The origins of friction and the growth of graphene, investigated at the atomic scale

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Chapter 2

The dissipation of a single-asperity contact: a problematic discrepancy

2.1 Background

In the previous chapter, the PT-model was introduced. This classical model is very successful in capturing many features of atomic-scale friction experiments. However, as was already indicated, there are two ways in which this PT-model does injustice to the physics that is crucial for friction. First of all, dissipation is not instantaneous. It must be governed by a finite dissipation rate that reflects the actual mechanism by which the excess energy is removed from the accelerated slider. Secondly, according to the fluctuation-dissipation theorem, the dissipation couples the slider to the thermal bath of the dissipating medium and, hence, introduces thermal noise.

Introducing these two elements, finite dissipation and noise, leads to an alternative and somewhat more sophisticated version of the PT-model, in the form of the Langevin equation given in Equation 1.2. In this picture, the effective mass represents the tip plus a significant part of the cantilever, which can be viewed as moving together with the tip. It can be estimated easily on the basis of the cantilever dimensions or it can be taken from the readily measured eigenfrequency of the cantilever, in combination with the spring coefficient of the cantilever. The effective spring coefficient differs from the cantilever spring coefficient. It can be measured directly from the 'stick'-part of a force-versus-displacement curve (cf. Figure 1.2d) and usually is dominated strongly by the flexibility of the tip apex, which acts as a soft spring. Typical values used for these parameters are an effective

mass in the order of 10^{-11} kg and an effective spring coefficient of 2 N/m.

The only free parameter left in Equation 1.2 is the dissipation rate γ, which is therefore used as a kind of fitting parameter in order to reproduce the experimentally observed data. The resulting value for γ is typically around 10^{-6} kg/sec[6, 11, 12, 21]. A more refined two-mass-two-spring model used by Maier [15] was applied using a similarly small 'tip'-mass of 10^{-12} kg and a dissipation parameter tuned such that the stick-slip patterns were observed. The value for the small mass was varied by a factor 10 and yielded no observable differences in the calculations[15].

Even though the Langevin equation (Equation 1.2) and the related numerical descriptions of the atomic-scale sliding motion seem to reproduce experimental observations successfully, they do not lead to a physical understanding of the friction mechanism: what does the effective mass mean and how do we have to interpret the values of the dissipation rate obtained from the fitting procedures? Actually, as our studies aim to understand the friction parameter, we do not want to use it as a fitting parameter, but prefer to argue its origin and try to predict its value.

2.2 The friction force on a single atom

In order to provide a better basis for understanding the dissipation rate, we will construct a simple, bottom-up description of the dissipation that is easy to quantify. By first studying the friction of a single atom, the dissipation rate of a small friction contact composed out of several atoms can be established. We will use this approach as the quantitative basis for our model calculations.

To get an estimate of the typical time it takes for an atom in the friction contact to lose its excess energy, we resort to related subjects in the field of surface science. Especially both experimental and theoretical studies on atomic and molecular vibrations and studies on diffusion and jumps of atoms and small molecules on surfaces are helpful[22–25]. These show that the motion of atoms and molecules on surfaces is approximately critically damped: the time it takes to dissipate excess momentum is in the order of the natural vibration frequency of the atoms or molecules on the surface, which frequency is in the order of 10^{12} Hz.

2.3 The dissipative force on a single asperity

We now assume that the momentum dissipation rate of an atom, γ_{at}, present at an atomically sharp tip, in contact with a substrate, is of the

same order of magnitude as the dissipation rate of a single atom on a surface. In addition, we assume that, in case the tip apex is not atomically sharp, the total dissipation rate scales linearly with the number of atoms in the tip apex that make contact with the substrate, N_c. This second assumption might seem to be crude and one might argue a sublinear dependence would be more appropriate as the dissipation per atom might degrade when more than one atom is constituting the friction contact. However, as will become clear at the end of our study, the possible overestimation of the total dissipation rate will make our conclusions even stronger.

Finally, we assume that the basic dissipation form in Equation 1.2, in which the dissipative force is proportional to velocity, remains valid. The combination of these three assumptions should enable us to produce a coarse prediction of the friction force on a complete asperity given its contact size, which is what we will perform in the next section.

