, concepts of domain walls or misfit dislocations are used instead of kinks, but the physics of the processes remains qualitatively the same. For example, the mechanism of motion of a finite chain (or

an island in the 2D system) is the following: a kink is created at one (free) end of the chain, then moves rapidly along the chain and annihilates at the chain’s other end; as a result, the whole chain is displaced by the distance of one lattice spacing [19].

Another extremely important concept of the FK model is the “incommensurability”. Namely, if the lattice constants of the chain a and the substrate a_s in the infinite system are incommensurate (i.e., their ratio $\chi = a/a_s$ is irrational), there always exists a critical value of the elastic constant of the chain g , such that for a higher rigidity the chain becomes effectively free of the substrate, i.e., the static friction falls to zero, and the kinetic friction becomes very small. This phenomenon (known in physics from the beginning of 1970th as the Aubry transition, or “the transition by breaking of analyticity” [20–24]) acquired an extreme actuality in tribology in connection with the prediction of “superlubricity” [25], i.e., the existence of lubricants providing extremely low friction. In the FK model the best condition for appearance of the state with $f_s = 0$ is achieved at the “golden ratio” incommensurability $\chi = (\sqrt{5} - 1)/2$. If the chain is placed between two 1D “surfaces”, the so-called “spiral ratio” of lattice constants turns out the best [26].

Thus, the simple models already provide answers to the basic questions of tribology, at least at a qualitative level. For example, it is clear that a solid lubricant could be the most effective: it should provide the maximal friction in the case of commensurate surface/lubricant interface (the so-called “cold welding of contacts”) and minimal (up to zero) friction, for the incommensurate interface.

3 MD Simulation of Friction

First of all we note that in order to obtain realistic values of the friction forces, the model system must be three-dimensional, because the basic mechanism of energy losses at sliding is excitation of phonons [1, 9]. The rate of this process is directly proportional to the density of phonon states which cannot be correctly described by a one- or two-dimensional model system. In other, the modelling of tribosystems is carried out by standard MD methods. The bottom and top substrates are modelled by one or a few atomic layers each, and the lubricant atoms (or molecules) are placed between the substrates (see Fig. 2). It is assumed that all atoms interact among themselves. The interaction is described, e.g., by the Lennard-Jones potential or by a more realistic for the given system potential. The periodic boundary conditions are used in the longitudinal directions x and y . The bottom substrate is fixed (immobile), and a load force and the driving force are applied to the top substrate, usually through a spring, whose end is driven

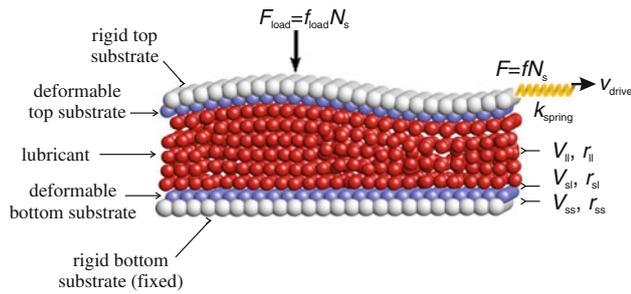


Fig. 2 A typical model for MD simulation of friction

with a given velocity v . During simulation the spring force, which corresponds to the friction force, as well as other parameters, such as the thickness of the lubricant film, its structure, distribution of temperature and atomic velocities through the contact, etc., are monitored.

The modelling of tribosystems has, however, two important features. First, as the number of lubricant atoms N is fixed, the results of simulation may be sensitive to this number, for example, whether the lubricant atoms form exactly two atomic layers or two layers and a half. To reduce related errors, one may make one or both surfaces “corrugated” as shown in Fig. 2 (that also is closer to real situations, where surfaces are often rough). This method was first realized by Persson and Ballone [27]. Besides, it is desirable to make a set of MD runs with different numbers N .

