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Towards thermo- and superlubricity on the macroscopic scale: from nanostructures to graphene and graphite lubrication

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Chapter 1

Introduction

Tribology - the study of friction, wear and lubrication of contacting materials - is a relatively new scientific discipline that has existed for only about 50 years. By contrast, the phenomenon of friction establishes one of the oldest engineering problems with roots dating back many thousands of years: first attempts to sledge on snow (10000 BC), the concept of the wheel (3500 BC), the transportation of massive pyramid stones by the Egyptians (2000 BC), etcetera. It has always remained a challenge to reduce that resistance of motion (friction) when one object rubs against another. Over time people have put much effort in understanding the nature of friction forces [1]. In particular, simple models were set up and phenomenological laws were derived [2], based on observations and measurements of friction forces for dry and lubricated interfaces. This has made it possible to provide a description of the behavior of simple mechanical systems on a practical level and to estimate the frictional energy losses. The experimental efforts got strengthened significantly by progress in microscopy techniques [3], which enabled research of the contacting surfaces down to the atomic scale and shed more light on the more microscopic aspects of the mechanisms of solid-solid interaction and lubrication. In turn, progress in the field of tribology contributes greatly to progress in many modern mechanical systems: wind turbines, brakes, hard-drives, valves, transmissions, wheels and others.

In the era of present-day technology, hand in hand with the development of nano- and microelectromechanical systems (NEMS/MEMS) [4] and other high-tech devices, the demand is increasing for a truly microscopic understanding and control of friction and wear. New, revolutionary approaches are called for at the nano- and microscale. On these small scales, most conventional lubricants (e.g. oil-based) fail to reduce friction for several reasons. Liquids introduce enormous capillary forces, while they bring in the risk of a variety of chemical reactions and they can add to the generation of wear debris [5].

In this PhD Thesis we demonstrate our first steps towards the reduction and control of dry, unlubricated friction on various geometrical scales, with solutions that have the promise to satisfy the boundary conditions set by some of the most pressing modern technological challenges. In Chapter 2 we describe the first stage of an experiment aimed at making the friction-lowering phenomenon of *thermolubricity* [6] active on the macroscopic scale. Our approach is based on earlier studies of friction, conducted in our research group with a Friction Force

Microscope (FFM). In these studies, it was shown that random thermal fluctuations change the familiar, atomic stick-slip motion of the atomically sharp tip of such a microscope into a thermal drifting motion, when these fluctuations are strong enough with respect to the barriers in the energy landscape. These excitations are concentrated in the last nanometers of the tip, because of the extremely small mass and the flexibility of the tip apex. Figure 1.1 shows a calculated dependence of the mean friction force on the true potential corrugation of the contact between the FFM tip and the substrate. The potential corrugation can be varied by adjusting the normal force on the contact [6]. Due to the thermal fluctuations of the tip apex there is a significant increase of the normal force required to see the friction force start increasing (Fig. 1.1).

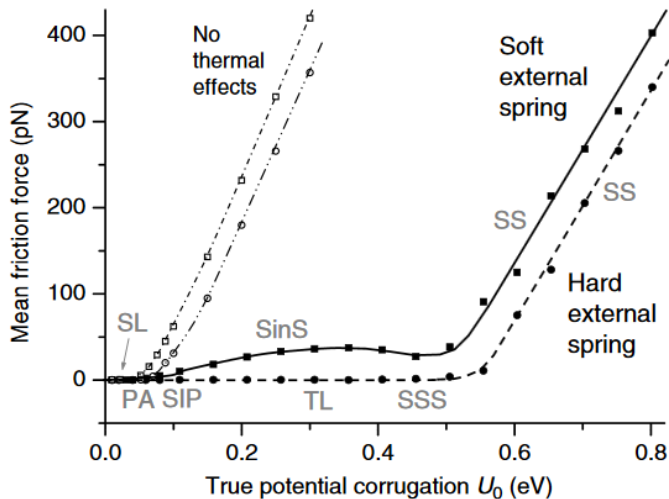


Figure 1.1. Mean friction force as a function of the corrugation of the interaction potential between the substrate and the AFM cantilever [6]. Note that the true potential corrugation is proportional to the actual normal load on the contact. Reprinted with permission from the authors [6].

