Frictional forces and Amontons' Laws: from the molecular to the macroscopic Scales, including different time and velocity scales, as studied by SFA, AFM, etc.

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There are a number of fundamental (but essentially empirical) laws of friction or tribology that were laid down many years ago by Leonardo da Vinchi, Amontons, Coulomb, Stribeck, and others that are still commonly used to describe the results of tribological phenomena and experiments, either qualitatively and quantitatively. The recent advent of sophisticated experiments such as the Surface Forces Apparatus (SFA) and AFM, and computer simulations, that can now directly probe tribological phenomena at the atomic (sub-nano) scale, as well as draw conclusions about scaling effects of length (area), load (pressure), time, and rate (sliding or shearing velocity), is leading to a reassessment of all of these 'fundamental laws', especially their scaling when taken over many orders of magnitudes. Nevertheless, Amontons' laws (and the Stribeck Curve) ***are*** often found to accurately describe experimental results (see, for example Fig. 1), although in most cases this is not due to the reasons that form the conceptual basis of these laws.

The early laws of friction are also surprisingly silent about some highly important tribological phenomena, including stiction, stick-slip friction, the sliding time, and 'previous-history' or memory effects, which are now recognized as being central to earthquakes and other seemingly unrelated phenomena (such as sensory perception, the sound of a violin, food texture) that now appear have a common tribological origin (see Fig. 2).

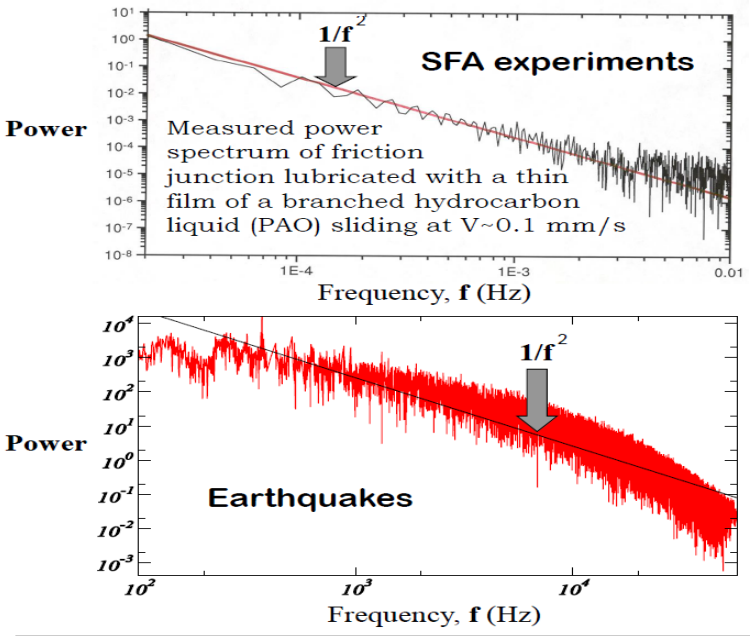


Figure 2. Similarity in typical experimental SFA measurements of chaotic friction and actual earthquake power spectra.

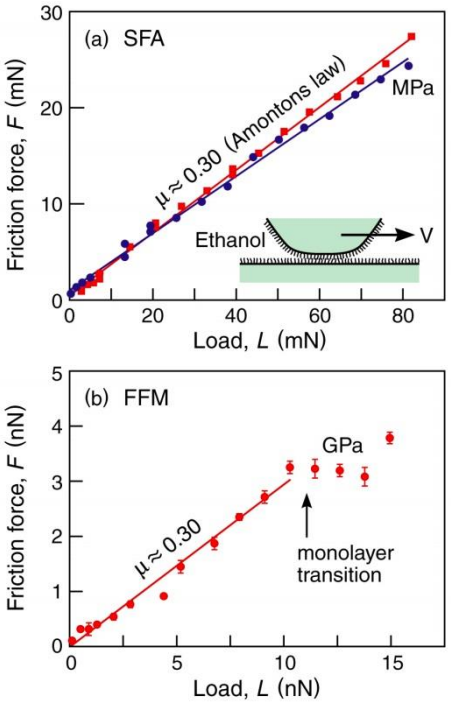


Figure. 1. Identical friction coefficients measured by SFA and AFM (FFM) for two surfactant monolayer-coated surfaces with up to six orderes of magnitude differences in the loads, pressures, and contact areas.

