Sliding friction
From microscopic contacts to Amontons’ law
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CHAPTER 5

Molecular Probes Reveal Deviations from Amontons’ Law in Multi-Asperity Frictional Contacts

5.1 Abstract

Amontons’ law[1] defines a material-dependent constant, the friction coefficient, as the ratio between friction force and normal force[2]. Amontons’ law is commonly explained with the two non-trivial assumptions that both the frictional and normal force depend linearly on the real contact area between the two sliding surfaces[3]. Most surfaces are rough[4, 3, 5, 6, 7, 8, 9] and experimental testing of frictional contact models has proven difficult, because few in-situ experiments are able to resolve this real contact area. In this chapter, we present a new contact detection method with molecular-level sensitivity, and independently probe the two relations that form the microscopic origin of Amontons’ law. While the friction force is proportional to the real contact area, we find that this real contact area does not increase linearly with normal force. Contact simulations performed on the identical surface show that the breaking of Amontons’ law is due to both elastic interactions between asperities on the surface and contact plasticity of the asperities. We exactly reproduce contact area and fine details of the measured contact geometry by including plastic hardening into the simulations. These new insights into contact mechanics pave the way for a quantitative microscopic understanding of contact mechanics and tribology.
5.2 Introduction

A third of the world energy consumption is due to friction[10], but our fundamental understanding of how this friction emerges is not complete. All frictional theories ultimately aim to understand how frictional dissipation emerges from the details of contacts between two sliding surfaces. Experimental testing of such contact theories for rough interfaces is crucial, but has proven very challenging. In the late 19th and early 20th century electrical conductivity has been used as a measure of the contact area between metal surfaces[11]. More recently, optical techniques such as phase-contrast microscopy[12], frustrated total internal reflection[13] or interferometry[14] have been used to gain insight into contact and friction mechanics. However, two important aspects of contact mechanics and their relation to friction have not been addressed by these experiments. First, can deformations of the roughness be elastically transferred from one contact point to another and thereby influence the contact area? Second, what is the relative importance of plasticity and elasticity in the formation of contact area and friction?

5.3 Experiment

Answering these questions requires a detailed experimental observation of the real contact area. To view the real contact area, we use a new optical technique[15], which employs rigidochromic molecules. When absorbing a photon, the rigidochromic molecules show excited-state deactivation along two distinct pathways[16, 15, 17, 18]. The first pathway is non-radiative (non-fluorescent) and triggered by rotation around a specific bond in the molecule. When this rotation is hindered by the confinement induced by a mechanical contact[15], the molecule is forced to follow the second, radiative, pathway: it fluoresces[19, 15].

In the experiments, we chemically attach such molecules to the surface of very smooth and flat glass cover slips[15] which are then inserted into our microscopy setup (Figure 5.1). A sphere is lowered into contact with the cover slip and the contact is illuminated from below, to excite the monolayer of rigidochromic molecules at the surface of the cover slip. The molecules fluoresce when the gap between sphere and cover slip becomes of the order of the molecule size (Figure 5.2 and Appendix B). The integrated fluorescence intensity is proportional to the number of confined molecules and weakly depends on the local contact pressure (see Appendix B). In the plane, we re-
solve the contact structure with diffraction limited microscopy (point spread function of 450 nm, see Appendix B). The fact that contact pixel intensities do not vary much spatially or with load, indicates that there is not much contact structure below this scale (see Appendix B).

Figure 5.1: Experimental setup (not to scale). A rheometer is mounted on top of an inverted confocal laser scanning microscope. We excentrically glue a rough sphere to the rheometer plate and make contact with a smooth and flat, float glass, cover slip. The rheometer measures normal and frictional forces on the contact. The inverted microscope excites a monolayer of rigidochromic molecules on the glass surface with 488 nm laser light and point scans images (at a large magnification: 63x, n.a. 1.4) of the resulting fluorescence that is emitted at the real contact area between the sphere and the glass. Two beam splitters and a long pass filter are used to collect the fluorescent light in a photomultiplier tube. To avoid strong light scattering and optimize image quality, we immerse the contacts in formamide and use transparent materials for the sphere: polystyrene (PS), poly(methyl methacrylate) (PMMA), polytetrafluoroethylene (PTFE) and Borosilicate Glass.

5.4 Results

In the experimental range of normal forces, the real contact area evolves from a discrete collection of asperities in contact at 4 mN to an almost Hertzian contact circle at 400 mN. During this evolution, existing contacts deform and increase their area while new contact patches emerge elsewhere. Quite surprisingly and contrary to the common interpretation of Amontons’ law, the overall contact area does not increase linearly with the normal force.
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(Figure 5.3a).