The above assumptions can be combined into the following equation for the friction force experienced by a tip that has N_c atoms in contact with the substrate and moves over the substrate at a velocity of $\dot{x}_t$:

$$F_{diss} = -\gamma_{diss}\dot{x}_t = -\gamma_{at}N_c\dot{x}_t \tag{2.1}$$

A subtle difference with typically used descriptions of the friction force, is the absence of the (effective) tip mass. The only (implicit) mass dependence is present in the factor $\gamma_{at}N_c$. One should note that in the approach presented here, we associate the actual dissipation entirely with the atoms that make up the frictional contact. It is their number and their velocity that count. The resulting dissipation force is independent of the mass of the rest of the tip, even if that moves at the same velocity as the contact atoms. Even though this may seem obvious, this approach is not followed generally.

Already at this stage, an estimate can be made of the dissipation rate γ_{diss} of a single asperity. In reports containing data from FFM-experiments, typically the size of the tip apex (the friction contact) is discussed[26–29]. In these experiments, point defects in the substrate lattice or step edges on crystal terrace were used to estimate the tip size. Typical sizes of the realized FFM friction contacts (N_c) vary from single-atom up to tens of atoms. For this moment, we will assume a contact size of 10 atoms. If tungsten is chosen to be the tip material, the characteristic lattice vibration frequencies are in the order of 10^{12} Hz. Taking these values, the momentum dissipation rate of the friction contact is in the order of 10^{-12} kg/sec.

The value estimated here for the dissipation rate that should be expected for a typical FFM friction contact is extremely much lower than

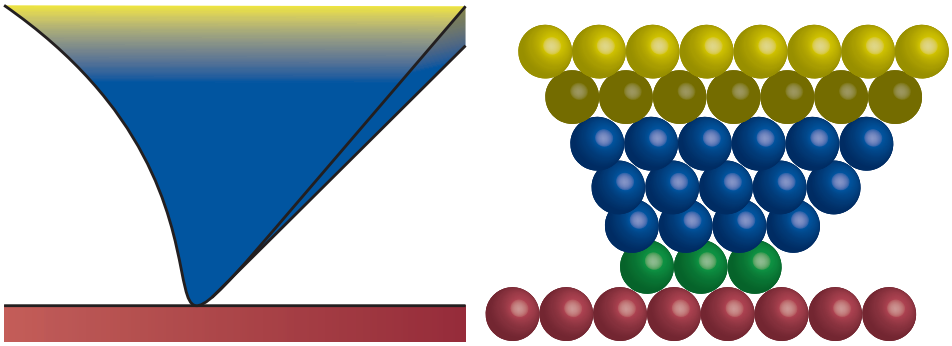

Figure 2.1: A (a) macroscopic and (b) nanoscopic schematic view of the deformation of the tip apex, prior to a slip event. The green tip apex atoms are the N_c atoms that make contact with the red substrate. The blue atoms share most of the lateral displacement of the green atoms. Together with the green atoms, they establish the N_d 'dynamic' atoms that will be accelerated most in the upcoming slip event. The lateral displacement of the yellow atoms is so modest, that these atoms are associated with the rigid part of the tip.

the values typically obtained when one uses the dissipation rate as the fitting parameter in the Prandtl-Tomlinson type Langevin equation shown in Equation 1.2 (cf. Section 2.1). The discrepancy is as large as six orders of magnitude. Introducing a sublinear relation between γ_{diss} and N_c, as a further element of sophistication in our description, would make the discrepancy even larger.

Using the low value for the dissipation rate, estimated above, the Langevin equation predicts heavily underdamped motion of the tip, resulting in a high probability for slip events over multiple lattice distances and thereby completely ruining the visibility of the atomic periodicity in the lateral force images.

2.4 Finding the origin of the discrepancy

When we re-inspect the Langevin equation (Equation 1.2), we recognize that the only parameter that is not defined rigorously, is the effective mass m_{eff}. We will now first discuss the character of this mass and then establish a mathematical derivation of this mass.

