

The nonlinear nature of friction

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Tribology is the study of adhesion, friction, lubrication and wear of surfaces in relative motion. It remains as important today as it was in ancient times, arising in the fields of physics, chemistry, geology, biology and engineering. The more we learn about tribology the more complex it appears. Nevertheless, recent experiments coupled to theoretical modelling have made great advances in unifying apparently diverse phenomena and revealed many subtle and often non-intuitive aspects of matter in motion, which stem from the nonlinear nature of the problem.

Friction plays a central role in diverse systems and phenomena that at first sight may seem unrelated, but which on closer scrutiny are found to display common features that are shared by all tribological processes, in technological, geological or biological areas. The development of durable and/or low-friction surfaces and thin lubricating films has become an important factor in the miniaturization of moving components in many technological devices. These include micro-electro-mechanical systems (MEMS), computer recording systems and miniature motors with small loads. The old, simple empirical laws of friction do not always hold in such systems; this is due to their high surface-to-volume ratio and the greater importance of surface chemistry, adhesion and surface structure or roughness. Conventional tribological and lubrication techniques used for large objects can be ineffective at the nanometre scale, which requires new methods for control. Another rapidly growing area of tribology is in biosystems, and particularly the lubrication mechanisms in joints. Through the process of natural selection, nature has produced water-based lubricant systems that far outclass the best oil-based lubricants of most man-made devices; to emulate these systems is one of today's great challenges.

At the conceptual and theoretical levels, however, recent advances have revealed the enormous complexity of even the simplest tribological process. Friction is intimately related to both adhesion and wear, and all three require an understanding of highly non-equilibrium processes occurring at the molecular level to determine what happens at the macroscopic level. Surfaces can be smooth or rough, hard or soft, elastic, viscoelastic or plastic, brittle or ductile, dry (unlubricated) or lubricated, and of very different chemistries. The multitude of asperities on two shearing surfaces are constantly coming into and out of contact, where the local pressure between them can fluctuate between ~ 1 Pa (10^{-5} atmospheres of pressure) and GPa (10^4 atmospheres) within microseconds. These are extreme conditions that cannot always be treated by simple 'linear' theories.

Modern views of friction

To understand the behaviour of two real surfaces in relative motion while still in contact, we need to look into what is going on at the 'single asperity' level. With the advent of the atomic force microscope¹ (AFM) and the surface forces apparatus² (SFA) it became possible to study individual sliding junctions at the molecular level. The AFM and SFA are ideal tools in nano-, micro- and macroscopic tribological experiments for measuring the normal and lateral forces, and wear, between (1) a nanometre-radius tip or micrometre-sized colloidal particle against a substrate surface, and (2) with the SFA, two macroscopic molecularly smooth or rough surfaces of measurable molecular contact area that confine a lubricant film of measurable thickness.

Three theoretical approaches that have been introduced to

investigate frictional forces in sheared systems are illustrated in Fig. 1: large-scale molecular dynamics (MD) simulations^{3–9}, phenomenological rate–state (RS) models^{10–15}, and 'minimalistic' models (MM)^{6–18}. Each approach has its advantages and disadvantages, and a different emphasis.

A tribological model is expected to recover certain key experimental observations, some of which are shown in the left panels of Fig. 2, as (1) structural transitions in thin lubricating liquid films induced by confining surfaces^{19,20} and how these are related to (2) periodic and chaotic stick–slip motion^{21–23} (Fig. 2a, b), where the shape of the stick–slip can be sawtooth or oscillatory²⁴, (3) transitions between 'smooth' and stick–slip sliding at certain critical sliding velocities or loads^{24,25} (Fig. 2c), (4) the very high effective viscosities of confined liquid films²⁶, and (5) the funicity, that is, dependence on the previous history, of friction forces^{21,27}.

Atomistic MD simulations (Fig. 1) have a wide range of applicability and have reached a high level of rigour and accuracy. They help us to understand liquid layering in nano-confinement²⁸, the relationship between static and kinetic friction^{6,10}, the nature of transitions between stick–slip and smooth sliding⁴, slippage at solid–liquid interfaces^{4,29,30}, shear thinning²⁹ and the friction of rough surfaces⁹. But MD simulations are currently limited to timescales no greater than tens of nanoseconds and length scales of tens of nanometres, which are too short for analysing many tribological systems⁷.