To achieve similar behavior in a macroscopic contact with an area well beyond that of the very specific tip-surface geometry of an FFM, we have shaped one of the two, macroscopic contacting surfaces in the form of a micro-fabricated array of silicon nanopillars, each with a well-defined spring coefficient, similar to that of a standard FFM tip. This pattern can be regarded as a large multitude of FFM-like tips, each one exhibiting the thermal fluctuations that we identified as

lubricating the single-tip motion in an FFM. We discuss our friction experiments with these fabricated patterns and address the role of the elasticity of the Si nanopillars, the specific choice of the counter-surface, the influence of humidity and that of the normal load. The fabrication process of the Si nanopillar arrays is discussed in detail in Chapter 3 and in ref. [7].

As the second approach to tackle the problem of dry, unlubricated friction at the macroscopic scale, we use a monolayer graphene coating in an attempt to observe the effect of the phenomenon, known as *superlubricity*, on the macroscopic scale. In previous experiments, our group has demonstrated that graphite can bring down friction in nanometer-scale contacts by orders of magnitude [8]. Superlubricity is based on the incommensurability of two contacting crystal lattices that are rotated out of registry. On the nanoscale this can render the friction forces almost negligible. Figure 1.2 demonstrates the average friction force acting at the interface between a graphite nanoflake and an extended graphite surface as a function of the rotational angle between their lattices. Chapter 6 describes our exploration of the potential of superlubricity in a practical context; the grand challenge is to evoke this lattice-mismatch behavior on the macroscopic scale. To this end, we use centimeter-scale, flat surfaces covered with a specially grown, high-quality, single-orientation, single monolayer of graphene. Friction force traces are measured with a novel friction force microscope. First results indicate that graphene indeed has a significant effect on macroscopic friction. In Chapter 6 we also discuss the effect of oxidation of the substrate on friction properties of graphene at the nanoscale.

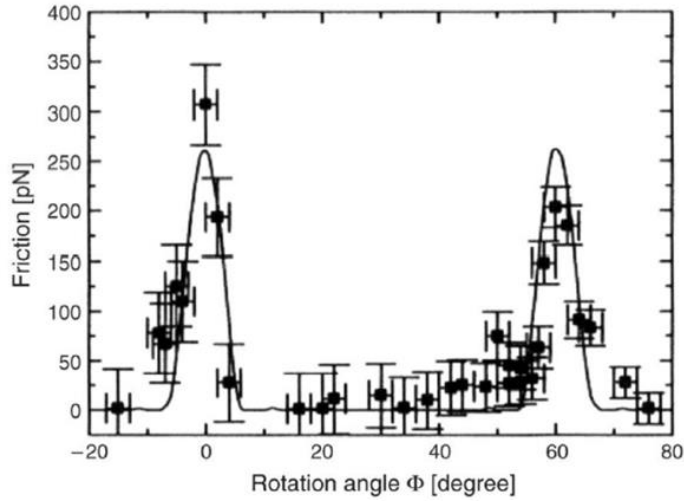


Figure 1.2. Friction force measured versus rotational angle by sensitive friction force microscopy between a nanometer-sized graphite flake and an extended graphite surface [8]. When the two graphite lattices are commensurate, at rotation angles of 0° and 60° , the friction force is high. When the lattices are rotated out of registry by only a few degrees, friction is low. This superlubricity effect has served as the source of inspiration for our experiments on friction between graphene-covered surfaces, described in Chapter 6. Reprinted with permission from the authors [8].

Interestingly, the signature of superlubricity can be associated not only with layered materials, such as graphite, molybdenum disulfide, boron nitride, etcetera [e.g. 9], but also with carbon-based, composite materials, such as Diamond-Like Carbon (DLC) coatings [10]. Even though DLC's form a well-known class of materials that reduce abrasive wear and friction, the origin of their low friction remains poorly understood. In Chapter 4 we report an experiment to explore the lubricating mechanism of DLC coatings. We describe our investigations of the frictional behavior of DLC coatings and draw conclusions about a possible influence of superlubricity in reducing the macroscopic friction of these coatings. For our experiments, we use a novel friction force microscope combined with more conventional surface characterization techniques. By micro-patterning one of the contacting surfaces, we have limited the region where mechanical contact can occur. This has enabled us to follow *all* changes in the contacting surfaces and directly observe the transformation of their structure and composition. In Chapter 5, we present an analysis of the frictional behavior of the micropatterned DLC

coatings as a function of relative humidity and sliding velocity, from which we draw conclusions about the lubricating mechanism of DLC and the role of wear particles, generated at the interface during sliding.

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