Parameter-free dissipation in simulated sliding friction

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Nonequilibrium molecular-dynamics simulations, of crucial importance in sliding friction, are hampered by arbitrariness and uncertainties in the way Joule heat is removed. We implement in a realistic frictional simulation a parameter-free, non-Markovian, stochastic dynamics, which, as expected from theory, absorbs Joule heat precisely as a semi-infinite harmonic substrate would. Simulating stick-slip friction of a slider over a two-dimensional Lennard-Jones solid, we compare our virtually exact frictional results with approximate ones from commonly adopted empirical dissipation schemes. While the latter are generally in serious error, we show that the exact results can be closely reproduced by a viscous Langevin dissipation at the boundary layer, once the backreflected frictional energy is variationally optimized.

The role of molecular-dynamics (MD) simulation in the theory of sliding friction and nanofriction can hardly be overestimated.[1,2] In a typical simulation, a slider is driven by an external force over a simulated solid substrate whose atoms, interacting through realistically chosen interatomic forces, vibrate and move according to Newton's law. Naturally, in order to attain a frictional steady state the Joule heat must be removed. Unfortunately, a realistic energy dissipation is generally impossible to simulate reliably, owing to size (and time) limitations. In a small-size simulation cell, the phonons generated at the sliding interface are backreflected by the cell boundaries, rather than propagated away to properly disperse the Joule heat. The ensuing problem is a spuriously accumulating phonon population in the slider-substrate interface region. The empirical introduction in the equations of motion of *ad hoc* Langevin viscous damping terms $-m\gamma_i \dot{q}_i$ (with m and $\dot{q}_i$ the mass and the velocity of the ith particle) and of an associated random noise, corresponding to some "thermostat" temperature T,[3] represents the handiest and commonest solution. However, both this procedure and the choice of thermostat and damping parameters γ_i are vastly arbitrary. The problem is not just one of principle. Dry friction generally involves stick slip,[1] and each slip generates a burst of phonons of very specific nature and composition. In realistic simulations, the overall effect on frictional dynamics of the partial backreflection of this burst must be minimized. Unless dealt with, backreflection will cause the simulated steady state and friction coefficient to depend on unphysical damping parameters. In the Prandtl-Tomlinson model,[4] for instance, damping is known to modify the kinetics and the friction, both in the stick-slip regime (including the multiple slips seen in atomic force microscopy[5]) and in the smooth sliding state. To a lesser or larger extent, this lamentable state of affairs is common to all MD frictional simulations.

Pursuing a viable solution, one wishes to modify the equations of motion inside a relatively small simulation cell so that they will reproduce the frictional dynamics of a much larger system, once the remaining variables are integrated out. Integrating out degrees of freedom is a classic problem,

largely analyzed in literature.[3,6–9] In the context of MD simulation, Green's function methods were formulated for quasi-static mechanical contacts;[10] approaches based on a discrete-continuum matching have also been discussed.[11,12] Among others, a formally exact dissipation formulation was given early on by Rubin.[6] Integrating out $N-1$ atoms in a linear harmonic chain yields a non-Markovian Langevin-type equation of motion of a single atom of interest. Extensions of this method were applied to a variety of problems, including the relaxation of impurity molecules in solids,[13,14] and atom-surface scattering.[15,16] Detailed formulations suitable for MD simulations were given by Li *et al.*[7] and by Kantorovich.[8,9,17] The idea to variationally minimize backreflection of individual phonons has also been put forward.[7] However, this body of theory has not yet found its way into realistic sliding friction MD simulations so that neither the importance of backreflection errors in dry friction nor a realistic way to get rid of them have really been demonstrated. Here we describe how both goals are achieved, in a simulated two-dimensional (2D) Lennard-Jones (LJ) system that exhibits a realistic stick slip. We find that exact stick-slip friction is indeed different from that of empirical damping schemes. However, the backreflected energy can be variationally minimized—although differently from Li *et al.*—the resulting approximate dynamics now reproducing quite accurately the exact benchmark stick-slip friction.