The second problem in modelling of friction is more serious. We remind that a tribological system is a “machine” to convert the energy of translational motion into heat. The driving pumps energy into the system, and if we do not remove it, soon the system will simply evaporate or burst. Therefore, the use of solely Newtonian equations of motion is impossible; it will be incorrect also to use artificial methods of removing energy (such as, e.g., the widely used method of renormalization of atomic velocities after a given number of MD steps), as the rate of energy removal will finally determine the kinetic friction force. A typical receipt in such situations is to model the substrates as made of many atomic layers, and then to use the Langevin equations with the damping coefficient smoothly increasing with the distance from the interface, thus modelling an effectively “infinite” substrate [17]. However, this method leads to a catastrophic increase of the system size, the more so unjustified, that in the end only the trajectories of lubricant atoms are of real interest.

The increase in computational time can be strongly reduced with the help of a multi-scale approach proposed by Persson et al. [28, 29]. In this method, the number of “atoms” in the substrate layers decreases with an increase of the distance from the interface, while the springs between the “atoms” become longer the further away from the interface they are located, and the “atomic” masses are

increased in such a way as to keep a fixed mass density of the substrate. This technique was successfully applied in contact mechanics as well as in a study of a (slow) squeezing of the lubricant away from the contact zone. Unfortunately, a study of dynamics of a (fast) sliding processes would lead to less reliable results, because this model cannot reproduce the substrate phonon spectrum rigorously.

Accurate results for the interface dynamics can be obtained with the help of a hybrid simulation method [30]. This method combines an atomistic treatment of the interfacial region with a finite-element method description of subsurface deformations. The dynamics in the two regions are coupled through displacement boundary conditions applied at the outer edges of an overlap region. The two solutions are followed concurrently but with different time resolution. Unfortunately, this method still requires a huge computer power—the time saving is about a factor of 20 in comparison with full MD simulation.

As a simpler solution of the problem, it was proposed to use the Langevin equations for all lubricant and substrate atoms, but with a “realistic” damping coefficient η , which depends on the coordinate r and velocity v of the given atom relative the surfaces in contact, and correctly describes the energy exchange between the moving atom and the substrates [31]. For the dependence $\eta(r, v)$, one can use the expression found for the adatom vibrating near the surface [32, 33]. Of course, the extension of the dependence obtained for vibration of a single adatom to the system of interacting moving lubricant atoms, may lead to some errors, but this approach should be much better than the one that uses an arbitrary constant for the damping coefficient η [6]. The use of the velocity-dependent damping coefficient requires in turn a substantial development of the method of stochastic equations, as has been done in [34].

The MD simulation showed [9, 31] that the basic factor which determines the behavior of a tribosystem, is the relation between the amplitude of interatomic interaction inside the lubricant V_{II} and the interaction of lubricant atoms with the substrate V_{sl} . In the case of traditional (e.g., oil) lubricants, the inequality $V_{II} < V_{sl}$ holds, i.e., the lubricant atoms are coupled to the surfaces much stronger, than among themselves; it is the so-called “soft” tribosystem. In the opposite case of the “hard” system, the interaction of lubricant atoms among themselves is strong, $V_{II} > V_{sl}$, and as a result, the lubricant remains in the solid state even during sliding.

3.1 Melting of a Thin Lubricant Film

As is well known from surface physics, mechanisms of melting of the monoatomic film adsorbed on a crystal surface essentially differ from those in bulk, and are

characterized by a large variety [18]. The same is true for the lubricant film confined between two surfaces. Typically, the temperature of melting of the lubricant film T_c is higher, than the bulk melting temperature T_v [5, 35]. The value of T_c monotonically decreases with the increase of the number of layers N_1 in the film and approaches the bulk value only for $N_1 > 5$. Such a behavior is related to the limitation of motion of lubricant atoms in the z -direction because of the contact with the surfaces.

The mechanisms of melting of the hard and soft lubricants are also different [35]. In the hard tribosystem, the lubricant atoms in contact with the substrates can vibrate with a larger amplitude than in the middle of the film; therefore, the melting begins from the boundary layers (this is an analog of “surface melting”). In the opposite case of the soft lubricant, where the boundary layers are strongly coupled to the substrates, the melting begins from the middle of the film. The $T_c(N_1)$ dependence obtained with the help of MD simulations can be well explained theoretically with the help of the Lindemann criterion [35].

Properties of the molten lubricant film differ from those of the bulk liquid—the former exhibits a well defined layered structure, which is preserved at sliding as well. Although the discovery of this fact caused a surprise in tribology community, from the surface physics viewpoint this phenomenon is obvious: the surfaces impose a structure to the near-by layers of the liquid lubricant.