An important issue, therefore, was how to reduce the large-scale, many-parameter MD simulations to simpler descriptions with only a few equations of motion. Various phenomenological RS models^{10–15} provided such a description (Fig. 1), where the coefficients of one or two dynamical equations are fitted to experiment variables and then used to describe a wide range of observed frictional behaviours, such as the dilation of a liquid under shear¹⁵ and the transition between stick–slip (regular or chaotic) and smooth sliding friction^{12,31}. However, most 'state variables' in RS models cannot yet be quantitatively related to physical system properties^{11,12}.

An additional understanding of friction came with the MM (Fig. 1) that focuses on a small number of the most relevant degrees of freedom of confined molecules but can nevertheless explain phenomena of high complexity^{16–18,32,33}. Moreover, the MM enabled predictions to be made that were later verified experimentally^{23,25}. The MM naturally led to two characteristic states of the embedded system when sheared in the presence of thermal noise: 'trapped' and 'sliding' states. These are the ingredients that lead to stick–slip and the transition to sliding and are therefore the essential requirements for successful modelling of friction³⁴. The MM emphasized the nonlinear nature of frictional dynamics that has led to a potentially new method for controlling friction and/or boundary slip via the external manipulations.

Control of friction

The ability to control and manipulate frictional forces is extremely important for many applications. One may wish to reduce or enhance friction, modify the chaotic regime, and so on. Such control can be technologically important for micromechanical devices and computer disk drives, where the early stages of motion and the stopping processes often exhibit unwanted stick-slip or damage³⁵. In contrast, chaotic stick-slip may be desirable, for example, in string instruments. The control of frictional forces has been traditionally approached by chemical means, usually by supplementing base lubricants with friction modifier additives.

A completely different approach for ‘tuning’ frictional response, which has attracted considerable interest recently^{8,36–40}, is to control the system mechanically via normal vibrations of small amplitude and energy (Fig. 3). In this case, the idea is to reduce the friction force or to eliminate stick-slip motion through a stabilization of desirable modes of motion. Figure 3a and b show some recent experimental results^{36,37}, and Fig. 3c and d show corresponding theoretical MM³⁸ and MD⁸ modelling of these systems. Calculations demonstrated that oscillations of the normal load could lead to a transition from a state of high-friction stick-slip dynamics to a low-friction smooth sliding state. Manipulation by mechanical excitations, when applied at the right frequency, amplitude and direction, pull the molecules out of their potential energy minima

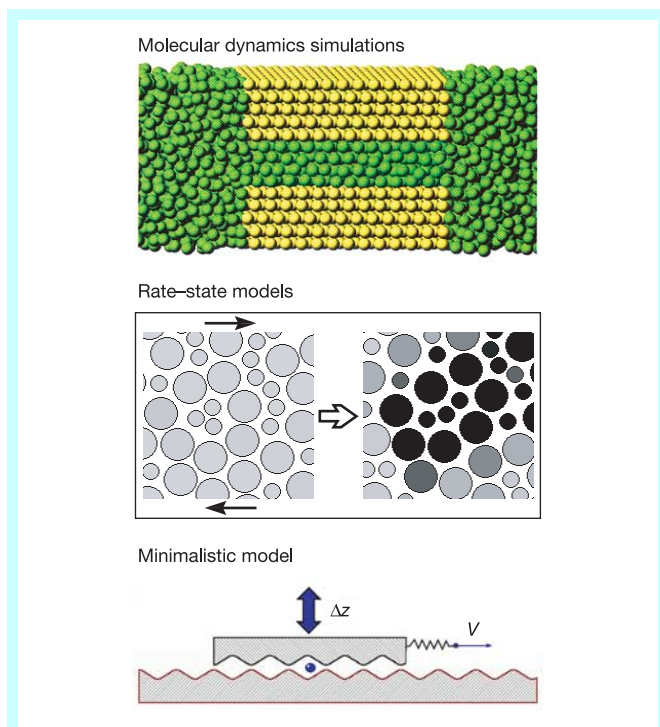


Figure 1 Three theoretical approaches to model friction. These are molecular dynamics simulations^{3–9}, rate-state models^{10–15}, and a ‘minimalistic’ model^{16–17,32,33}. MD simulations⁹, here showing shear-ordered lubricant molecules (green) between gold surfaces, follow the trajectory of molecules in space and time by solving the equations of motion as determined by the interatomic potential functions. RS models assume that the system is composed of ‘local phases’ (domains or grains) that can be described by a small number of ‘state variables’ that characterize the deformations, molecular rearrangements^{10,12}, dilation¹⁵ and other (statistical) properties of the interfacial material. Darker grains indicate a larger shear displacement with time. MM reduces the system to its bare essentials, for example, representing an embedded system by a single particle (which can also describe a system of many non-interacting particles) between two surfaces. Most of the experimental observations are qualitatively recovered by the MM.