We focus on a model sliding system made of a semi-infinite two-dimensional crystalline substrate with N_z monatomic layers each of N_x atoms, and of a slider made of a single chain of N_x' atoms. All atoms interact via a LJ potential (cutoff for simplicity to first neighbors). The slider is pressed against the substrate by a normal "load" force F_0, and is driven along x (parallel to the substrate) through a spring of strength k, whose end is pulled at constant velocity v_0. Following similar earlier formulations,[7–9] the ideal infinitely thick substrate is divided as in Fig. 1(a), into three regions: (i) a live slab comprising N_z-1 atomic layers whose motion is fully simulated by Newton's equations; (ii) the dissipative boundary (N_zth) layer, whose motion includes the effective non-Markovian Langevin terms; (iii) the remaining
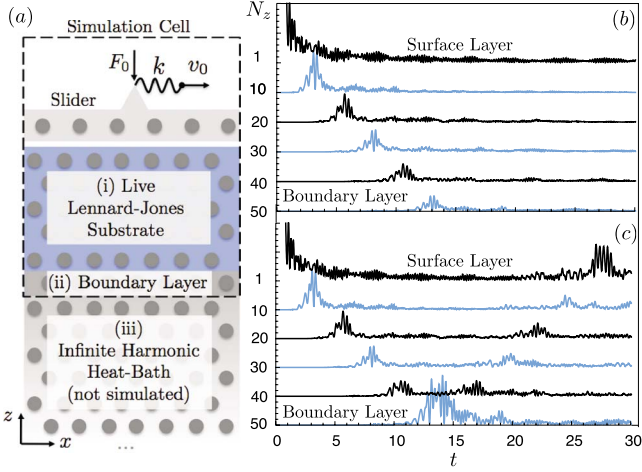
FIG. 1. (Color online) (a) Sketch of the simulated system. (b) and (c) Layer-averaged kinetic-energy time evolution of a surface-injected phonon burst. Phonon backreflection is strong without dissipation at the boundary layer (c) but accurately canceled once the correct dissipative kernels are included (b).

semi-infinite solid acting as a phonon absorber, or heat bath, whose degrees of freedom are integrated out, providing the source of effective damping terms in (ii). In our practical implementation, we substitute the full LJ potential within region (iii) and between (ii) and (iii) with its harmonic approximation. This approximation, necessary in order to get an analytical form for the effective forces on the boundary atoms, is all the more accurate the weaker the intensity of frictional phonons. In practice, for crystal substrates above quantum freezing and below the Debye temperature, this level of accuracy can always be attained by a sufficient thickness $N_z - 1$ of the live simulation cell (i). The harmonic heat bath (iii) is decoupled by diagonalizing the dynamical matrix $D_{\mu\nu}^{kl}$ (k, l denoting atoms), obtaining eigenvalues ω_k^2 and eigenvectors $\boldsymbol{\lambda}_k$. The equations of motion for the boundary layer atoms are[7,8]

$$m\ddot{q}_\mu^i(t) = -\frac{\partial U_{sub}}{\partial q_\mu^i} - m\sum_{j,\nu}\int_0^t ds K_{\mu\nu}^{ij}(t-s)\dot{q}_\nu^j(s) + R_\mu^i(t)$$
$$+ \sum_{j,\nu} q_\nu^j(t)[K_{\mu\nu}^{ij}(0) - D_{\mu\nu}^{ij}], \qquad (1)$$

where i and j denote boundary layer atoms, μ and ν indicate x/z components, U_{sub} is the LJ interaction between the ith boundary atom and those inside the simulation cell (i). The second term is non-Markovian and nonconservative, introducing an effective damping proportional to the velocity of the jth atom, through a time convolution with the memory kernel functions $K_{\mu\nu}^{ij}(t)$. Standard kernels are built from the harmonic eigenvalues and eigenvectors of the heat-bath dynamical matrix and from the coupling vectors $\boldsymbol{\phi}_\mu^i$ containing the harmonic coupling constants $D_{\mu\mu}^{ik}$ and $D_{\mu\nu}^{ik}$ of the ith atom of region (ii) with the kth heat-bath atom $K_{\mu\nu}^{ij}(t)$ $= \Sigma_k[(\boldsymbol{\lambda}_k \cdot \boldsymbol{\phi}_\mu^i)(\boldsymbol{\phi}_\nu^j \cdot \boldsymbol{\lambda}_k)/\omega_k^2]\cos(\omega_k t)$. They oscillate and decay with time, with power-law tails due to the bath acoustical phonon branches. Periodic boundary conditions along the x