It is interesting that in the solid state at $T < T_c$ the film structure also substantially differs from that in the bulk: although the state is “solid”, as the shear module is non-zero, mobility of atoms in the film considerably exceeds that in the bulk, as characteristic for the same temperature. It is related to the presence of a large number of defects (e.g., vacancies) in the confined film, so that the state of the film is closer to a glasslike than to the ideally crystalline.

The lubricant film may also be melted not because of the rise of temperature, but due to sliding (the sliding-induced melting). The mechanism of this melting, however, is different from that described above [36]. For a submonolayer lubricant film driven over a substrate periodic potential, sliding-induced melting was first discussed by Persson [37]. It was found that the film may split into molten (liquid) and solid domains; this fact is important for a mesoscopic model of friction as discussed below in Sect. 4.

3.2 Kinetic Friction

Although it may seem strange, the problem of static friction is considerably more difficult, than the problem of kinetic friction. For example, theory predicts [38] that the static friction should almost always be zero for the contact of two elastic substrates, in conflict with known experiments. The difficulty of f_s calculation emerges because the static friction

is determined by the concrete structure of the contact, which may be quite complicated and poorly defined (for example, it is supposed that it rather corresponds to a glasslike structure). Besides, the values of thresholds may differ from contact to contact, as well as change in time (typically f_s grows with the time of stationary contact—the so-called aging of contacts—probably, because of their plastic deformation). Moreover, at a nonzero temperature the sliding persists at any nonzero shear stress (the so-called creep motion) so that formally $f_s = 0$ [7]. In general, an accurate description of static friction may be done within a mesoscopic model discussed in Sect. 4. Therefore, below we discuss the problem of kinetic friction only, i.e., the regime of smooth sliding, when the system is in the well defined steady state. We remind that this regime corresponds to quite high sliding velocities, $v > v_c \sim 1$ m/s.

In the case of a traditional (oil-based) lubricant, or the soft tribosystem, the boundary layers of the lubricant film are strongly coupled to the surfaces and, therefore, sliding must begin with rupture of bonds somewhere in the middle of the film. As a result, the film is melted with the onset of motion, and remains liquid in the smooth sliding regime (at $v > v_c$) as well as in the sliding phase of the stick-slip regime at $v < v_c$. In the latter case, the film freezes again during the stop of motion; such a mechanism of stick-slip motion is known as the melting-freezing mechanism [39, 40]. In the smooth sliding regime, the liquid state of the film is supported due to its heating because of sliding. However, a detailed MD study shows that there are two different sliding regimes, the “liquid-sliding” (LS) regime described above, and the “layer-over-layer” sliding (LoLS) regime, when the lubricant layers keep an ordered structure at sliding (these two regimes remind the turbulent and laminar flows in hydrodynamics). The friction coefficient in the soft system takes on intermediate values of the order of $\mu \sim 0.1$, and μ is directly proportional to the lubricant viscosity, which for a thin film may be either lower or 2–3 times higher than the bulk viscosity [9, 31].

On the other hand, in the case of a solid lubricant, or the hard tribosystem, the sliding takes place at the surface/lubricant interface. And, if the surface and the hard lubricant have an ideal crystalline structure, we get the system with extremely low friction. Indeed, the substrate and lubricant are typically made of different materials, therefore they have different lattice periods, incommensurate in a general case. But even if the periods coincide, the commensurate interface in the 2D contact can be formed only if the axes of these two lattices are strictly aligned, because any, even the smallest dismiss of the axes will result in incommensurability of the lattices. Then, the substrate and lubricant are usually rigid enough, so that their rigidities are above the Aubry threshold. Therefore, the regime of extremely low friction should be realized practically always if, we emphasize, the substrates and the solid

lubricant have the crystalline structure [9, 31]. Namely this fact explains very good characteristics of the graphite-based lubricants as well as other layered materials such as MoS_2 and Ti_3SiC_2 . Extremely low friction is indeed observed experimentally, for example, during scanning of the W(011) tip on the Si(001) surface [41], or for sliding of a graphite flake on the graphite surface [42, 43].

