SCALE DEPENDENCE OF FRICTION

M. SCHERGE
IAVF Antriebstechnik AG, Im Schlehert 32, 76187 Karlsruhe, GERMANY; e-mail: matthias.scherge@iavf.de

SUMMARY
Our microtribological research has shown that basic friction mechanisms crucially depend on the length and force scale under investigation. The tests were performed with a microtribometer covering the entire force range from millinewtons to nanonewtons for the tribological pairing of a silicon ball with a diameter of 1 mm versus a silicon flat. The ball is attached to an especially adapted glass double leaf spring. The flat sample is connected to a sample holder that is oscillated. When both samples are engaged friction was measured during oscillation. To detect the friction-induced deflection of the spring a laser interferometer was used. The investigation shows that the overall friction signal is composed of several contributions acting on different length and force scales. Whereas in the range of millinewtons friction is dominated by topography, water film thickness determines the magnitude of friction in the micronewton range. When the dimensions are reduced further ordering effects become detectable that result in increased viscosity of the liquid. When the liquid is removed, mutual attraction due to cohesion is the main contribution to friction in the nanonewton range. All contributions act simultaneously, however, each mechanism is superimposed by the mechanism acting on the following higher force scale.

Keywords: microtribology, friction, capillarity, ordering, scale effects

1 INTRODUCTION
This paper deals with the evaluation of friction in a micromechanical system. Since the beginnings of micro- and nanotribology the question was raised if these new methods – atomic force microscopy or microtribometry – can be used to simulate macroscale friction systems [1]. The approach is tempting, since the macrocontact can be considered as the sum of a vast number of microcontacts. Thus, the apex of the AFM or the micrometer-sized probe of the microtribometer can be used to simulate a single-asperity contact [2 - 4]. Very soon it was realized that the friction coefficient obtained with these methods has little to do with the coefficient obtained in macrosystems. One of the reasons for the discrepancies is lack of statistics. This means that a single-asperity contact is just a single contact isolated from all neighboring contacts and all forms of mutual interaction. The other reason is the insufficient comprehension of mechanisms that contribute to the overall picture of friction. It can be shown that different scale levels with respect to length, height, width and force are accompanied by distinct friction mechanisms. By separating these mechanisms their range of action can be evaluated.

To cover the force range from mN to nN a microtribometer was used [5]. The results were obtained for a model system consisting of a silicon ball and a silicon flat [6]. The system was investigated in an unlubricated state. However, due to adsorption a liquid film that mainly consists of water covers the surfaces [7]. Since the samples were measured in air, silicon is oxidized and, therefore, hydrophilic. The following section describes the microtribometer and the samples. In section 3 results of friction measurements on the milli-, micro- and nanonewton scale will be presented. Section 4 summarizes the results and provides conclusions.

2 MECHANICAL SETUP
In this section a microtribometer is described that optically detects the deflection of a double-leaf spring allowing the measurement of forces in the range from mN to nN for a sample size that can range from micrometers to several millimeters [8]. The following technical requirements are met by the microtribometer:

i) The forces are measured directly by detecting the deflection of the spring with nanometer resolution.

ii) The stiffness of the employed bending beam is exactly known both for vertical (normal force) and horizontal direction (tangential force).

iii) The force measurement has a resolution of nanonewtons covering a range from nN to mN.

2.1 Spring selection
Photo-structurable glass was used as the base material for high-precision bending beams. Glass was chosen because of its supreme mechanical properties with respect to the elastic constants [9]. By means of photolithography 1 mm thick glass wafers (Li2O—Al2O3—SiO2) were structured [10]. When the glass is exposed to ultraviolet radiation, the amorphous structure becomes crystalline. Using buffered hydrofluoric acid the exposed areas were removed after annealing. This technique ensures a high aspect ratio. Two 100 µm wide and 1 cm long glass beams serve as displacement elements. The etching process is controlled to preset the spring constant by realizing the appropriate ratio of length and thickness of the bending beams. Mechanical stressing of the springs showed no degradation of the elastic properties, even after $10^8$ deflections over a distance of 400 µm at 5 Hz. Fig. 1 shows the double-leaf spring. Based on construction, the spring is stiffer in the z-direction ($k_z = 100$ N/m). The stiffness in the x-direction ($k_x$) is 50 N/m.
2.2 Mechanical and optical set-up

A polished silicon ball with a diameter of 1 mm was used to probe the flat counter sample. Since ball and flat are made of the same material, identical surface preparations were performed. This ensured comparable test results. In the experiments the ball was glued to the spring as shown in Fig. 1.