![Diagram of contact area and simulations](image)

**Figure 5.2:** The real contact area measured and simulated at increasing loads. 

a), Fluorescence intensity images of the contact geometry. The average contact pressure, defined as the ratio of contact force and contact area, rises from roughly 100 MPa at the beginning of the experiment to 250 MPa at the highest loads. b), Elastic Greenwood and Tripp (GT) bearing area calculation. c), Purely elastic simulation. d), Elasto-plastic contact hardening simulation. Experiments and simulations were carried out on the sphere whose roughness is shown in Appendix B. Simulated contact geometries are convoluted with the point spread function of the microscope (see Appendix B).

If the contact area links the normal force to the friction force, this observation would imply that Amontons’ law is broken. To induce frictional slip and measure the friction coefficient, we rotate the rheometer plate (Figure 5.1) at
5.4 Results

Figure 5.3: Amontons’ law and the real contact area. a) Real contact area vs. normal force. Symbols show experiments on three PS spheres that have similar roughness. Solid lines show values obtained from theory as well as linear fits to the penetration hardness model, with $p_Y$ the penetration hardness, and the fully elastic simulation, with $p_{\text{rough}}$ the constant contact pressure. The inset shows the same data, but on a logarithmic scale. Experimental contact is reproduced by the contact hardening model that considers long range elastic asperity interactions and local plasticity at contact. Other models either underestimate the contact area or do not describe the deviation from linearity found in the experiment. b) Static friction force of contacts like those in (a), measured at different normal forces. Symbols show experiments on two PS spheres, the red solid line is the hardening simulation fitted onto the friction data by multiplication with the interfacial shear strength. The agreement shows that the static friction force is proportional to the contact area. The constant of proportionality, or interfacial shear strength, is 50 MPa, close to the bulk shear strength of PS. Inset: the friction force $F$ between a PS sphere and a glass substrate as a function of applied strain $d$ measured using a rheometer. Through rotation of the rheometer plate (Figure 5.1), a constant strain rate of $\sim 1 \mu m/s$ is imposed on the contact. The friction force builds up until slip occurs. The static friction is then defined as the maximal friction force at the onset of slip, measured at different normal forces, $N$, shown in the inset. Friction and contact data recorded during the event indicate that there is no stick slip behaviour at the imposed sliding velocity.

a constant velocity of $1 \mu m/s$ resulting in a linear build up of friction force, caused by the finite stiffness of the measurement system (inset Figure 5.3b). Once the applied force exceeds the static friction, the contacts break and slip. We indeed observe that Amontons’ law is broken; the static friction force is
proportional to the contact area but not to the normal force (Figure 5.3b). This means that the friction coefficient is ill-defined, it depends on the normal force.

The experiments thus show that friction is controlled by the contact area, but not what sets the contact area. Many of our present-day insights into the mechanics of rough contacts come from theoretical considerations. Early models assumed surfaces deform purely plastically[4, 3]. In these models, surface roughness causes the contact area to be small and therefore the contact pressure to be large. This enormous pressure leads to irreversible, plastic deformation of the contact points. The real area of contact \( A \) is then proportional to the load \( N \) pushing the surfaces together, \( A = N/p_Y \) with \( p_Y \) the penetration hardness of the material. It was argued that after the first, irreversible deformation of the material, it would respond purely elastically; this led to the development of sophisticated multi-asperity models[5, 21, 6, 7]. The first multi-asperity theory by Greenwood and Williamson[5] approximated surface roughness as a collection of identical, non-interacting, spherical summits of varying height. Greenwood and Williamson showed that if these asperities follow Hertzian[20] contact mechanics and have a Gaussian height distribution, the real contact area grows in proportion to the load. This result demonstrated that linearity between contact area and load can also exist in rough elastic contacts, despite the fact that a single sphere-on-flat contact area grows with the load to the 2/3 power according to Hertz theory of elasticity[20]. Since the pioneering work of Greenwood and Williamson, asperity theory has been refined to describe more complex asperity shapes, but the base assumption has always remained that a rough surface is a discrete collection of independent asperities. Persson’s recent scaling theory[22] alternatively, uses a description with an arbitrary form for the roughness, taking into account the fractal nature of surfaces that causes smaller asperities to exist on top of larger ones. The overall real contact area predicted by both theories is very similar and follows the form \( A = N/p_{\text{rough}} \) with the characteristic contact pressure exerted on the roughness \( p_{\text{rough}} \approx h_{\text{rms}}' E^*/2 \) now determined by the elastic contact modulus[20] \( E^* \) and the root mean square slope \( h_{\text{rms}}' \) of the surface roughness.

