

Low-Speed Atomistic Simulation of Stick–Slip Friction using Parallel Replica Dynamics

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Received: 2 April 2009 / Accepted: 14 May 2009 / Published online: 2 June 2009
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Abstract Atomic stick–slip friction has been predicted by molecular dynamics simulation and observed in experiments. However, direct quantitative comparison of the two has thus far not been possible because of the large difference between scanning velocities accessible to simulations and experiments. In general, the slowest sliding speeds in MD simulations are at least five orders of magnitude larger than the upper limit available to experimentalists. To take a step toward bridging this gap, we have applied parallel replica dynamics, an accelerated molecular dynamics method, to the simulation of atomic stick–slip. The method allows molecular simulations to run parallel in time in order to extend their duration, thereby enabling lower scanning velocities. We show here that this method is able to predict atomic stick–slip friction accurately and efficiently at scanning speeds several orders of magnitude slower than standard molecular dynamics simulations. The accuracy and usefulness of this method is illustrated by correct prediction of the logarithmic dependence of friction on velocity.

Keywords Nanotribology · Stick–slip · Friction mechanisms

1 Introduction

Friction on the atomic scale has been at the forefront of scientific interest in recent years due to the miniaturization of mechanical components as well as to experimental and computational advances that have enabled measurements with atomic-scale resolution. Of particular focus has been atomic stick–slip. Atomic stick–slip describes the phenomenon of two contacting surfaces that do not slide smoothly over each other when subjected to an external force; instead the atomic features of the surfaces cause them to repeatedly stick together, and then slip. This results in a non-steady friction trace that is difficult to predict and can lead to large, fluctuating energy dissipation. Besides being fundamentally interesting, the ability to understand and predict this frictional behavior is critical to the commercial success of small-scale tribological components.

Atomic-scale friction can be measured and characterized experimentally using an atomic force microscope (AFM). Modeling and simulation can supplement these results by providing a detailed atomistic picture and mechanistic interpretations of the observations. Therefore, it is beneficial to use experiment and modeling in conjunction to investigate atomic scale friction. However, thus far direct quantitative comparison has been difficult or impossible because of a significant time-scale gap that results from the large difference between scanning velocities accessible to simulations and experiments.

On one hand, experimental studies of atomic stick–slip friction typically use scan speeds in the range of 10^{-9} – 10^{-6} m/s [1, 2]. Lower velocities are often not feasible due to unsuitably large amounts of distortion from thermal drift, and higher velocities are prevented by excitation of mechanical resonances in the scanning apparatus. On the other hand, recent molecular dynamics (MD) simulation

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studies on this topic that take advantage of powerful computational resources report velocities in the range of 0.22–800 m/s [3–6]. This range is characteristic of MD simulations for which the lower velocity limit is typically at least five orders of magnitude larger than the upper limit available to experimentalists.

The factor that limits typical MD simulations to relatively high sliding speeds is actually the length of the simulation. MD models atomic movement and interactions. To do so accurately, it must capture the thermal motion of individual atoms, the timescale of which is on the order of picoseconds. This requires that the time step of the simulation be on the femtosecond scale, which limits the total duration of the simulation to nanoseconds. However, many physical phenomena that can be observed experimentally, including atomic stick–slip under realistic driving forces, occur over much longer times. The usual solution to the problem is simply to drive harder (e.g., increase the speed and/or the temperature), which can be expected to corrupt the results. Long timescale information can also be inferred from simplified dynamics in some specific limits (e.g., $T \rightarrow 0$ effective potential energy surfaces [7]). However, a description of nanoscale friction in terms of activated stick–slip dynamics will, in general, require full atomistic models.