direction guarantee translational invariance so that $K_{\mu\nu}^{ij}(t)$ is a function of $|i-j|$ only. As kernels inherit their symmetry properties from those of the heat-bath dynamical matrix, one can show that $K_{\mu\nu}^{ij}(t) = -K_{\nu\mu}^{ij}(t)$ and $K_{\mu\nu}^{ij}(t) = K_{\mu\nu}^{ji}(t)$. As $|i-j|$ grows, $|K_{\mu\nu}^{ij}(t)|$ decrease, but again not exponentially, so that correlations must be included up to large distance.[7] By cutting kernels off at time τ_c, one could limit the time integrals in Eq. (1), which need to be calculated at each time step. By increasing both τ_c and the live simulation cell size N_z, one refines the dissipation scheme accuracy as much as desired, although at increased computational cost. The third term in Eq. (1) is a Gaussian stochastic force present at nonzero temperature, responsible for the energy transfer between the heat bath (iii) and the live substrate (i), with $\langle R_\mu^i(t)\rangle = 0$ and $\langle R_\mu^i(t)R_\nu^j(t')\rangle = mk_B T K_{\mu\nu}^{ij}(t-t')$, where brackets denote ensemble average, k_B is Boltzmann's constant, and T the temperature. As is well known,[3] this relationship fulfils the fluctuation-dissipation theorem—the scheme represents a well-defined thermostat in contact with the simulation cell. The last term in Eq. (1) is the harmonic coupling between atoms ith and jth within the boundary layer, where the coupling constant $D_{\mu\nu}^{ij}$ is modified by $K_{\mu\nu}^{ij}(0)$.

As a first step before simulating friction, we implement the set of Eq. (1), along with ordinary Newton's equations governing the live cell in an MD simulation, to test phonon reflection. Starting with the equilibrated system ($N_z = 30$ close-packed layers and $N_x = 10$ atoms per layer) at temperature T, a test burst of phonons is introduced at the topmost layer and time evolved. Results in Fig. 1(b) show how a relatively thin $N_z = 30$ layer substrate (i+ii) is able to mimick, within the exact dissipation scheme, the full ideal semi-infinite system (i+ii+iii). Layer-resolved kinetic energies inside the simulated substrate show the group of phonons propagating below the surface. Upon reaching the boundary layer, the phonons are perfectly absorbed as if they propagated into the (integrated out) semi-infinite crystal (iii). For comparison, Fig. 1(c) shows the same phonons massively backreflected once the memory kernels are removed from the boundary layer.

We next simulate stick-slip sliding friction by driving a slider, here consisting of a LJ chain of $N_x' = 9$ atoms, over the same live substrate as above.[18] The instantaneous friction force is measured by the spring elongation $F(t) = k(x_{CM} - v_0 t)$, x_{CM} being the slider center of mass position. The sawtooth force profile typical of stick-slip friction is obtained [Fig. 2(a)]. The friction coefficient, obtained by averaging over a stationary sequence of stick slips is $\langle F\rangle/F_0 = 0.116 \pm 0.002$. The stick-slip pattern is irregular, with a periodicity similar but not exactly matching, a substrate lattice spacing. The highest spikes signal the forward jump of most slider atoms, smaller ones involve only about 2/3 of them. A measure of the distribution of the spike heights is the variance of $F(t)$, i.e., $\sigma = \frac{1}{\tau_s \langle F\rangle^2}\int_0^{\tau_s}[F(t) - \langle F\rangle]^2 dt$, where τ_s is the total simulation time. These simulations using the full Eq. (1), and the corresponding frictional results can be considered essentially exact, and represent our benchmark reference of stick-slip friction with a correct Joule heat removal. The numerical implementation of this standard scheme in a generic three-
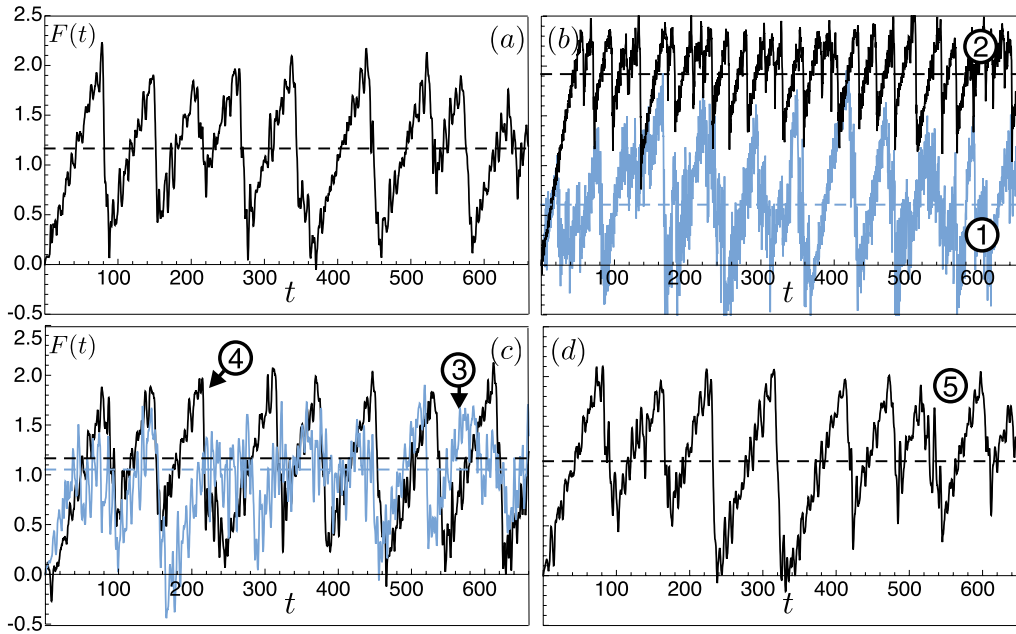
FIG. 2. (Color online) (a) Simulated friction force $F(t)$ for the "exact" non-Markovian dissipation scheme of Eq. (1), and (b)–(d) for empirical Langevin schemes with a $-\gamma\dot{q}$ damping applied to tip **L1**, whole substrate **L2** and bottom layer **L3**. The γ value is identified by numbers 1–5 in Fig. 3. Dashed lines: mean value $\langle F \rangle$.