![Diagram of tester setup](image)

Fig. 1: Tester setup. The silicon ball is attached to the spring. The flat silicon sample is oscillated in horizontal direction to initiate friction and in vertical direction to set the normal force. The piezo-electric transducers necessary to move the sample holder are not shown. By means of a single-beam laser interferometer the deflection of the spring is determined with nanometer resolution. By multiplication of deflection and spring constant the force values were obtained. With a spring constant in tangential direction of 100 N/m a force resolution of 100 nN was achieved.

The mechanical part of the tester consists of a piezo-electric transducer (x-piezo) for dynamic application up to 400 Hz with a maximum expansion of 35 µm. The piezo-electric transducer is powered by a low-voltage amplifier (-10 V – 150 V), which is controlled by an input voltage-ramp to achieve a linear expansion, retraction and constant sliding velocity. A second piezo-electric transducer (z-piezo) is used for engagement and serves to set the normal force. After the contact is established further expansion of the z-piezo deflects the spring in the vertical direction. The spring is stiffer in this direction and higher normal forces (compared with the tangential force) can be set. Micropositioners are used first to make the ball approach towards the flat sample. The final engagement is accomplished by using the z-piezo.

To detect the deflection of the spring with nanometer resolution an interferometer was applied. The ideal solution for microtribological applications consists of a single-beam interferometer and a spring with attached reflector (see Fig. 1). The set-up involves a circular aperture to evaluate the passing interference pattern at just one point. The beam is then sent through a beam splitter to obtain two sub-beams for sine and cosine signal generation. First, both sub-beams are equal in phase. Therefore, the 90° phase shift has to be introduced by polarization. Out of a variety of possible methods the superposition of orthogonal circular polarized light was chosen, leading to a phase-stable sine and cosine signal independent of the shape of the interferogram. Since the interferogram is evaluated at just one point, a laser beam of small diameter can be used and small reflecting areas are sufficient to serve as a mirror.

Since all mechanical parts are compliant with vacuum requirements, the tester can be placed in a UHV chamber with an ultimate pressure in the 10⁻⁹ mbar range. The deflection of the spring is measured by passing the laser beam through a glass window onto the reflector.

To measure friction, one single-length signal was measured. This was the deflection of the spring. Due to friction the ball and flat initially stick until the back driving force \( F_s = -k_x \Delta x \) generated by the spring equals the friction force. Up to this point the relative velocity is zero. After exceeding the static friction force \( F_s \) the ball starts to slide. When the direction of the horizontal drive is reversed, ball and flat sample stick again until the spring force equals the friction force. The peak-to-peak value is equal to twice the static friction force \( F_s \). The force in the vertical direction can be determined by taking the deflection of the spring in the z-direction.

2.3 Sample preparation

Silicon (100) was used in all experiments. The roughness was about 1 nm for the polished sample and 1200 nm for the rough samples. Before the tests the samples were rinsed in isopropanol and methanol for 5 minutes followed by a dip in distilled water. The thin native oxide layer (thickness 1.2 nm), which is always present, is receptive for water. Auger Electron Spectroscopy (AES) was used to monitor the chemistry of the surface. AES was used as a measure of the oxide coverage, thus being an indirect indication for the degree of hydrophilicity. From the spectra it was deduced that the oxygen intensity of the hydrophilic (oxidized) sample is about 3 times larger than of a hydrophobic (oxide free) reference sample. A small silicon peak in the spectrum of hydrophilic silicon is characteristic for the replacement of silicon atoms by oxygen. The Auger line shape was used to confirm that silicon was oxidized. This can be shown in the LMM transition at about 90 eV. The shift to lower energies (from 92 eV for silicon to 78 eV for oxidized silicon) and the line shape are typical for oxidized silicon.

The same procedure was applied to the ball. The ball has a diameter of 1 mm and received the same surface finish as regular silicon used in integrated chip manufacturing. To prevent any effect caused by faceting, a ball made of polycrystalline silicon was chosen.
3 RESULTS

In this section results of friction experiments on different length scales are presented. The analysis starts from rough surfaces and proceeds to smooth surfaces where subtle influences like ordering and cohesion were detected.