These studies have also revealed interesting effects such as how anisotropically structured (commensurate or incommensurate) surfaces and linear lubricant molecules or anisotropic nanoparticles become aligned or reoriented druring sliding, and the profound and subtle effects this can have on their (unisotropic) friction, and the relationship between adhesion and stick-slip-friction and wear-initiation (Fig. 3).

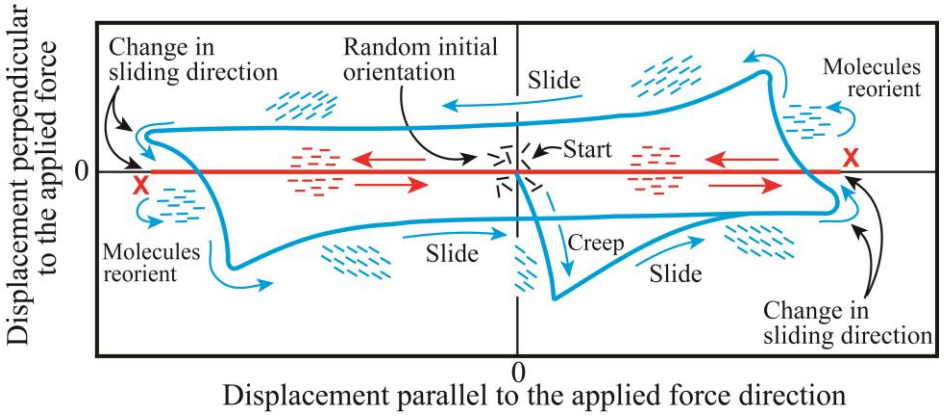


Figure 3. Using a modified '3D Surface Forces Apparatus' that can apply and measure forces and motion in all three directions in space, we measured the friction forces generated by nano-thin liquid films of linear hydrocarbon chain molecules (hexadecane) between two mica surfaces whose crystalline surface lattices could be oriented at different 'twist angles', and where the moving surface was allowed to move in any direction in space. The anisotropic lubricant molecules and anisotropic surface lattices (at the atomic or sub-nano-scale) resulted in 'anisotropic, off-axis, friction forces' and a complex path taken by the surfaces (see blue curve) manifested by a non-zero velocity at all stages of back-and-forth sliding, and absence of damage even after prolonged shearing. The red line and dashes show the conventional straight line path and the orientation of the trapped lubricating molecules during conventional reciprocating (back and forth) motion where the surfaces come to rest at points **X**. The blue curve and dashes show the complex anisotropic 'off-axis' path and molecular orientations of this system, where the anisotropic molecules and surfaces never stop moving, and thereby avoid stiction damage to the surfaces.

This talk will provide concrete examples of how experiments and modeling over the last 30 years into all of the above phenomena have provided completely new insights and interpretations of these phenomena, both quantitatively and qualitatively, and have given the field of tribology a more physically rigorous, rather than empirical, basis.

Friction from Atomic to Nano Scales: *In-Situ* Observation in TEM

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In the study on the mechanical properties of nano structures, experiment with in-situ observation is important because the properties depend tightly on the dynamic change in their shapes and sizes [1]. Thus, in-situ observation was performed by a transmission electron microscope (TEM) with a help of small actuators, such as a MEMS actuator [2]. In our experimental system, named MEMS-in-TEM, MEMS devices with micro electrostatic actuators were operated inside an ultra-high-vacuum TEM specimen chamber, $5x10^{-8}$ Pa. The precision of the micro electrostatic actuator was better than sub-nm order and its stability was 0.1 nm in standard deviation for several tens of minutes. The MEMS device was inserted into a TEM specimen chamber using a custom TEM sample holder. The driving voltages for actuators were applied using feed-throughs equipped in the holder. The spatial resolution of TEM observation was 0.2 nm. The images were recorded as a video at 30 fps.

We have conducted real-time in situ imaging of nanometer-scale asperity-pair friction at high normal pressure (> 1GPa) during low-speed sliding (~0.01 nm s^{-1}) [3] – a situation in close analogy with plate asperities. A pair of nanometer-scale crystalline silicon apexes were compressed and rubbed with TME monitoring of its evolution. This operation induces decrystallization, atomic inflow, and plastic deformation at the junction (initial diameter: 6 nm) between the apexes. Our further analysis using molecular dynamics (MD) simulations showed that this dynamical process helps gradual (continuous) dissipation of a large portion of the energy accumulated at the interface and hence reduces the chance of sudden (discrete) energy release or catastrophic destruction.