We have implemented parallel replica dynamics (ParRep) [8], an accelerated MD approach for infrequent-event systems, to reduce the discrepancy between experimental and simulated atomic stick–slip velocities. The ParRep method is based on the assumptions that the system being modeled advances to new configurations via infrequent events and that there are well-defined instantaneous rate constants for these events that are independent of the driving rate [9]. The method distributes the simulation time across multiple processors and therefore adequately samples the various possible state-to-state pathways accessible to the system, as would a standard, single-processor MD simulation run for a very long time. The molecular simulations are run parallel in time in order to extend their total duration, thereby enabling lower scan speeds. In this article, we briefly present the method, and then some illustrative results indicating that this approach can be used to complement experimental work with molecular simulations run at consistent, physically representative, low speeds.

2 Method

The following is a brief summary of the ParRep implementation procedure. The key steps of the process are illustrated in Fig. 1. First, the current configuration of the system is replicated on multiple processors and minimization is performed to generate a reference configuration

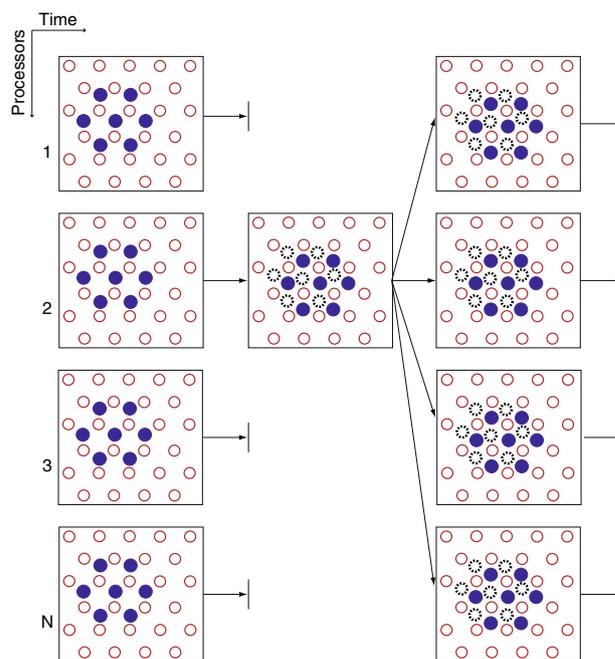


Fig. 1 Heuristic representation of the parallel replica dynamics (ParRep) method in which the atomic stick–slip simulation is run parallel in time across multiple processors. Circles represent atomic positions: red—substrate, solid blue—tip at current time, dashed line—tip at previous time. Note that this illustration does not include the critical ParRep phases of minimization, dephasing, and decorrelation, which are described in the text. (Color figure online)

for transition checks. On each processor, atomic momenta are randomized and dynamics are carried out for a short period of time in order to eliminate correlations between replicas. Once this step is completed, standard MD is carried out on each replica. During MD, each processor tracks the total shear force and also monitors for a transition event. When a processor detects a transition in its replica (which could correspond either to a slip or to some other topological change such as an adatom diffusion event), all other processors are stopped. The simulation clock is then advanced by the sum of the accumulated MD times on each replica. On the processor where the transition occurred, MD is continued slightly longer (in this work we use 2 ps) to allow for additional, correlated, transitions to occur. The final state in which the system settles is then broadcasted to all other replicas and the process is repeated. One can show that the proper statistics of transition times is recovered if each replica is driven N_{rep} times faster than the targeted physical driving rate, where N_{rep} is the number of replicas. A detailed description of these steps and the rationale behind selection of the various parameters of the method are given in the original articles [8, 9].