3.1 Roughness-induced friction

It was shown previously [7] that capillarity has the strongest impact on friction in the micronewton range. Capillarity acts in the same direction as the normal force, thus, increasing the load.

The strength of the capillary bridge is determined by the amount of adsorbed water as well as by the curvature of the surface [11]. Therefore, roughness strongly influences friction mediated by a liquid. As a consequence, frictional fluctuations can be correlated to the irregularities of topography. Fig. 2 shows an example. Up to 1000 nm of sliding distance ball and flat stick followed by stochastic fluctuations of the friction signal. The fluctuations are caused by the formation and interruption of capillary bridges expressed in a changing normal force. With increasing roughness the curvature of the asperities decreases. This leads to a decrease of capillary force according to

\[ F_c = 4\pi\gamma R, \]

where \( \gamma \) is the surface energy and \( R \) is the radius of curvature. Therefore, with increasing roughness friction decreases. Roughness is the controlling factor.

When the rough surface is replaced by the smooth surface, roughness effects can be neglected. Therefore, the action of capillarity is confined to the influence of the liquid film thickness. This can be underlined by measuring friction as function of the relative humidity. Fig. 3 shows this effect. For low humidity friction is small. A strong increase can be induced by increasing the water coverage of the sample. This effect is supported by capillary condensation in the slit that is formed between ball and flat. When the sample has achieved equilibrium between adsorption and desorption, friction keeps constant. The formation of the equilibrium is not an instantaneous process as underlined by the transition between 50 % and 60 %.

3.2 Molecular order-induced friction

Chemical surface modification (oxidation or hydrogen termination) and variation of the environmental conditions are means to alter the thickness of the water film on silicon samples. However, the quantitative determination of the water film thickness is rather complicated. The following experiments quantitatively investigate the interactions of the hydrophilic silicon samples to determine the dependence of friction force on the water film thickness in vacuum [12]. In these experiments, the fact was used that water desorbs as the pressure drops, finally forming a single monolayer or less. The friction force was measured initially in air and subsequently as a function of the total pressure in the vacuum system.

Parallel to the friction measurement the thickness of the water film was measured by scanning tunnelling microscopy (STM) as explained in [13]. With decreasing pressure the water film decreases in thickness. The measured water film thickness is the sum of the water film on the Si sample and on the tungsten STM tip, which is also oxide-covered. At a pressure below 10\(^{-7}\) mbar the last monolayer of water desorbed. This was confirmed by photoelectron spectroscopy, which indicated a binding energy of oxygen caused by the bond with silicon. In the case of water coverage the oxygen peak would have been shifted to the value caused by the bond with hydrogen. This was confirmed byphotoelectron spectroscopy, which indicated a binding energy of oxygen caused by the bond with silicon. In the case of water coverage the oxygen peak would have been shifted to the value caused by the bond with hydrogen. 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This current is caused by ionization. When the tip is in close proximity with the solid surface the tunnelling effect increases the current. The distance between the onset of the ionization current and the exponentially increasing tunnelling current was interpreted as water film thickness, see Fig. 5. The offset in water film thickness due to tip coverage was eliminated by a thickness measurement versus a water-free hydrophobic reference sample. The obtained values are too large due to the effect of high tip voltages.

As the pressure dropped, the equilibrium point between liquid and vapor constantly changed. Lower water film thickness leads to lower capillary force and, thus, to lower friction (see Fig. 6). To elucidate the dynamics of the process the friction force curves as a function of time were also taken into consideration (see insets). When the water film is thick the frictional behavior appears to be liquid-like. The magnitude of the friction force is solely dominated by capillarity. As the pressure continues to decrease, capillarity vanishes and more subtle influences, for instance increased viscosity due to ordering were detected. After some time of liquid-like sliding behavior, solid-like sliding suddenly appeared, indicated by stick/slips. This occurred after passing the first minimum.

Stick/slips are characteristic for either dry sliding conditions or for ordered quasi-crystalline liquid layers. Then, friction increased up to the first local maximum. From this point one more minimum and maximum followed where the decrease in friction force was always accompanied by liquid-like sliding and the intermediate increase by solid-like sliding. At sufficiently low pressure, the friction became constant with a low degree of fluctuation [14]. Finally, the friction force curve left its main minimum and increased (not shown in Fig. 6).