We coated the apexes with Ag and conducted the same experiment [4]. Stepwise discrete deformations were observed for an Ag nano-junction under shear stress; this is similar to the observation of stick–slip motion at the microscale. The length of discrete deformation were the integral multiple of 0.3 nm. The theoretical value of a sliding distance due to the atomic structure of Ag is estimated to be 0.29 nm [5], which is the distance between neighboring atoms on a slip plane of Ag. Furthermore, the shear force was calculated as the product of the stiffness of the arm (2.1 N m^{-1}) and the difference between the displacement before and after junction formation. We demonstrated that the energy loss associated with a shear fracture event is strongly correlated with the increase in total surface energy of the two surfaces formed here after the fracture.

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Scaling of Friction With Contact Area in Single Asperity Contacts

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Models of friction commonly assume a simple relation between friction forces and the load or area of contact. Adhesive models assume friction proportional to area and nonadhesive models often assume area proportional to load. Previous simulations have observed both types of behavior but have typically been limited to contacts that are at most a dozen atoms across. We use an efficient Greens function method to study the friction in contacts with radii ranging from a nanometer to a micrometer with a range of interaction strengths and loads. Commensurate, incommensurate and amorphous surfaces are studied. We identify three regimes of scaling. Small contacts are effectively rigid. In this limit commensurate contacts have a friction proportional to area or load, while the friction in incommensurate or amorphous contacts rises as a lower power of area or load. At large loads the friction becomes independent of area or load and can be related to the Peierls stress for dislocation motion along the interface. The crossover regime between the two regimes depends on the nature of interactions and the geometry. Scaling exponents are found for commensurate surfaces in the limits of nonadhesive and adhesive interactions. The latter case is consistent with previous work by Hurtado and Kim and Gao. A new exponent is derived for repulsive interactions. The connection to the Mindlin Model for single asperities and simulations of multiasperity contacts will be discussed.

What does the coefficient of friction depend really on? – (Some Philosophical Thoughts about the Past and Future of the Physics of Friction)

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It is known that the coefficient of friction generally depends on a large number of system and loading parameters. Alreagy Coulomb presented experimental evidence that the static coefficent of friction may depend on time, on normal force, on the contact size, and on the presence of intermediate lubricant layers. For sliding coefficient of friction, he presented dependencies on the sliding velocity as well as force and size dependencies [1],[2]. Later research has shown that the friction coefficient is very sensible to the presence of oscillations (including self-excited vibrations). Due to the large number of governing parameters, no generalized laws of friction or empirical procedures for measuring and representing the law of friction have been developed so far, which included at least *each* of the following four parameters: velocity, force, system size and form, and time.

In the present paper, we discuss the question of how the dimension of space of governing parameters can be reduced and if a small set of "robust giverning parameters" of friction can be identified. We argue that the property which most directly and robustly determines the contact configuration is the *indentation depth*, whereas the parameters of the contact configuration which are relevant for friction are the *surface gradient* and the characteristic *size of micro-contacts*. Both parameters depend only on the indentation depth, though this dependence is relatively weak. Another parameter which is of importance is the characteristic *relaxation time* of the medium which only depends on temperature. The suggested structure of the coefficient of friction allows to apply the phenomenological "generalized master curve procedure" by measuring the coefficient of friction at different velocities, temperatures and indentation depths and applying the shifting procedure [3]. This master curve procedure is based on the following property of friction: if we plot the logarithm of the coefficient of friction as function of the logarithm of velocity, for different values of the indentation depth and the temperature, all curves will "look the same" and it is possible to move them by some vertical shift factor depending solely on the indentation depth as well as horizontally by the shift factor which is the additive superposition of a function of indentation depth and a function of temperature.

The indentation depth determines most directly not only the coefficient of sliding friction but also the *pre-sliding* (initial displacement before the gross sliding starts) and is the natural scale also for the *influence of oscillations on friction*.

As the indentation depth is not a parameter which normally can be controlled in experiment, the suggested frictional law has to be completed by a contact mechanical derivation connecting the indentation depth with the normal force, shape of the bodies and their materials parameters.

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Theory of Amontons' Law of Friction

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It is widely believed that Amontons' law, that is the frictional force is proportional to the loading force and does not depend on the apparent contact area, holds well for dry sliding systems. But the mechanism of the law is still in controversy. Bowden and Tabor established the adhesion theory for the occurrence of the friction [1]. In the adhesion theory the Amonrtons' law holds because the actual contact area is proportional to the loading force, e.g. by the yielding of actual contact points, and the frictional force is proportional to the actual contact area. Greenwood and Williamson showed that the law is derived by considering the height distribution of asperities even for the elastic systems also by assuming that the frictional force is proportional to the actual contact area [2]. But it is now well known that the frictional force is not proportional to the actual contact area for the incommensurate or random systems as claimed by Robbins et.al. [3].