We applied this method to the case of atomic stick–slip; a snapshot of the simulation cell is shown in Fig. 2. All atoms are copper and modeled by an Embedded Atom Model potential fitted by Mishin et al. [10]. Regions

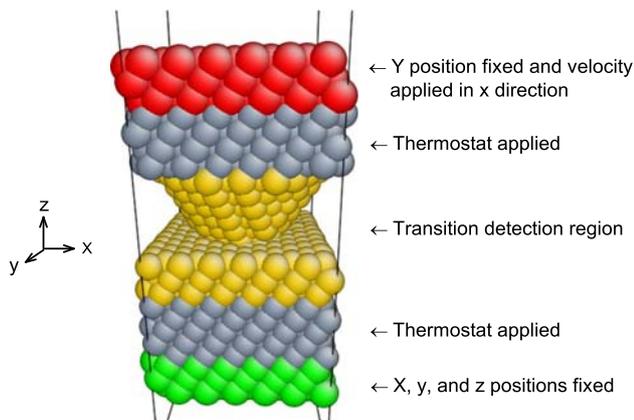


Fig. 2 Snapshot of the simulation cell illustrating the different atomic regions that are treated differently in the model in order to most correctly capture the behavior of the corresponding physical system. (Color figure online)

subjected to separate treatment in the molecular model are differentiated by color in Fig. 2. The system is driven (i.e., AFM tip moved over the substrate) by applying a velocity to the atoms in the uppermost region (red) of the tip. Except for this imposed displacement, the atoms in this region are restricted from any movement in the x - y plane. However, they are allowed complete freedom of movement in the direction normal to the substrate surface (z -direction). The bottommost atoms of the system (green) are fixed in all directions to provide structure to the simulation cell. There are no positional constraints on any other atoms in either the tip or substrate. Temperature is controlled in the simulation by applying a Langevin thermostat [11] to the atoms in the middle regions (grey) of the model tip and substrate with a target temperature of 200 K. The thermostat is intentionally not applied to the atoms near the interface (yellow) to minimize the effect that it might have on the stick-slip behavior.

A critical part of the ParRep implementation is to reliably identify transitions that occur during the simulation. In the present framework, transitions are associated with topological changes in the relaxed state of the system. Specifically, if, after minimization of the energy of the system (at a fixed x position of the tip), one or more atomic bond lengths differ by more than 20% from their previous reference length, a transition is declared. In the following, we assume that all atoms for which the equilibrium distance is less than 0.3 nm are bonded. This transition detection method makes our implementation completely general: we do not assume that the only relevant transitions are concerted slips of the contact but rather explicitly consider all topological changes of the system, be it diffusive jumps of adatoms on the tip or more complex slip mechanisms mediated, for example, by dislocation propagation.

3 Results

To extract a friction trace from the ParRep simulation, we average the total force on the fixed substrate atoms (green atoms in Fig. 2) over the number of processors. Note that averaging over processors in ParRep is equivalent to averaging over a block of time, as is done in standard MD simulations.

For this study, we ran the simulation parallel in time across up to 256 processors. This allowed us to obtain repeatable friction data at velocities as low as 0.001 m/s in approximately 72 h using the ParRep code. An example of typical results is shown in Fig. 3. Using standard MD, an equivalent simulation would require more than a year.

The representative simulation-predicted friction trace shown in Fig. 3 clearly exhibits the “saw-tooth” pattern characteristic of simulated atomic stick-slip. The sudden force drop-offs correspond to concerted slips at the interface; as expected given the small contact size, we did not observe dislocation-mediated slips. The long and short segments of the friction trace correspond to face-centered-cubic (fcc) and hexagonal-close-packed (hcp) registry between tip and substrate, respectively. Given that the tip was initially in fcc registry with the substrate and that the effective lateral stiffness of such a small tip is very large, the hcp registry is energetically disfavored here; hence it makes a small contribution to the friction force trace. The regular nature of the friction trace indicates that the shape of the tip does not significantly vary with time. Given its minimal surface area, slip along the (111) contact plane is energetically favored over slip along other (111)

