# Describing Nanoscale Friction with Fully Atomistic Simulations and Rate Theories

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Because of its fundamental importance in the design of reliable nano-mechanical devices, nanoscale friction (i.e., the dissipation of energy at driven nanoscale interfaces) has been an area of extreme interest from both the fundamental science and engineering perspectives. However, because of the difficulties inherent in simulating the process in full atomistic accuracy over sufficiently long time and length scales, and because of the challenges associated with handling high-dimensional theories, a complete understanding of friction is still lacking. By using a combination of direct simulations using AMD and a fully parameterized high-dimensional rate theory, we demonstrate that the physics of friction can be captured in a way that allows for quantitative predictions over a wide range of conditions. In contrast, popular simplified theories are shown to provide only a qualitative description of friction.

The characterization of the velocity and temperature dependence of friction forces has been of particular focus in nanoscale friction. Experimental studies using friction-force microscopes have shown that mean friction is significantly affected by both velocity and temperature. However, the specific form of these dependencies is still a subject of debate and the understanding of the underlying physics remains imperfect. Theoretical approaches employed to investigate this problem can be broadly divided into two classes: (1) the direct simulation of fully atomistic models using molecular dynamics (MD), and (2) the development of rate theories based on a simplified, low-dimensional representation of the system. The former is typically being used to elucidate the atomistic detail of the process, while the latter is relied upon to provide analytical guidelines to assist in the interpretation of measurements. Indeed, an advantage of rate-theory-based models is that they enable one to make predictions over experimentally relevant time and length scales that are inaccessible to direct MD simulations. However, in contrast to MD simulation, rate theories require a large amount of non-trivial information about the system in order to be parameterized. Because of that, parameters are usually extracted from greatly simplified models of the experimentally relevant physical systems, which makes comparison with experimental results difficult. Therefore, at the present time, our theoretical descriptions of friction are either very accurate, but valid in a very limited range of conditions, or far-reaching, but with a limited accuracy.

In order to address this serious issue, we adopt a two-pronged approach: (1) using state-of-the-art accelerated MD (AMD) simulations methodologies developed at LANL, we significantly extend the time scales accessible to direct fully-atomistic simulations, and (2) using the same atomistic model we then fully parameterize a rate-theory description of the system that contains no adjustable parameters. This enables us to assess the accuracy of the full high-dimensional rate theory, as well as that of some of its common low-dimensional approximations [1].

The model system we simulate, shown in Fig. 1, corresponds to the interfacial region of a friction force microscope (FFM), the most commonly used apparatus to study friction at the nanoscale. In a real FFM, the tip is attached to a large elastically compliant cantilever. The deflection of the cantilever is measured as the tip is dragged along the surface, and the corresponding friction force at the interface is inferred. Because of the size limitations of MD, we cannot explicitly model the cantilever; instead we use a rigid support that is elastically tied to the tip in order to account for the proper elastic response.

AMD simulations were carried out with the Parallel Replica Dynamics (ParRep) method [2,3]. ParRep can be seen as a specialized parallelization scheme that operates in the time domain, instead of in the space domain as is usually the case. This way, long time scales can be simulated on small systems, something that is not possible with standard approaches. Using this technique, we extend the time scales amenable to direct simulations by a factor of a few hundred. In parallel with AMD simulation, we use the same atomistic model to parameterize a rate theory description of the friction process. Our description is based on a master equation with rates computed from the high-dimensional Transition State Theory in the harmonic approximation (h-TST) [4]. This requires the calculation of energy barriers and vibrational spectra



*Fig. 1. Atomistic configuration used in the present study. Pink atoms (bottom three layers) are fixed, and red atoms (top three layers) are tied one-to-one to the green support atoms (smaller atoms in the top three layers). Blue atoms (middle) are unconstrained.*
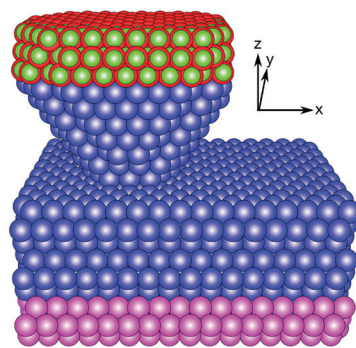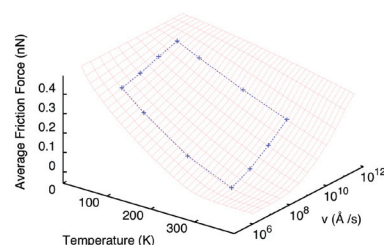
*Fig. 2. Average friction force as a function of temperature and scanning velocity. Red surface: rate-theory model; blue symbols and dotted line: ParRep simulations. Error bars on the atomistic results are comparable to the symbol size.*

for many positions of the tip along the surface. Once these parameters are numerically obtained, the average friction force is calculated by a numerical integration of the master equation for a given temperature and scanning velocity.

The comparison of the AMD simulations with the rate theory results is shown in Fig. 2. These results clearly demonstrate that a full rate theory captures the essential physics of friction. Indeed, the rate theory results agree with the direct AMD simulations to within few percent-error bars. This offers a convincing demonstration that rate theory is the proper theoretical tool to investigate friction. This is an important finding because, once parameterized, rate theories can provide predictions for a very wide range of conditions, often far out of the range amenable to direct simulations with MD or even AMD.

In order to assess the validity of commonly used simplified rate theories, namely the Ramped Creep (RC) model of Sang et al. [5] and the Thermal Drift (TD) model of Krylov et al. [6], we compare their predictions to our full rate theory. As can be seen in the insets of Fig. 3, these models provide a reasonable qualitative description of the high-velocity/low-temperature regimes (in the case of RC) and of the low-velocity/high-temperature regimes (in the case of TD). However, significant deviations can still be observed. We were able to determine that these deviations stem from intrinsically high-dimensional effects that cannot be captured by simplified models in a few dimensions. This indicates that, while they are useful for providing a first-order description of friction, they cannot provide quantitative predictions.

In conclusion, by using a combination of fully atomistic AMD simulations and high-dimensional harmonic Transition State Theory, we have demonstrated that the physics of nanoscale friction can be captured essentially exactly by rate theories when such a theory is fully parameterized from an atomistic model. We also show that simplified rate theories based on low-dimensional representations of the system capture the basic features of friction, but lack the ingredients to provide a quantitative description of the phenomena. Our work greatly extends the reach of the theoretical description of friction at the nanoscale and opens the door to a vigorous interaction between theory and experiment.
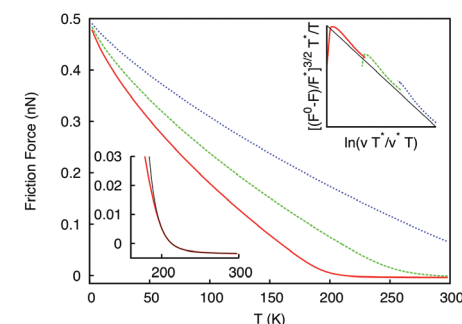


*Fig. 3. Average friction force as a function of temperature for different scanning velocities, as inferred from the rate theory. Red continuous line: 104 Å/s; green dashed line: 106 Å/s; blue dotted line: 108 Å/s. Bottom inset: blowup of the high-temperature region. The thin black line is a high-temperature fit to the Thermal Drift Model. Top inset: scaling plot of the Ramped Creep Model. The thin black line is a guide to the eyes and shows the expected Ramped Creep scaling.*

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