However, the above-described advantages of the solid lubricant disappear completely if the contacting surfaces are not ideal, for example, if there are steps, asperities or other defects on the surfaces, where the pinning (hooking) of the surfaces takes place. In particular, a rather detailed study due to Persson et al. [28] showed that relatively small surface roughness (of an amplitude $> r_{\text{sim}} 0.1 \text{ \AA}$) may completely kill the superlubricity. At depinning from the defects, the lubricant may be melted, and then, during the stop, it will be solidified again, but now with an amorphous-like structure, as the cooling of the confined film is very rapid due to a good thermal contact with the substrates. In the case of imperfect (amorphous or glasslike) structure of the solid lubricant film, the friction is rather large, larger than for liquid lubricants [9, 31].

Nevertheless, the good tribological characteristics of the solid lubricant can be recovered by the careful choice of its parameters. We remind that the lubricant is heated to some temperature T_{slide} at sliding, and also that its melting temperature T_c is proportional to the amplitude of the interatomic interaction V_{II} . If we choose the V_{II} parameter so that T_{slide} is close to T_c (but $T_{\text{slide}} < T_c$), structural defects of the film may be annealed, and the film may self-order during sliding. The MD simulation [44] confirms the possibility of such a scenario: starting from an imperfect film, we observe the stick-slip motion. The film temperature sharply rises during the phase of sliding, the film self-orders remaining in the solid state and, after a few stages of sliding, the system passes to the smooth sliding regime with a very low friction. For a realization of the self-ordering mechanism, it is necessary to choose the amplitude V_{II} of the interaction large enough, so that the film is not melted during sliding, but not too large, so that the rate of annealing of defects is not too low.

The results of MD modelling also allowed us to develop the phenomenological theory of kinetic friction [9, 45], by which it is possible to predict analytically the behavior of the tribosystem with the change of its parameters.

4 From Atomic-scale to Meso- and Macroscopic Friction

The described above atomic-scale mechanisms of friction are important in designing nano-mechanical devices. However, there arises a question of are they in any relation

with the friction at the meso- and macro-scales. First of all, the value of the critical velocity of transition from stick-slip to smooth sliding causes suspicion: experimentally, the transition is observed at velocities $\sim 1\text{--}10 \mu\text{m/s}$ [1, 46], while the MD simulations lead to values $v_c \sim 1 \text{ m/s}$ and, as proved in [17], this value cannot be made substantially lower. The second problem is related to the viscosity of the thin lubricant film: MD simulations predict that the viscosity of the film may be higher than the bulk value by a factor of 2–3 only, while the experiment shows their difference by many orders of magnitude. These two problems, however, are linked together. Indeed, viscosity of the thin film is formally defined as fd/va_s^2 , where d is the film thickness and a_s is the substrate lattice constant [9]. If we insert into this expression, instead of v , the value v_c taken from experiments, we obtain the huge increase of viscosity as declared by experimentalists.

Both of these contradictions can be resolved within the model originally introduced to describe earthquakes [47] and then adopted to friction by Persson [48]. Physics of both processes is qualitatively the same, but differs by the spatio-temporal scale—nanometers and seconds to hours in tribology in comparison to kilometers and years to centuries in geology.

In the earthquake (EQ) model, the sliding interface is treated as a set of N_c “contacts” with the average rigidity $\langle k_c \rangle$ as shown in Fig. 3. The contacts represent, for example, asperities for the interface of rough surfaces [4], or patches of lubricant or its domains (“solid islands” [37]) in the case of lubricated friction. The i -th contact connects the slider and the bottom substrate through a spring of the elastic constant k_i . When the slider moves, the contact position changes, and the contact’s spring elongates or shortens, so that the slider experiences from the interface the force $-F = \sum f_i$, where $f_i = k_i x_i$ and $x_i(t)$ is the spring length. The contacts are coupled frictionally to the slider and behave according to MD simulations or FFM experiments. Namely, as long as the force $|f_i|$ is below a certain threshold f_{si} (corresponding to plastic flow of the entangled asperity, or to local shear-induced melting of the boundary lubrication layer), this