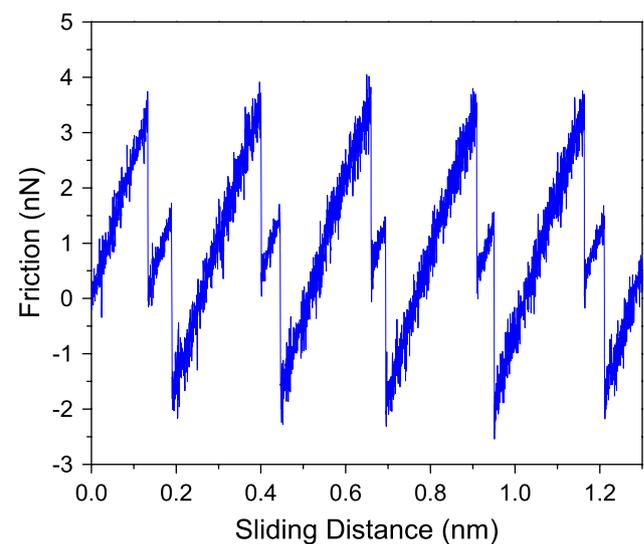


Fig. 3 Friction force in the sliding direction as a function of the position of the tip, predicted by the ParRep simulation at a velocity of 0.005 m/s

planes in the tip, the occurrence of which would modify the tip and contact geometry. This indicates our system is relatively insensitive to adhesion. This is in contrast to systems with larger interfaces for which the relative magnitude of slip barriers along different planes are expected to be much smaller, leading to stronger adhesion and shearing of the tip, a phenomenon we observe in regular MD simulations of much larger Cu–Cu systems.

Characteristics of atomic stick–slip are often investigated in terms of the mean friction force. However, there is some error associated with using ParRep for mean friction force calculations due to the fact that transition checks only occur at set intervals (in this case 1 ps). That is, when a slip event occurs on a given replica, it will not be detected until the next transition check. During the time between an actual slip event and when it is detected, the other replicas will continue running and erroneously contributing to the total force. We introduce a correction factor to address this issue. The mean force is effectively the integral of the force over the periodicity of the stick–slip. We assume that, on average, a transition occurs one half of the transition check interval before it is detected. Then the “extra” force is the distance each replica moves after a slip event (velocity \times number of replicas \times one-half the transition check time interval) multiplied by the change in force during a slip event. This approximation provides a known factor that can be subtracted from the measured ParRep mean force to correct for the discrete interval of time between transition checks. The correction factor is velocity dependent and contributes between 0.5% and 5% to the mean force for speeds of 0.001–0.1 m/s, respectively. This is consistent with the observation that the tip will travel further in a given time at faster speeds.

As mentioned previously, the ParRep method is based on the assumption that the system is quasi-static during the “stick” phase of the process. If this assumption is violated, which can occur at large velocities, the method will yield error observable as variation in the mean force with the number of processors on which ParRep is run. It is expected, and we observe (see Fig. 4), that the error will decrease as the velocity is decreased. As a reference point, we have also shown on Fig. 4 the error due to typical MD simulation noise (estimated from four independent simulations run on the same number of replicas). It can be observed that the error decreases with decreasing velocity and is less than the simulation noise once the velocity is slower than about 0.04 m/s (corresponding to about 4 m/s on each replica). This indicates that, while it may not be possible to eliminate the error completely, its effect is negligible for the small velocity cases that the ParRep simulation is intended to address.

An extremely important characteristic of atomic stick–slip friction that has been studied often both experimentally