dimensional (3D) case will, in principle, be possible but time consuming, and far from handy. Long-range non-Markovian correlations imply a general scaling as N_x^2 so that simulations for large-size 3D sliding systems may pose a practical challenge of parallel computing.

The virtually exact frictional dynamics just obtained can now be compared with empirical Langevin schemes commonly adopted in friction simulations, where a viscous damping $-\gamma\dot{q}$ is arbitrarily applied to the motion of some atoms in the system, for instance to slider atoms (**L1**, dashed line in Fig. 3) or to all substrate atoms (**L2**, dotted line) or to the boundary atomic layer only (**L3**, solid line). As anticipated, the stick-slip friction simulated with all these empirical schemes depends vastly on γ, and generally deviates systematically from the correct benchmark. The closest agreement is obtained in case **L3** where the viscous damping $-m\gamma\dot{q}_\mu^i(t)$ is applied to the ith boundary layer atom motion, $m\ddot{q}_\mu^i(t) = -\frac{\partial U}{\partial q_\mu^i} - m\gamma\dot{q}_\mu^i(t) + R_\mu^i(t)$, with the appropriate Gaussian stochastic force $R_i(t)$ with $\langle R_\mu^i(t) \rangle = 0$ and $\langle R_\mu^i(t)R_\nu^j(t') \rangle = 2mk_BT\gamma\delta_{\mu,\nu}\delta_{i,j}\delta(t-t')$. Since the parameter γ is adjustable, we may seek to optimize it in order to approximate the exact results. Li *et al.*[7] considered variationally minimizing a group-velocity weighted phonon reflection. In the actual friction simulation, the steady-state internal energy increase $W = \langle E \rangle - \langle E_0 \rangle$ over the equilibrium value E_0 is easily computable. Our proposed scheme is to variationally minimize W, a quantity generally positive and proportional to the Joule frictional heat. In absence of backreflection, W is minimal whereas partial energy reflection at the boundary will cause Joule heat to artificially accumulate in the slab and increase W so that for any arbitrary dissipation scheme $W/W_{exact} > 1$. As a function of γ, a variational minimum of W/W_{exact} occurs because backreflection of frictional phonons is large *both* when the boundary layer damping γ is too small
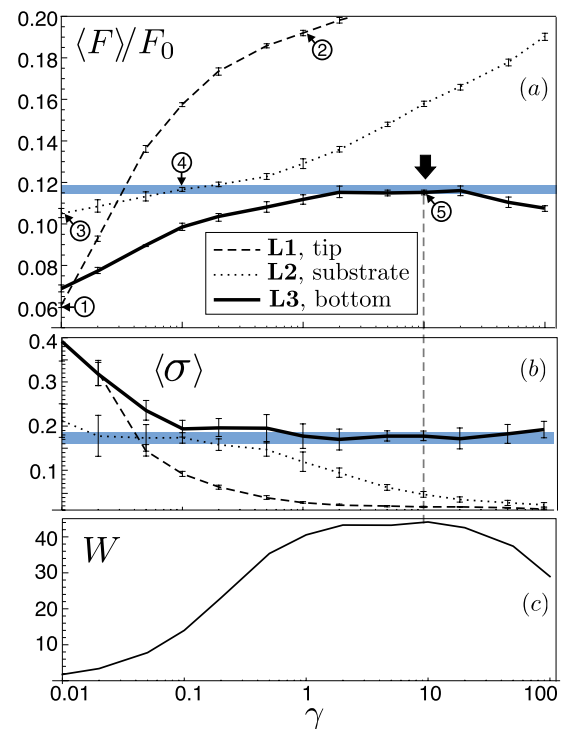