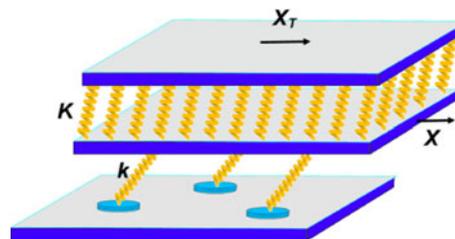


Fig. 3 The earthquake model—third from the basic models used in tribology

contact moves together with the slider. When the force exceeds the threshold, the contact detaches from the slider, and then re-attaches again in the unstressed state after some delay time τ . Thus, with every contact we associate the threshold value f_{si} , which takes random values from a distribution $\tilde{P}_c(f)$ with the mean value f_s and the standard deviation Δf_s . The spring constants are related to the threshold forces by the relationship $k_i = \langle k_c \rangle (f_{si}/f_s)^{1/2}$, because the value of the static threshold is proportional to the area A_i of a given contact, while the transverse rigidity k_i is proportional to contact's size, $k_i \propto \sqrt{A_i}$. When the contact is “reborn” (i.e., re-attached to the slider), new values for its parameters have to be assigned. Also one may take into account the elastic interaction between the contacts, so that the relaxation of one contact causes redistribution of forces on the other contacts, and that can provoke their relaxation as well or even cause an avalanche of relaxations.

Rather than studying the evolution of the EQ model by numerical simulation it is possible to describe it analytically [49]. Let $P_c(x)$ be the normalized probability distribution of values of the thresholds x_{si} at which contacts break; it is coupled with the distribution of threshold forces by the relationship $P_c(x)dx = \tilde{P}_c(f)df$. To describe the evolution of the model, we introduce the distribution $Q(x;X)$ of the stretchings x_i when the bottom of the solid block is at a position X . Let us consider a small displacement $\Delta X > 0$ of the bottom of the solid block. It induces a variation of the stretching x_i of the contacts which has the same value ΔX for all contacts. The displacement X leads to three kinds of changes in the distribution $Q(x;X)$: first, there is a shift due to the global increase of the stretching of the asperities; second, some contacts break because the stretching exceeds the maximum value that they can withstand; and third, those broken contacts form again, at a lower stretching, after a slip at the scale of the asperities, which locally reduces the tension within the corresponding asperities. These three contributions can be written as a master equation (ME) for $Q(x;X)$: $Q(x;X + \Delta X) = Q(x - \Delta X; X) - \Delta Q_-(x; X) + \Delta Q_+(x; X)$. The first term in the r.h.s. of this equation is just the shift. The second term $\Delta Q_-(x; X)$ designates the variation of the distribution due to the breaking of some contacts. It can be written as

$$\Delta Q_-(x; X) = P(x)\Delta X Q(x; X), \tag{1}$$

where $P(x)\Delta X$ is the fraction of contacts that break when the position changes from X to $X + \Delta X$. According to the definition of $P_c(x)$ the total number of unbroken contacts when the stretching of the asperities is equal to x is given by $N_c \int_x^\infty P_c(\xi) d\xi$. The contacts that break when X increases by ΔX , are those which have their thresholds between x and $x + \Delta X$, i.e., $N_c P_c(x)\Delta X$. Thus,

$$P(x) = P_c(x) \int_x^\infty d\xi P_c(\xi). \tag{2}$$

The broken contacts relax and have to be added to the distribution at $x = 0$, leading to the third term in the master equation, $\Delta Q_+(x; X) = \delta(x) \int_{-\infty}^\infty d\xi \Delta Q_-(\xi; X)$. Finally, taking the limit $\Delta X \rightarrow 0$, we get the integro-differential equation

$$\begin{aligned} \frac{\partial Q(x; X)}{\partial x} + \frac{\partial Q(x; X)}{\partial X} + P(x)Q(x; X) \\ = \delta(x) \int_{-\infty}^\infty d\xi P(\xi)Q(\xi; X), \end{aligned} \tag{3}$$

which has to be solved with the initial condition $Q(x;0) = Q_{ini}(x)$.