The effective mass comprises all atoms that move with approximately

the same velocity as the N_c atoms of the contact. This includes the N_c contact atoms and all other atoms that can be regarded as effectively forced to move together with these contact atoms. Typically, this mass is interpreted as being both the tip and a sizeable part of the cantilever, resulting in a value of around 10^{-11} kg. However, the relatively soft effective spring coefficient, by which the contact is connected to the support ($k_{\text{eff}} = 2\,\text{N/m}$), suggests that an alternative choice for this mass could be more appropriate, namely that of a small portion of the tip apex, as schematically shown in Figure 2.1a. In this figure, the tip apex is drawn while it is deformed by the shear force exerted by the substrate. Most of the deformation is concentrated near the very end of the tip.

A more detailed schematic of the tip apex is shown in Figure 2.1b. In this schematic, the tip is atomically resolved. The N_c tip atoms that are in direct contact with the substrate are coloured green. Atoms close to the friction contact, which are part of the deformed tip and hence are part of the effective mass too, are coloured blue. For reasons that will become even more clear later, the amount of atoms taking part of the effective mass (the green and blue atoms in Figure 2.1b) is denoted as N_d, the amount of 'dynamic' atoms. We propose to replace the combined effective mass m_{eff}, used in Equation 1.2 to describe the motion of the contact and the resulting dissipation, by the dynamic mass m_d.

2.5 Estimating the dynamic mass

We can easily derive the effect that the dynamic mass has on the experienced dissipation force in the following way. During a slip event, the apex of the tip is accelerated under the combined influence of the spring force that it experiences and the interaction force with the substrate. Over the slip distance this provides the tip apex, i.e. the dynamic mass, with an amount of kinetic energy that is independent of its own mass. The maximum velocity that this kinetic energy corresponds to, is therefore inversely proportional to the square root of m_d. According to Equation 2.1, we should thus expect that:

$$F_{\text{diss}} \propto 1/\sqrt{m_d} \tag{2.2}$$

We see that the combination of the observed, high friction forces with our low expectation value for the microscopic dissipation rate indicates that the dynamic mass is microscopically small. The earlier discrepancy of six orders of magnitude is resolved when we assume a value for m_d of 10^{-23} kg, i.e. twelve orders of magnitude lower than the typical value of 10^{-11} kg for

the tip plus part of the cantilever. This small dynamic mass corresponds to a very modest number of atoms, in full accordance with the notion, introduced above (Figure 2.1), that it is associated with the very end of the tip apex.

The reduction of the dynamic mass to such a small size, means that only the very end of the tip apex is deformed significantly during the stick phase. According to Equation 1.3, the observed effective spring coefficient k_{eff} gives a good estimate of the flexibility associated with the dynamic mass. The typical experimentally found value of k_{eff} ($2\,\text{N/m}$) agrees with our picture of a very small, flexible end hence dynamic mass that is performing the SS-motion. For this reason, from here, we will denote the spring coefficient of the dynamic mass by k_{d} instead of k_{tip}.

An alternative, but equivalent line of argumentation to estimate the size of the dynamic mass, is based on the two typical timescales involved. On the one hand, we have the eigenfrequency of the dynamic mass. This sets the timescale for each slip event in terms of the dynamic mass and the effective spring coefficient, via the following relation:

$$t_{\text{slip}}^{-1} = 2\omega_{\text{d}} = 2\sqrt{\frac{k_{\text{d}}}{m_{\text{d}}}} \tag{2.3}$$

The other timescale is given by the time the friction contact needs to dissipate the excess energy. This timescale is set by a damping rate, denoted by η_{diss}, expressed in $[\,\text{sec}^{-1}]$. This damping rate is related to the dissipation rate, via

$$t_{\text{diss}}^{-1} = \eta_{\text{diss}} = \gamma_{\text{diss}}/m_{\text{d}} \tag{2.4}$$

where $\gamma_{\text{diss}} = N_{\text{c}}\gamma_{\text{at}}$, as before. We express the atomic dissipation rate γ_{at} as $\gamma_{\text{at}} = m_{\text{at}}\eta_{\text{at}}$, in which η_{at} is that atomic damping rate.