(and thereby reduce friction) (at other frequencies or amplitudes the friction can be increased).

Friction and lubrication in biology

Animals, insects, their internal organs, tissues and biological microstructures and microorganisms experience much the same friction and lubrication forces in their movement as do machines. These can involve both lubricated (Fig. 4a) and unlubricated surfaces (Fig. 4b). The main difference between man-made and natural (biological) lubricants is that the former are usually ‘oil-based’ while the latter are ‘water-based’. Water-based lubricant systems function well with hydrophilic surfaces where the surface charge provides an electrostatic ‘double-layer’ repulsion, in addition to the ‘steric’ repulsion of the hydration layer of tightly bound water molecules⁴¹. Recent experiments⁴² show that brushes of charged polymers (polyelectrolytes) attached to surfaces rubbing across an aqueous medium result in superior lubrication even at low sliding velocities and at pressures up to several atmospheres. Some biolubricating systems, such as in the eyes, may be similarly mediated by brush-like polyelectrolyte layers.

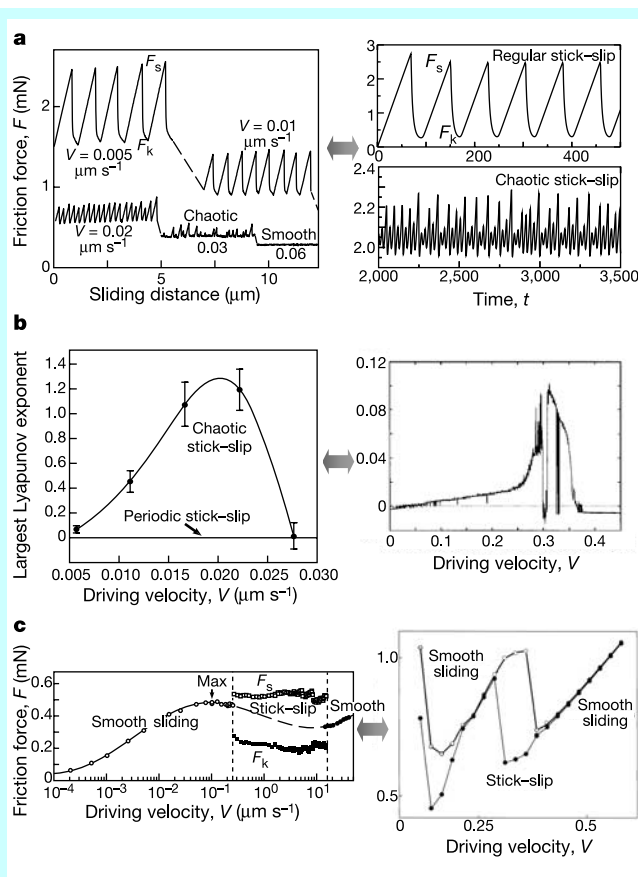


Figure 2 Examples of complex tribological effects of friction forces. Such effects versus sliding distance or time are shown as measured (left panels) and modelled (right panels) in dimensionless units. **a**, Friction traces of ultra-thin films of the model lubrication-oil squalane ($C_{30}H_{62}$) between two shearing mica surfaces showing a typical transition from periodic stick-slip to smooth sliding via a chaotic stick-slip regime, as measured²³ and modelled by the RS model³¹. F_s and F_k are the static and kinetic friction forces in the stick-slip regime. **b**, Positive Lyapunov exponent—an indicator of ‘chaos’ as opposed to random or ‘stochastic’ motion—as measured²³ and modelled by the MM model^{16,17}. **c**, Example of measured²⁵ and MM modelling³² of a transition from smooth to (inverted) stick-slip sliding and to smooth sliding again with increasing velocity. Open and closed symbols show the maximum and minimum friction forces in the inverted stick-slip regime. An RS model has also successfully explained this phenomenon²⁵.

Water-based lubricants are more efficient but cannot function at high temperatures owing to the high volatility and oxidative reactivity of water. But with the development of MEMS devices that are often made of ceramic materials and designed to operate at ambient temperatures, it is likely that we will learn from biology how to use water-based fluid lubricants. Another important differ-

ence between biological and man-made lubrication systems is that in the former the lubricant is often chemically attached to the surface, as occurs at the cartilage surfaces of joints. The recent development of computer disk surfaces, using 2–4-nm thick perfluoroether polymer layers chemically grafted to the carbon surfaces, is an example of this trend.