The evolution of the system in the quasi-static limit where inertia effects are neglected shows that, in the long term, the initial distribution approaches a stationary distribution $Q_s(x)$ and the total force F becomes independent of X . The final distribution is independent of the initial one (the mathematical proof of this statement for a simplified version of the EQ model was presented in [50]). The statement is valid for any distribution $P_c(x)$ except for the singular case of $P_c(x) = \delta(x - x_s)$. The steady-state, or smooth-sliding solution of Eq. 3 can easily be found. It can be expressed as

$$Q_s(x) = C^{-1} \Theta(x) E_P(x), \tag{4}$$

where $\Theta(x)$ is the Heaviside step function, $E_P(x) = \exp[-U(x)]$, $U(x) = \int_0^x d\xi P(\xi)$, and $C = \int_0^\infty dx E_P(x)$.

The dynamics of the EQ model is determined by the following factors: by the ratio $D = \Delta f_s/f_s$, by the relation between the rigidity of the driving spring K (or the elasticity of the slider) and the interface rigidity $K_c = N_c \langle k_c \rangle$, and by the delay time τ . If the interface is soft, $K_c < K^*$, where $K^* \sim KD$, the force $F(X)$ always approaches the stationary value $\langle F \rangle < N_c f_s$ corresponded to the “kinetic” friction force, undergoing oscillations of the amplitude decaying as $\propto \exp(-D \langle k_c \rangle X/f_s)$. Thus, the amplitude of oscillations is large when $D \ll 1$, while in the case of $D \sim 1$ the force $F(X)$ quickly achieves the value $\langle F \rangle$. On the other hand, for the hard interface, $K_c > K^*$, the block undergoes the elastic instability [8] and, if $\tau > 0$, the function $F(X)$ becomes periodic [51–53]; this corresponds to the stick-slip motion of the slider.

It is important also to incorporate the contact's aging in the EQ model—the threshold f_{si} should increase with the contact lifetime counted off from the moment of its last sliding. The aging leads to a dependence of system dynamics on the driving velocity [51]. Indeed, at a low velocity, when all contacts have enough time to “grow

old” and attain approximately the same threshold value, depinning of contacts takes place almost simultaneously over the whole system, i.e., their motion is synchronized. It is the stick-slip regime observed in macroscopic experiments. At a high sliding velocity, the threshold values f_{si} for different contacts are different, therefore they move asynchronously and, as a result, the averaged spring force is approximately constant. In the macroscopic experiment, such a motion looks like “smooth sliding” at which, however, the contacts themselves undergo the stick-slip motion. Moreover, the gradual breaking (melting) of different microscopic contacts (domains) slows down the detachment which initiates the slip [53]. As a result, the slowing of slips in the stick-slip regime is not determined by the huge lubricant viscosity η as was supposed recently [54]. Thus, the EQ model with a distribution of static thresholds resolves both the problems mentioned above related to the viscosity of the thin film and the critical velocity of the stick-slip to smooth sliding transition.

The distribution $\tilde{P}_c(f)$ can be straightforwardly calculated for the contact of rough surfaces [4]. However, in the SFA (surface force apparatus [55]) or SFB (surface force balance [56, 54]) experiments, where the sliding surfaces are made of mica, the interface may be atomically flat over a macroscopic area. But even in this case the lubricant film cannot be ideally homogeneous throughout the whole contact area—it should be split into domains, e.g., with different orientation, because this will lower the system free energy due to the increase of entropy. Domains of different orientations have different values for the thresholds f_{si} , i.e., they play the same role as asperities in the contact of rough surfaces.

The described above ME formalism can be extended to take into account various generalizations of the EQ model. For example, this approach can accommodate temperature effects which enter through their effect on the distribution $P(x)$, because thermal fluctuations allow an activated breaking of a contact which is still below the threshold [48, 52, 57, 58]. When $T > 0$, the friction force depends on the sliding velocity as $F(v) \propto v$ at low speeds (this effect may be interpreted as an effective “viscosity” of a thin lubricant film which, however, has nothing in common with the bulk viscosity) and $F \propto \ln v/v_0$ [48, 7, 57] or $F(v) \propto (\ln v/v_0)^{2/3}$ [52, 58] at intermediate velocities. Also, the ME formalism can incorporate the delay in contact formation [52, 57, 58] and the aging of the contacts [51, 52, 58]. In this case the master equation for $Q(x, X)$ must be completed by an equation for the evolution of $P_c(x)$, which in turns affects $P(x)$. Typically, due to aging effects the friction force should decrease when the sliding speed increases (roughly as $F \propto v^{-1}$), i.e., these effects are concurrent to the temperature ones. Taking also into account that the delay times and the rate of aging