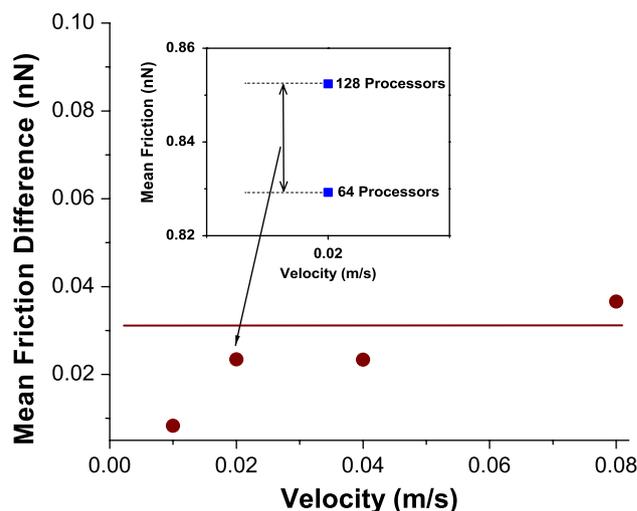


Fig. 4 Variation in mean friction predicted by ParRep when run on 128 replicas vs. 64 replicas (circles and inset) and expected due to typical MD simulation noise (solid line)

and via-simulation is its velocity dependence. A logarithmic relationship has been observed experimentally and supported theoretically [12]. However, MD simulation-predicted relationships have been inconsistent both in whether friction increases or decreases with velocity and the rate of that increase or decrease (see for example [4, 13, 14]).

For the Cu (111) system we employ, two highly cited papers have addressed the question of velocity dependence: the experimental work by Bennewitz et al. [1] and the MD study of Sorensen et al. [13]. Bennewitz et al. observed a logarithmic increase of friction force with scan velocity (Fig. 5 of that publication) while Sorensen et al. predicted that friction would ultimately decrease at higher velocities (Fig. 10 of that publication). Although the Sorensen et al. work was ground-breaking and helped explain a great deal about the physical mechanisms underlying atomic stick–slip, it did not correctly predict the experimental velocity dependence later observed. This is most likely predominately due to the seven orders of magnitude difference between the simulation and experimental velocities.

Using ParRep, we are able to model velocities three orders of magnitude closer to those reported by Bennewitz et al. We therefore repeat the velocity dependence analysis to determine if this impacts the resultant friction trend. In Fig. 5, we report the mean friction force at velocities from 0.001 to 0.1 m/s predicted by the ParRep simulations. This figure clearly illustrates the logarithmic dependence of friction on velocity. Driven ParRep has been used before to predict this trend in a study of grain boundary shearing [15].

The physical mechanism underlying the logarithmic velocity dependence of friction has been described by analytical models. Gnecco et al. [2] introduced a modified Tomlinson model based on reaction rate theory to show

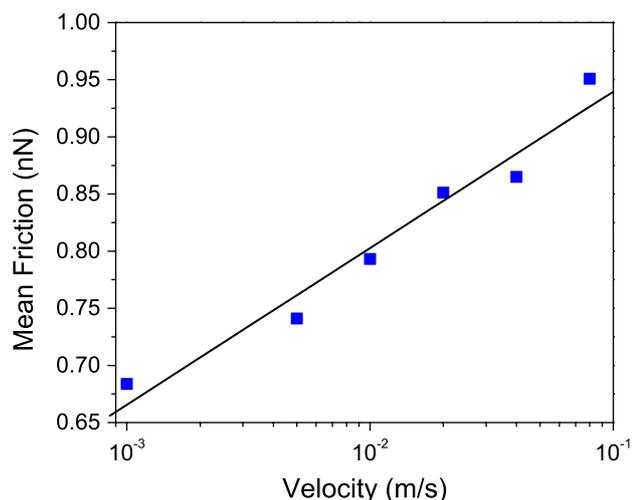


Fig. 5 Mean friction force as a function of velocity: *symbols* represent ParRep simulation data and the *solid line* is a logarithmic fit to that data

that the velocity dependence is due to thermal activation of the slip events. This concept of slip as an activated process is also part of the theoretical basis of the ParRep simulation method, which further supports our use of this method to describe atomic stick–slip.