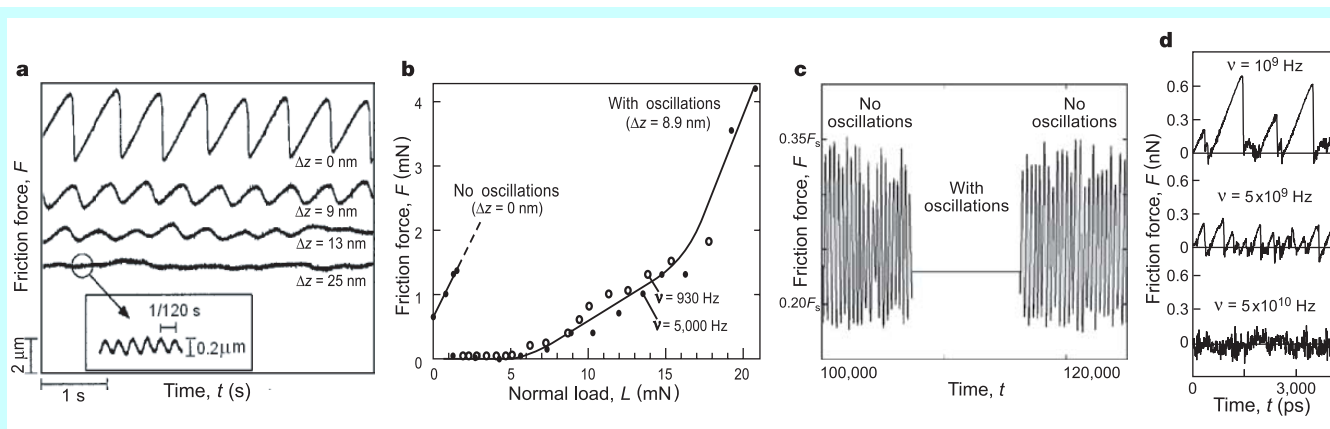


Figure 3 Reduction of friction and stick-slip by mechanical excitations. **a, b**, Experiments^{36,37} showing a drastic reduction in the stick-slip amplitude ($F_s - F_k$), the friction force F , or the friction coefficient $\mu = F/L$, by applying low-energy oscillations of frequency ν and small amplitude Δz perpendicular to the sliding direction. **c**, MM modelling³⁸ shows the elimination of stick-slip during the oscillations.

The friction force is given in units of static friction; time in dimensionless units. **d**, An MD simulation of F versus time at three different frequencies for a system of two organic surfactant-coated surfaces as in **b** showing the reduction of stick-slip and F with increasing ν .