To investigate why in the experiments proportionality between contact area and normal force is not observed, we perform simulations\(^3\) in which different effects can be considered or left out. Prior to the contact experiment, we use Atomic Force Microscopy (AFM) to obtain a three dimensional map of

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\(^3\)The contact simulations were performed by Till Junge and Lars Pastewka from the Karlsruhe Institute of Technology.
the sphere roughness at the exact same location that is pressed onto the glass (Appendix B). Since we use materials for the sphere that are significantly softer and rougher than the glass, the deformation of this roughness map completely determines the experimental contact area and therefore forms the ideal input for contact simulations. We first consider elasticity and start with the rough-sphere multi-asperity model of Greenwood & Tripp (reference [21] and Appendix B) to compute the dependence of the contact area on normal force. Surprisingly, the contact area resulting from this calculation is five times smaller than that found in the experiment (Figure 5.2b). By reducing rough surfaces to a collection of discrete asperities, multi-asperity theories such as the Greenwood & Tripp model ignore strain transmitted from asperity to asperity through the bulk.

The omission of such interactions from multi-asperity theories is considered to be problematic[22], because many surfaces are fractal; smaller asperities exist on top of larger asperities implying that in contact, asperities have to transmit strain to one another. We therefore compare the Greenwood & Tripp model to a full numerical calculation of the contact area using a Green’s function method[23] that treats the elastic interaction exactly on all length scales. This simulation ignores nonlinear elastic effects but constitutes the exact solution of the problem that both multi-asperity theories and Persson’s analysis approximate. There are no adjustable parameters in the elastic simulation, because sphere radius, sphere roughness and modulus are all independently measured (see chapter 2 and Appendix B). We observe that the inclusion of asperity interactions leads to a different contact patch distribution (Figure 5.2c) compared to that of the Greenwood & Tripp model. The contact morphology obtained by elastic simulation is closer to the experiment, leading us to conclude that asperity interactions are required to more accurately predict the real contact area. The real contact area from the elastic simulations, however, is still linear in normal force and still significantly smaller than in the experiment (Figure 5.3a).

If one estimates the stresses at the contacts from the measured forces and real contact areas, one obtains values on the order of 200 MPa. Since the penetration hardness the PS is of the same order[24], irreversible, plastic deformation of the asperities may occur in addition to elastic deformation. To confirm that plasticity is indeed important in the experiment, we measure the surface topography by AFM after the contact experiment; we observe that indeed the contact points have been permanently deformed (Appendix B). We therefore add plasticity into the simulation, first by using the canonical plasticity model of contact mechanics: We allow contact points to flow
above a penetration hardness $p_Y$ (see Appendix B). $p_Y$ is set to 10% of the PS elastic modulus, three times higher than the yield strength of PS under compression[3]. Although the resulting contact area (Figure 5.3a) is about twice the size of that predicted by the purely elastic simulation, it is still significantly smaller than that measured in the experiment. By varying the only adjustable parameter, the penetration hardness, the match between experimental and simulated contact patterns cannot be improved significantly (Appendix B). More importantly, the real contact area still depends linearly on the load, in disagreement with our experimental findings.

The likely solution comes from carefully looking at the experimental data, and comparing to the purely plastic model discussed above. From the latter, one would expect the contact pressure to remain constant at the value of the penetration hardness of the material. However, due to the sublinear dependence of the contact area on the load, the average contact pressure rises during the experiment from roughly 100 MPa at the lowest loads to 250 MPa at the highest loads. This strongly suggests that the contacts become harder to deform at large strains; such strain hardening is generally observed for the materials employed here[25]. To capture this effect, we introduce simple linear hardening of $p_Y$ with local plastic displacement $h^{pl}$, $p_Y = kh^{pl}$, into our calculation (for details see Appendix B). This is the simplest constitutive equation for a strain hardening model; we adjust the single empirical parameter $k$ to match our experimental contact area vs. load curves, giving $k \approx 4$ MPa nm$^{-1}$.

The hardening simulation predicts contact geometries that are almost indistinguishable from the experiments (Figure 5.2), including also the deviation from linearity of contact area with load (Figure 5.3a). Using the surface topography map as input, we can now predict exactly where contact will occur, and where not.

5.5 Conclusion

In summary, the first determination, with molecular resolution perpendicular to the plane, of the real contact area in a frictional contact shows that (as commonly assumed) the static friction is directly proportional to the real contact area. However, we also observe that this contact area does not grow linearly with normal force, which is the result of elasto-plastic deformation of the asperities that constitute the roughness. The deformation of the asperities and the resulting contact geometry can only be predicted accurately by taking
into account the elastic interactions between contact points and combining them with a strain hardening plasticity law. The former is commonly ignored in multi-asperity models and the latter in most numerical calculations. We expect the elastic behavior observed here to be applicable to most other materials, while the surface plasticity may be more material dependent. In any case, the hardening model presented here also describes the contact mechanics of PMMA (Appendix B) and the nonlinear contact area and friction is also observed in PTFE and glass spheres. We anticipate these results to lead to a better understanding of many tribological problems such as the running-in of frictional contacts[26] that is key in engineering applications and slip-weakening[27, 28] in geology. Both are related to contact plasticity that determines the initial change in friction coefficient and surface roughness of a tribological system that controls much of its subsequent tribological properties.
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