Here I extend the Greenwood-Williamson theory and consider explicitly the contribution of each asperity to the maximum static frictional force of the system. I successfully derive the Amontons' law for the maximum static frictional force without assuming that the friction force is proportional to the actual contact area. I further extended the theory to include the characters of each asperities, such as shape, dimension, the degree of plasticity and so on. I also take into account the effect of the finite range attractive interaction between asperities. Moreover the effects of correlation among the deformations of asperities are also taken into account. Even in those cases the Amontons' law holds for the systems in which the actual contact area is small enough compared to the apparent contact area and the conditions noted below are satisfied.

In the above argument we assumed the loading and driving conditions are uniform for each asperities. This is a reasonable assumption for the mesoscopic systems. In many macroscopic systems, however, the nonuniformity of the pressure at the sliding surface resulting from the torque induced by the driving force can play an important role. The nonuniformity induces the local precursor slips before the onset of bulk sliding. The precursor can make Amontons' law break and instead new friction law can hold [4]. The static friction coefficient decreases with the increasing loading force with the power -1/3. This behavior is confirmed by the experiment [5]. Law of friction depends on the scale of the system.

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Analytical Calculation of Macroscopic Friction Coefficient for Viscoelastic Materials

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In many sliding systems consisting of solid object on a solid substrate under dry condition, the friction force does not depend on the apparent contact area and is proportional to the loading force. This behaviour is called Amontons' law and indicates that the friction coefficient, or the ratio of the friction force to the loading force, is constant. Here, however, from analytical calculations of a simple model, we show that Amontons' law breaks down systematically under certain conditions for an viscoelastic materials experiencing a friction force that locally obeys Amontons' law. The macroscopic static friction coefficient, which corresponds to the onset of bulk sliding of the object, decreases as pressure or system length increases. The results depends on the type of the viscoelasticity. This decrease results from precursor slips before the onset of bulk sliding, and is consistent with the results of our recent experiment and a numerical analysis [1,2].

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Nanoscale friction and wear phenomena studied by combining in-situ tribometry and atomistic simulations

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Sliding induces severe changes of the material with respect to topography, composition and microstructure by mechanical mixing of the tribocouples with the lubricants and additives ("third body" , [1]). This third body strongly influences the friction and the wear performance of sliding components yet the generation and the materials properties are barely understood. In our recent work we combined in-situ tribometry with atomistic simulations in order to improve our understanding of nanoscale interfacial processes in metallic tribo couples and coatings. Experiments were performed using a novel experimental platform for the on-line correlation of friction, wear and topography under lubricated sliding [1] and by AFM nanoscratching. The third body was characterized ex-situ by Focused Ion Beam (FIB) analysis, Transmission Electron Microscopy (TEM), X-Ray Photo Electron Spectroscopy (XPS) Nanoindentation and Micropillar compression. Then, in order to elucidate the atomistic level processes which contribute to the observed microstructural evolution in the experiments, classical molecular dynamics were carried out. The combined experimental and simulation data allowed a look at the third body formation of tungsten and tungsten carbide [3,4], diamond-like carbon coatings [5] as well as wear particle formation due to folding in copper [6].

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Title: The tribological triplet - a framework accross the scales.

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Abstract: In the 80 ‘s, Godet and Berthier build several powerful tribological concept to face each tribological problem. Around the concept of the third-body, a.k.a the interface which separate the body in contact, they build the concept of third body flows and the tribological triplet.

This last one proposes to see a «contact» as the combination of the two bodies which interact (the first and the second body, called the first bodies), the third body and the mechanism which maintain them.

Even if this representation seems natural, no real used have be done of such powerful tools.

Through three examples (space tribology, braking and geophysics), one proposed to deal with the concept of tribological triplet in situation where the characteristic lengh ranges from nanometer to meter, combining experimental and numerical approaches.

Neutron Scattering: Useful Method to Understand Molecular Level Origin of Friction and Lubrication

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Neutrons are electrically neutral and have the ability to deeply penetrate matter, and they can probe structural properties of a buried interface. They interact with nuclei and neutrons, unlike X-rays, can distinguish light elements such as hydrogen. Neutron wavelengths range from 0.1 Å to 1000 Å, which is comparable to the distance of neighboring atoms in condensed matter. This makes them an ideal probe of atomic and molecular structures from the order of angstrom to sub-micrometer scale. The energies of neutrons are of the same magnitude as the diffusive motion in solids and liquids, the coherent waves in crystals (phonons and magnons), and the vibrational modes in molecules. It is useful to detect any dynamical properties of matters between 1 neV and 1eV, corresponding to femto-sec and micro-sec.