In [6] a model for the molecular arrangement between the ball and the flat was proposed. The model is based on lateral ordering of water molecules [15] due to double layer forces [16 - 18]. Thus, when two solids are brought together the ordered layers of water constitute the contact. All additional water forms a capillary neck surrounding the contact region. In the course of the experiment this additional water vanishes by desorption but the double-layers remain in direct contact. When the first minimum is passed the sliding is solid-like, showing a distinct stress value of static friction. Then a transition to liquid-like sliding occurs. The static friction and the stick/slip amplitude decrease, see Fig. 7. After passing the next minimum solid-like sliding becomes again dominant. It can be concluded that the first transition – from liquid-like to solid-like sliding – appears when the water film has reached a stable arrangement of four monolayers. In this state the interdiffusion of double-layer water and bulk water ceases. Viscosity-induced friction instead of capillary-controlled friction is now the dominating factor. Further desorption seems to destroy the stable four-layer arrangement and the sliding takes on a more liquid-like property. In the transition...
period between this stable arrangement and the next stable state – probably the three-monolayer arrangement – the friction decreases, since the viscosity is lowered due to the intermediate disordering of the water layers. When the stable three-layer arrangement is achieved, the stick/slips appear again, indicating that solid-like sliding was re-established. Viscosity is again high, though on a decreased level. Finally, the same scenario takes place for the transition from three to two layers (not resolved in the experiment).

**Fig. 7: Stick/slip amplitude versus time.**

### 3.3 Cohesion-induced friction

As mentioned before friction increases for low water coverage after long pumping times. Here, the normal desorption is supported by friction-induced desorption leading to sub-monolayer coverage. The increasing friction is caused by cohesion between both solids [19].

To study these effects in greater detail, the silicon samples were stored in the vacuum chamber. After achieving the ultimate pressure the friction process was initiated to desorb the last monolayer. Then, ball and flat were disengaged and kept in this state for a certain amount of time. When the ball was re-engaged initially high friction followed by a strong decrease was detected (Fig. 8).

Calculations have shown that the water coverage of a free hydrophilic surface in uhv is much lower than half of a monolayer and can be estimated to $10^{-8}...10^{-6}$. The coverage is a result of the competition of two processes: adsorption from the gas phase and desorption. Adsorption rate is a function of molecule concentration while desorption depends on adsorption energy and surface temperature. At very low pressure the adsorption rate is very low, but the desorption rate remains constant. Therefore, at low pressures desorption is the dominating process and the free surfaces almost completely lack in physically adsorbed molecules.

When ball and flat come in contact the slit forms a trap for adsorbed molecules. The depth of the potential well doubles its magnitude which is crucial for the desorption rate, because of its exponential dependence on adsorption energy. As a result, the desorption rate decreases by several orders of magnitude and becomes comparable to the adsorption rate. Thus, the adsorbed layer grows. While free surfaces are practically free of adsorbates, the slit triggers the appearance of a considerable water film. After contact the locally increased concentration of adsorbate molecules spreads along the surface (or piles up) and molecules desorb until the adsorption-desorption equilibrium is established.

**Fig. 8: Friction force as a function of sliding time in ultra high vacuum. Before engagement ball and flat were not in contact. Due to capillary condensation in the slit formed between ball and flat water gathers shielding the surface atoms of both solids. As a result, cohesion drops and friction decreases.**

It is necessary to take into account the high mobility of adsorbed water molecules on the surface. Since the sliding velocity was rather low and comparable with diffusion rate of molecules, the majority of trapped molecules seems to be moved together with the ball along the surface. It looks like moving a finger on the glass with water drops. The water accumulates around the finger and moves together with it. The molecules which are locked on defects or impurities cannot move together with the ball desorb. Generally, accumulation rate seems to be higher than desorption – at least until coverage reaches a certain steady value, corresponding to equality of accumulation and spreading processes.