As the motion of single atoms is close to critically damped (see above), the atomic damping rate can be expressed as follows:

$$\eta_{\text{at}} = 2\omega_{\text{at}} = 2\sqrt{\frac{k_{\text{at}}}{m_{\text{at}}}} \tag{2.5}$$

where ω_{at} is the atomic vibration frequency and k_{at} the characteristic atomic bond stiffness. Based on these relations, the damping rate of the contact can be estimated to be

$$t_{\text{diss}}^{-1} = \eta_{\text{diss}} = \frac{\gamma_{\text{diss}}}{m_{\text{d}}} = \frac{N_{\text{c}}}{N_{\text{d}}}\frac{\gamma_{\text{at}}}{m_{\text{at}}} = \frac{N_{\text{c}}}{N_{\text{d}}}\eta_{\text{at}}. \tag{2.6}$$

How are the two timescales, t_{slip} and t_{diss} related? The mere observation in FFM experiments of atomic lattices characterized by stick-slip motion of

the tip apex, indicates that the excess, potential energy that is stored in the effective spring during the stick-phase is released and dissipated fully during the slip-phase of the tip apex. All potential energy is converted to kinetic energy, which is dissipated via the sliding tip. This requires the dissipation time t_{diss} to be smaller than or equal to the slip time t_{slip}. Apparently, the system has to be at least critically damped. This allows us to connect those two timescales explicitly. For convenience, we define the following relative damping rate:

$$D = \frac{\eta_{\mathrm{diss}}}{2\omega_{\mathrm{d}}} = \frac{\gamma_{\mathrm{diss}}}{2\sqrt{m_{\mathrm{d}} k_{\mathrm{d}}}} \tag{2.7}$$

so that $D = 1$ corresponds precisely to the critically damped case. The relative damping rate D can be rewritten in a form that depends only on numbers of atoms:

$$D = \frac{N_{\mathrm{c}} m_{\mathrm{at}} \eta_{\mathrm{at}}}{2\sqrt{N_{\mathrm{d}} m_{\mathrm{at}} k_{\mathrm{d}}}} = \frac{2N_{\mathrm{c}} m_{\mathrm{at}} \omega_{\mathrm{at}}}{2\sqrt{N_{\mathrm{d}} m_{\mathrm{at}} k_{\mathrm{d}}}} = \sqrt{\frac{N_{\mathrm{c}}^2}{N_{\mathrm{d}}} \frac{k_{\mathrm{at}}}{k_{\mathrm{d}}}} \tag{2.8}$$

The condition that the motion of the contact is at least critically damped, can thus be expressed as:

$$N_{\mathrm{d}} \leq \frac{k_{\mathrm{at}}}{k_{\mathrm{d}}} N_{\mathrm{c}}^2 \tag{2.9}$$

This rather simple inequality has a strong implication. As the factor $k_{\mathrm{at}}/k_{\mathrm{d}}$ is close to 1 for typical FFM experiments, the dynamic mass has to be extremely small. This means that for a typical 10-atom contact the dynamic mass consists of at maximum 100 atoms, corresponding to a microscopic mass in the order of 10^{-23} kg. As discussed before, this mass is some twelve orders of magnitude below the effective mass that is typically associated with FFM experiments.

The ultra small size of the dynamic mass that is exploring the substrate lattice continuously and is performing the stick-slip motion, confronts us with a fundamental change of interpretation of its behaviour. Due to its size, its eigenfrequency is in the terahertz regime. As a result, the tip apex is exploring the lattice in trajectories in which it reaches extremely high velocities. This enables the tip apex to dissipate the released slip energies with extreme efficiency.

The estimates and scaling relations, presented above, ask for verification by use of adequate numerical simulations. In order to investigate and interpret this new friction scenario further, a combined 2-mass-2-spring model is necessary in which both the extremely high dynamics of the tip apex and the slow response of the cantilever are taken into account. This new

model is introduced in the next chapter, after which the results and their interpretation are given in Chapter 4.

2.6 Summary

By an estimation of the friction force on a single atom, we were able to calculate the typical dissipation rate of a friction contact. This value was found to be approximately six orders of magnitude lower than values typically used in the literature. The solution of this discrepancy was found in the choice for the mass that is deformed significantly during the stick phase and that moves significantly during the slip phase. We argued that this mass corresponds to a very modest number of atoms that is associated with the very end of the tip apex. The experimentally found value for the effective spring constant agrees with our picture of a very small, flexible and dynamic mass that has a value in the order of 10^{-23} kg. Due to this small mass, the tip apex acquires extremely high velocities, which enables the tip apex to dissipate the released slip energies with extreme efficiency. Our findings are in agreement with the observed SS-behaviour in experiments, that predicted an approximately critically damped system.