Because of these characteristic features, neutron scattering could be useful to investigate tribology. Neutron Reflectometry has already been used for such works, for example, to investigate thickness and density of adsorbed layer on metal surface in lubricant. [1] Small-Angle Neutron Scattering has been intensively used to investigate a relation between structural properties and rheological behaviors. [2, 3] Quasi-elastic Neutron Scattering could give information about motion of molecules and molecular assemblies which should be an origin of internal friction of tyre rubbers. [4] By using these neutron scattering methods, rich information will be obtained on friction and lubrication.

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Resonance Shear Measurement for Tribology

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There are increasing interests in clear understanding of friction and lubrication behavior at the atomic and molecular scale. The physics and chemistry to explain the fundamental origin of friction and lubrication is not sufficiently established because of the complexity of the friction behavior. We developed the resonance shear measurement (RSM)[1,2], which employs the surface forces apparatus, for studying a thin liquid film confined between solid surfaces with controlling the thickness of the liquid at a sub-nanometer resolution. The frequency and the amplitude of the resonance peak are highly sensitive to the viscosity and the frictional properties of the confined liquids. The method has been modified for studying also the friction of gel[3].

This paper reports our RSM studies include: (1) development of the measurement techniques such as Fourier transform measurement for quick acquisition of resonance curves[4], and a new mechanical model for full analysis of the resonance curve[5]; (2) typical examples of the RSM measurements on water[6,7]; (3) systematic study on phenylether lubricants[8] and ionic liquids[9,10]; (4) two component system of a model lubricant (liquid crystal,6CB) and a model additive (dye, Sudan Black) which exhibits the stick-slip transition under the shear[11].

The friction between an elastomer and a hard surface typically has two contributions; i.e., the interfacial and deformation components. The friction of viscoelastic hydrogel materials has been extensively studied between planar gel and planar substrate surfaces from the viewpoint of an interfacial interaction. However, the geometry of the contact in practical applications is much more complex. The geometric and elastic deformation term of a gel to friction could not be neglected. We used the resonance shear measurements (RSM) for characterizing the shear response of a glass sphere on a flat polymer hydrogel, a double network (DN) gel of 2-acrylamide-2-methylpropanesulfonic acid and *N,N*-dimethylacrylamide, and have found that the friction between a flat DN gel and a silica sphere was dominated by the elastic term due to the local deformation by contact with a silica sphere[3].

These studies indicate that more precise design of lubricants should be possible for controlling the friction.

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Glasslike Dynamics of Liquid Lubricants in Confined Geometries

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When liquid molecules are confined in a narrow gap between two solid surfaces, their dynamic properties are completely different from those of the bulk. The molecular motions are highly restricted and the system often exhibits solid-like responses when sheared slowly. The dynamic properties of a variety of confined liquid lubricants were investigated experimentally using the surface forces apparatus (SFA) technique. Simultaneous and precise measurements of friction force (both kinetic and static), sliding film thickness and real contact area using the SFA revealed the molecular-level mechanisms of the solidification dynamics. Two important experimental observations are described in this talk: i) universal shear-thinning behavior of confined liquid lubricants in their hard-wall states [1-4]; and ii) freezing-melting transitions of confined lubricants in stop-start dynamics [5-8]. Both observations are explained from the Williams-Landau-Ferry (WLF) theories of bulk glass-forming materials modified for confined liquid systems, indicative of a glasslike nature of the solidification dynamics.

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Rotation of Frictional Force in Translational Sliding

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Abstract

Fundamentals of friction have been examined for a long time in simple translational sliding. In most of a number of models on sliding friction, consciously or subconsciously, the two velocities (V_1 and V_2) of contacting surfaces are supposed to be in the same direction, which naturally makes researchers focus on the "magnitude" of frictional force. However, if the directions of V_1 and V_2 are different, even if the misalignment angle (φ) is quite small, frictional force rotates continuously with change in the magnitude of V_1 or V_2, which sheds light on the other side of the frictional force as a vector: the "direction" of the frictional force. In this presentation, I will show how the rotation of frictional force affects strongly on the dynamics of translational sliding.