### 4 CONCLUSIONS

#### 4.1 Forces on different scales

Microtribometry using equipment that covers several orders of magnitude in tangential force ensures to detect force contributions acting in the milli-, micro- and nanonewton range. The main findings can be summarized as follows:

- Reducing dimensions of the tribological system increases the multiplicity of influences, which determine friction, adhesion and wear. Besides mechanical interlock, wettability, surface termination, local confinement of liquids and cohesion become decisive factors.
- From all influences on the microscale capillary forces in combination with topography expose the strongest action provided that electrostatic charging was eliminated.
Fig. 9: Loss mechanisms acting on different length scales. 
(a) Topography and capillarity have the strongest contribution to friction on the microscale. (b) On the lower microscale capillary bridging depends on the water film thickness. (c) Thin liquid layers under confinement are subject to increased viscosity, leading to higher friction. (d) Frictional interaction and loss on the atomic scale is mainly accompanied by molecular interaction (cohesion).

- The capillary effect increases the normal force and can be expressed by $F_c = 4\pi\gamma R$. However, the real contact situation must be analyzed accurately, since the given equation fails for limiting cases such as ultra-thin coverage.
- Liquid layers in direct contact with a solid surface lead to the balance of the electrical charges available on the surface and cause a water double layer.
- If two solids are brought together, then the contact formation is made first by the water double layers. Within the force area of microtribology this arrangement is retained (i.e., the water film is not penetrated by the solids) and experiences a further increase due to local confinement in the tribological contact.
- Additional water forms a capillary neck, which includes both double layers. In this state capillary forces dominate completely the tribological behavior.
- Variations of the sample temperature as well as the relative humidity affect the thickness of the water film. Friction rises with increasing humidity for hydrophilic samples.
- When the capillary bridge is removed, the order of water in the double layer arrangement leads to the increase of viscosity and thus to the increase of the friction.
- Viscosity increases – accompanied by solid-like friction – was detected for the case of the quadruple, triple and double layer arrangement. In the transient areas the friction was liquid-like, i.e., showed less fluctuations.
- If the water coverage is reduced to a monolayer then friction is small, since this arrangement shields van der Waals forces and/or chemical bonds between the solid. With penetrated monolayer friction increases, since both solids come into direct contact.

### 4.2 Comparison of micro- and macrosystems

When macro- and microsystems are compared the crucial question is whether the microtribological contact can simulate the effects on the macroscale:

- For macroscopic contacts one must differentiate strictly between the geometrical and the real contact area. While the geometrical contact is determined by the outer dimensions of the bodies, the real contact area is influenced by elastic and plastic deformations as well as fracture mechanics. The real contact area is smaller than the geometrical contact area. This effect can almost be neglected for microsystems, the geometric contact area is very often equal to the real contact area.
- Since the macroscopic compressive stress can be simulated by selecting suitable microscopic counterbodies, the temperature in the tribological microcontact should also resemble the temperature in a tiny fraction of the macrocontact. However, since the microcontact is isolated from other contacts, effects like heat dissipation or the whole concept of flash temperatures deviates strongly [20].
- On the macroscopic scale friction is dominated by roughness effects (when wear can be neglected). The most common loss mechanisms are vibrations (phonons) and mechanical deformation. When two opposite asperities are regarded separately, then the capillary bridge existing between them is responsible for the friction loss. Further in the dimensions reduced, viscosity effects appear due to local ordering phenomena. On atomic scale interatomic interactions play a role. All events mentioned prevail simultaneously, however the effect dominating on a certain force level is covered in its action by the mechanism acting on the next following force level. With suitable measuring techniques these mechanisms can be separated and regarded individually.

### 4.3 Validity of macroscopic laws at the microscale

- Macroscopic laws as of Amonton or Coulomb lose their validity, if single contacts are regarded. Experiments show that the size of the contact area has a crucial influence on the friction. Also the relation of friction to normal force loses its proportionality. It can be concluded that the coefficient of friction on the microscale has only limited meaning, because of strong fluctuations. These fluctuations, exactly like the influence of the individual asperities, average themselves on the macroscale.
- The dependence of the friction force on normal force obeys the Bowden Tabor model. The contact area occurring in the model can be determined by means of contact theory.

### 4.4 Lubrication effects as function of scale

Lubrication shows just like the other tribological entities a strong scale dependence. On the macroscale water can be classified as lubricant. In the microrange capillary
action dominates and water can be regarded as friction-increasing. Within the area of a few monolayers lubricating and friction-increasing effects alternate. In the state of a monolayer coverage water works again lubricating, since the monolayer shields van der Waals forces and suppresses the formation of chemical bonds.

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6 REFERENCES