FIG. 3. (Color online) (a) and (b) Friction coefficient $\langle F \rangle/F_0$ and variance $\langle \sigma \rangle$ as a function of a viscous damping γ for different empirical Langevin dissipation schemes, compared with exact non-Markovian values (thick horizontal stripes). (c) Average internal energy W per substrate atom for the Markovian thermostat **L3** divided by the internal energy W_{exact} per substrate atom for the non-Markovian scheme (1). The exact and empirical frictional behavior nearly coincide when the boundary layer parameter γ is such as to variationally minimize the relative total backreflected energy W/W_{exact}.
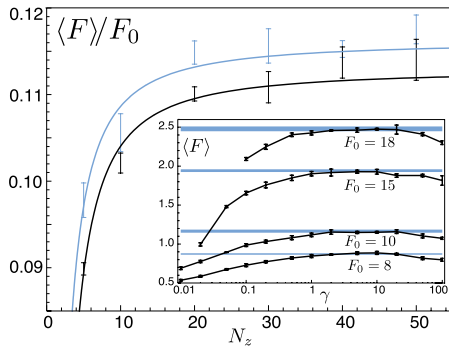
FIG. 4. (Color online) Friction coefficient as a function of the simulated cell thickness N_z for different driving velocities $v_0=0.01$ (blue, upper curve) and $v_0=0.02$ (black, lower curve) obtained with the non-Markovian dissipation scheme. The inset reports the average friction force for different loads F_0 obtained with the non-Markovian approach (thick horizontal stripes) and the comparison with the Markovian Langevin scheme **L3** at different γ (black curves).

and too large. This ratio is shown in Fig. 3(c). The agreement between the exact frictional results, where no phonons are backreflected and the variational one at $\gamma=10$ is excellent. (We note incidentally that at the optimal γ the friction coefficient also peaks.) To confirm the variational result, we changed system parameters, including sliding velocity, and load. The variable load results of Fig. 4 (inset) show that the coincidence of optimal and exact friction is systematic. Fig-

ure 4 also shows a dependence of calculated friction on the thickness of the simulated substrate portion (i+ii), converging for sufficiently large N_z. The required thickness of a few tens of layers depends on velocity, and is reached once the substrate inertia grows large enough to stop interfering spuriously with the frictional dynamics.

In conclusion, we demonstrated frictional MD simulations with a non-Markovian dissipation, enacting the correct disposal of generated phonons, even in the relatively violent stick-slip case. Using the virtually exact frictional results so obtained as a reference, we benchmarked common empirical viscous dissipation schemes, finding them generally wanting. There exists, however, an optimal viscous damping γ_{opt} which, once applied to the cell boundary layer, yields results nearly indistinguishable from the exact ones. The optimal damping parameter variationally minimizes the total backreflected energy at the cell boundary, and can be identified even without any exact reference calculation. This optimal damping scheme, which unlike the exact one does not require a knowledge of the substrate vibrational properties, is a good candidate for adoption in future practical MD frictional simulations.

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[18] Sliding simulations are performed at temperature $k_BT=0.035$, roughly corresponding to $T/T_{melting}=0.06$ (LJ units used throughout). The load is $F_0=10$, the average sliding velocity $v_0=0.01$, and the spring constant $k=5$. Periodic boundary conditions are applied to substrate and slider along x. To favor sliding, the strength of the slider-substrate LJ interaction is reduced from 1 to 0.6. Equations of motion are integrated by a modified velocity-Verlet algorithm with a time step of $\Delta t=5\times10^{-3}$. Memory kernels are cutoff at $\tau_c=5\times10^3$ time steps, with good accuracy of the frictional pattern in all its aspects, including phonon dissipation, our main concern.