We present here a brief overview of the modified Tomlinson model. Consider the case of a tip dragged at velocity v on the surface. During the stick phase, the coupled tip–substrate system is vibrating in a potential energy well. In order to slip, the system must overcome a potential energy barrier. That barrier ΔE , varies as a function of position of the tip, and hence as a function of time. In the harmonic approximation of the transition state theory, the instantaneous rate of slip is given by:

$$k(t) = v \exp\left(-\frac{\Delta E(t)}{k_B T}\right), \quad (1)$$

where v is the vibrational prefactor, which for simplicity, is taken to be independent of time. The probability flux of slipping at time t is thus simply given by:

$$\frac{dp(t)}{dt} = -k(t)p(t), \quad (2)$$

where $p(t)$ is the probability that the system has not yet slipped by time t . Changing variables from time to lateral force F_L , one gets:

$$\frac{dp(F_L)}{dF_L} = -k(F_L) \left(\frac{dF_L}{dt}\right)^{-1} p(F_L). \quad (3)$$

Since the force is seen to be linear in displacement (c.f., Fig. 3), we obtain $dF_L/dt = k_{\text{eff}}v$, where k_{eff} is the effective stiffness of the tip and v is the velocity. Further, in the vicinity of the critical force F_L^* at which slip occurs

spontaneously, the barrier ΔE can also be assumed to be of the form:

$$\Delta E(F_L) \simeq \lambda(F_L^* - F_L)^\alpha. \quad (4)$$

Although later articles reported α to be $3/2$ [15–17], the original Bennewitz et al. article uses $\alpha = 1$ to fit their experimental data for copper. To facilitate comparison and for simplicity, we also assume $\alpha = 1$. Using these assumptions and solving for the force F_L^m which maximizes the slip probability flux, we finally get:

$$F_L^m = F_L^* + \frac{k_B T}{\lambda} \ln\left(\frac{vk_{\text{eff}}}{vk_B T}\right) \quad (5)$$

The modified Tomlinson model thus predicts that the maximal friction force (and hence the mean force) vary logarithmically with the scanning velocity v .

We fit our ParRep data to the logarithmic relationship and find that the velocity dependence parameter is $\lambda = 0.29$ eV/nN. This is very reasonable in comparison to the parameter we infer from Bennewitz et al. [1] experimental data for a nm-scale copper tip on Cu (111), $\lambda = 0.2$ eV/nN. Even though our tip size and shape are almost certainly not the same as in the experiment, this good agreement between the simulated and experimental λ is meaningful. If the tip–surface contact area is small enough that dislocation mechanisms are not important, such that it is a good approximation that all tip atoms travel in perfect concert to the new position when a slip event occurs (which is what we observed here), then we expect that the energy barrier ΔE and the critical force F_L^* are each proportional to the number of atoms in the contact area. Then λ , which is the ratio of these two quantities, will be independent of the number of atoms in the contact. So if our interatomic potential for Cu is accurate enough and our driving rate slow enough, we should be able to predict an accurate value for λ and therefore the dependence of stick–slip friction on velocity.

4 Conclusion

We have introduced the use of an accelerated MD method, ParRep, to simulate atomic stick–slip in friction-force microscopy. This approach has enabled us to access velocities at least two orders of magnitude slower than standard MD simulation, thereby bringing us closer to the scan speeds of the experiments we are modeling. We have shown that the ParRep method predicts atomic stick–slip friction both accurately and efficiently and we anticipate that this method can be used going forward to bridge the gap between experiment and simulation. Experiments and simulations, run at consistent sliding speeds, can be used together to understand and ultimately predict the unique phenomenon of atomic stick–slip friction.

Acknowledgements We are grateful for the contributions of Drs. Robert Carpick, Yuri Mishin, and Vladimir Ivanov and to the National Science Foundation for its support via award CMMI-0758604. Work at Los Alamos National Laboratory (LANL) was supported by the United States Department of Energy (U.S. DOE) Office of Basic Energy Sciences, Materials Sciences and Engineering Division, and by the LANL Laboratory Directed Research and Development Program. LANL is operated by Los Alamos National Security, LLC, for the National Nuclear Security Administration of the U.S. DOE under Contract No. DE-AC52-06NA25396.

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