depend on temperature, one may obtain a nonmonotonic temperature dependence of friction with a peak at cryogenic temperatures [58]. Moreover, one can take into account the elastic deformation of the interface by introducing a position-dependent distribution $Q[x, X(\mathbf{r})]$ where \mathbf{r} denotes the position of an asperity on the interface, and $X(\mathbf{r})$ the local translation of the interface averaged over a mesoscopic scale. The master equation must then be coupled to an equation describing the elastic deformation of the interface, subjected to the contact forces at each point \mathbf{r} . A preliminary work in this direction [59] already allowed us to describe a nontrivial scenario of the onset of sliding. This illustrates the new viewpoint introduced by the ME approach which describes the phenomena at an intermediate scale between the microscopic scales of the contacts and the macroscopic scale of the displacement of the solid block. The ME equation only has a meaning if, on this intermediate scale, there are many individual contacts, allowing us to study them as a statistical distribution.

5 Conclusion

Thus, the complex problem of friction is split into sub-problems that can be studied separately, the statistical properties of the contacts, and the evolution of the distribution Q which is described by the master equation, coupled to additional equations representing different effects such as the elastic interactions between asperities, the aging of the contacts or temperature fluctuations. The atomistic MD simulations provide the information on microscopic properties of individual contacts (asperities, domains, etc.), namely, the distribution of static thresholds $P_c(x)$ and its evolution with time due to aging effects. Then, the evolution of the system on the mesoscopic scale may be described with the help of the master equation approach. An incorporation of the elastic deformation of the substrates should finally result in a complete theory of wearless friction.

The majority of the results described above, as well as many others not included in this mini-review because of the lack of space, were obtained just in the last decade. This indicates the fast progress in nanotribology. However, further improvement of experimental methods, able to fix not only the average friction force but provide also a detailed information about processes inside the lubricant film, is still extraordinarily actual. In this context, the use of methods already well developed in surface physics, looks very promising. For example, an important information on the energy exchange in adsorbed films can be obtained by the quartz crystal microbalance method [60]. From other interesting new experimental methods, we

mention the ones used in works [61–66], and also a recently developed technique of “levitating substrate”, where a sliding block holds out above the surface by the magnetic field [67].

From the problems not considered above, first of all we must mention the searching of methods to control and operate the friction either chemically by adding specially chosen molecules to the base lubricant [68], or by mechanical methods, for example, using special nanopatterned surfaces [69] or applying an external oscillating force to the system [70–77].

We also did not touch the important question of the shape of lubricant molecules. In the case of superlubricity, the form of molecules of the hard lubricant is not important, as the interface structure plays the main role. But in the case of traditional (soft) tribosystems, the kinetic friction force is directly proportional to viscosity of the liquid lubricant; therefore, the lower is the viscosity, the smaller should be the friction. An example known from the times of ancient Egypt but recently acquired the special actuality in connection with development of nano-mechanical devices, is the use of water [78, 79] or water solutions [80] as lubricants.

One has to note also that in the process of evolution, the nature chose namely water solutions as lubricants in living organisms. However, daily experience says just the opposite—it prompts that a liquid with a high viscosity is often a better lubricant. This is related to squeezing of the lubricant out of the contact area: the higher is the viscosity, the slower is the rate of squeezing out [1]. More rigorously, the important issue is not the lubricant viscosity, but the length L of lubricant molecules: the longer is the molecule, the by the greater number of the atoms it holds on the surface, and the more difficult is to remove it out from the contact area [81]. In some systems, however, the dependence $\mu(L)$ may be nonmonotonic [82]. Another interesting idea is to use advantages of rolling friction, e.g., to use the spherical molecules C_{60} (fullerenes) as a lubricant [83–85]—as is well known, in macroscopic systems the friction coefficient of rolling is $10^2 - 10^3$ times lower than that of sliding friction. For example, lately it becomes actual to design micro-bearing using carbon nanotubes or fullerenes as rollers.

In conclusion we emphasize that the problem of friction requires the coordinated intra-disciplinary efforts from the side of physicists, chemists, material scientists, and mechanics, and then we may expect a great progress in tribology in the nearest future.

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