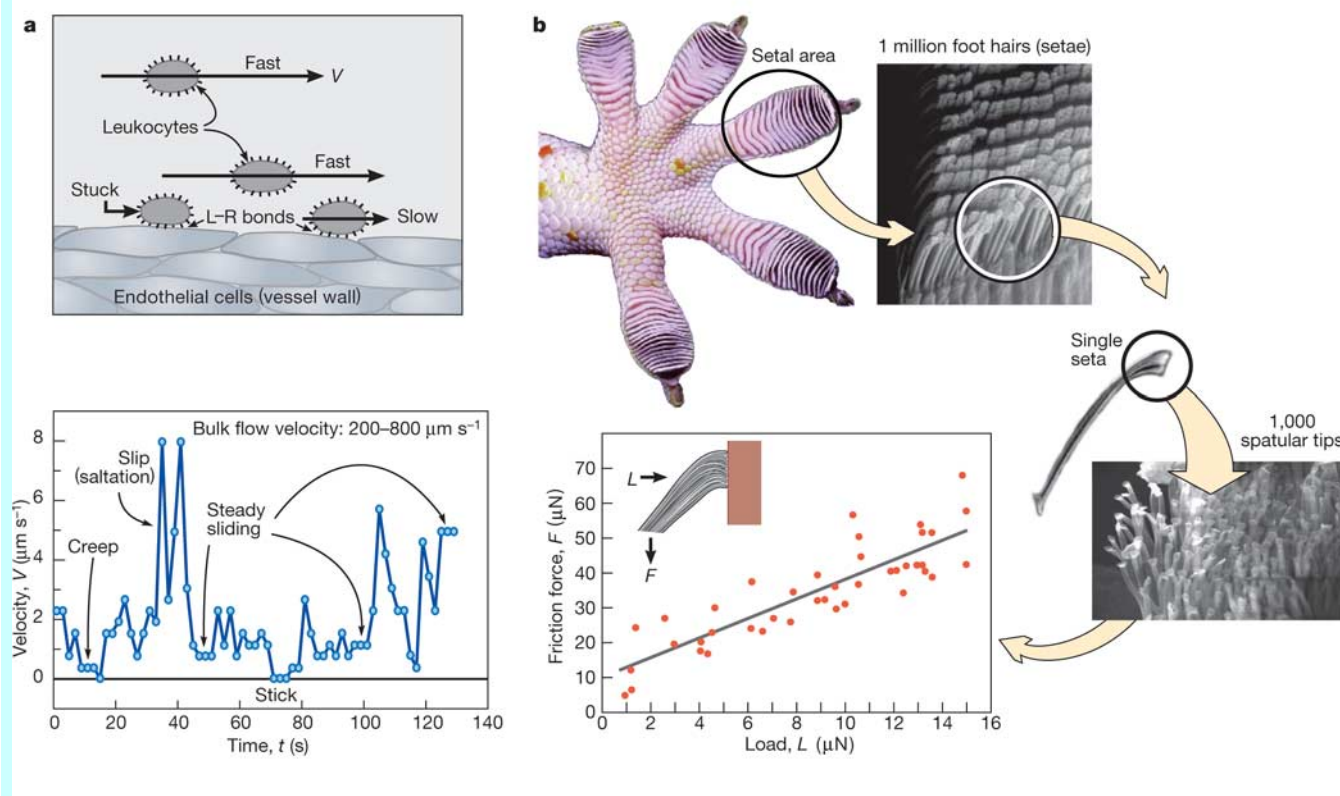


Figure 4 Two examples of friction and lubrication forces in living systems. **a**, *In vitro* 'rolling' of leukocyte cells while in contact with the endothelium (top) resembles chaotic stick-slip behaviour (bottom⁴²) involving sticking ($V = 0$), slow sliding (referred to as 'rolling' or creep), steady sliding, slip or 'saltation', and free or bulk flow ($V = 200$ –

$800 \mu\text{m s}^{-1}$). **b**, The high adhesion and friction forces between the pads on gecko feet and surfaces allow them to climb up walls. The truly amazing aspect of gecko locomotion is their ability to both attach and detach, that is, control high binding and unbinding forces, within a few milliseconds. Figures kindly supplied by K. Autumn.

Theoretically, the differences between water-based and oil-based systems are probably more quantitative than qualitative, and there is no reason to believe that existing nonlinear dynamical models which apply to machines and nano-devices are not also applicable to biological systems, such as those shown in Fig. 4. Thus, the saltatory motion of white blood cells along the endothelium surfaces of blood capillaries (Fig. 4a, top) follows a chaotic-like stick-slip motion (Fig. 4a, bottom). This behaviour has been successfully modelled in terms of the thermally controlled binding and unbinding of specific ligand-receptor bonds on the two surfaces^{43,44}.

Figure 4 shows examples of how adhesion and friction are intimately coupled in complex macromolecular systems. This coupling is also found in other biological systems, such as protein unfolding, which often follows a stick-slip process⁴⁵. The close relationship between adhesion, stick-slip and friction, which ultimately involve the making and breaking of bonds, is at the heart of recent theoretical and experimental studies^{46–50} that suggested a universal $[\ln V]^{2/3}$ - dependence of the height of stick-slip spikes on the driving velocity V .

Outstanding fundamental questions

What one hopes for is a unified approach to energy-dissipating systems that encompasses most tribological but also other phenomena, for example, in biology and geology. The models should be able to explain complex but common observations in terms of meaningful physical quantities and unravel the origin of energy dissipation which underlies all friction processes. Later, one would like to use these models for making predictions. More specifically, some of the important questions are:

- (1) Why is 'static friction' so universally observed between solid objects?
- (2) How are friction and wear related? And why does surface damage often occur at the start of motion?
- (3) How are the static and kinetic friction forces, and the characteristic transition velocities between smooth and stick-slip sliding, determined by the molecule-molecule and molecule-surface interactions and, in macroscopic systems, asperity-asperity or grain-grain interactions?
- (4) Are the stick and slip regimes indicative of different phase states (liquid, solid, glassy) of the confined films or interfaces?
- (5) What 'hidden' information is contained in chaotic as opposed to periodic motion (compare Fig. 2)? This is particularly important for predicting earthquakes.
- (6) And finally, how can we control friction in practice, most often to reduce it or eliminate stick-slip at all pressures and velocities? But there are also situations when one wants high friction, as in clutches and brakes, or stick-slip, to enrich the sound of a violin and improve the feel or 'texture' of processed food as sensed during biting and chewing. □

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