

Simple Model of Microscopic Rolling Friction

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Rolling friction at a microscopic scale is studied with the help of a simple two-dimensional model. Molecular dynamics simulations show that rolling of spherical lubricant molecules exists only for concentrations lower than the concentration of a close-packed layer. At concentrations higher than a critical one due to jamming of lubricant molecules the rolling of nearest neighboring molecules is hindered. An optimal concentration exists which provides the minimum of kinetic friction. Methods for avoiding jamming and increasing the range of operation of rolling mechanism of friction are discussed.

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The problem of friction between two substrates in a moving contact separated by a thin lubricant film is very important technologically, as well as very rich physically [1]. Typical lubricants provide a friction coefficient on the order of $\mu \geq 0.1$ in a smooth-sliding regime (the “tribological” friction coefficient is defined as $\mu = f/f_{\text{load}}$, where f is the driving force and f_{load} is the load). A very promising way may be the use of solid lubricants. In an ideal case of the contact of two rigid crystalline incommensurate surfaces, the static frictional force f_s is zero [2] (moreover, the same is true even for commensurate surfaces if they are not perfectly aligned). Similarly, if the lubricant film has a crystalline structure and is confined between two flat substrates with an ideal structure, the friction coefficient may be as low as $\mu \sim 10^{-3}$ – 10^{-2} or even lower [3].

On the other hand, it is known that coefficients of rolling friction are generally 10^2 to 10^3 times lower than those of sliding friction for corresponding materials. The main source of friction in rolling is the dissipation of energy involved in deformation of the objects. In this context the following intriguing question emerges [4]: may a similar mechanism work at a microscopic scale; i.e., may ball-shape molecules such as almost spherical C_{60} molecules (fullerenes) work as a “molecular bearing”? It could be very promising for realization in nano- and micromachines. Unfortunately, these anticipations have not been confirmed in experiments. The lowest friction coefficient between two C_{60} films was found to be of order $\mu \sim 0.15$ [5]. The aim of the present work is to clarify why experiments do not demonstrate the desired low rolling friction. We show that the microscopic rolling is hindered because of jamming of lubricant molecules.

A large scale molecular dynamics simulation of fullerenelike lubricant, where each molecule itself consists of 60 atoms, is extremely complicated, in particular, because intermolecular potentials are complicated and not yet well established [6,7]. For these reasons in the present work we use a simplified two-dimensional (2D) model which, nevertheless, allows us to catch all the important physics of the problem and leads to qualitatively correct results.

Model.—We use molecular dynamics simulations for a 2D system, where all atoms have the unit mass ($m = 1$) and interact via the 6–12 Lennard-Jones (LJ) potential $V_{\text{LJ}}(r) = V_0[(R_0/r)^{12} - 2(R_0/r)^6]$ with the amplitude $V_0 = 1$ and the equilibrium distance $R_0 = 1$ (this defines our dimensionless system of units). Each molecule is constructed of $L + 1$ atoms (see Fig. 1), namely, one central atom and L atoms on a circle of radius $R_m = R_0/2 \sin(\pi/L)$ so that the equilibrium distance between the circular atoms is R_0 . The circular atoms of a given molecule are coupled with the central atom, additional to the LJ potential, by stiff springs of the elastic constant K_m , $V_{\text{stab}}(r) = \frac{1}{2}K_m(r - R_{\text{stab}})^2$, where the distance $R_{\text{stab}} = R_m + (12V_0/K_m R_m)[(R_0/R_m)^6 - (R_0/R_m)^{12}]$ is chosen so that the total potential $V_{\text{LJ}}(r) + V_{\text{stab}}(r)$ has the minimum at $r = R_m$. Such a construction is typical in simulation of polymer molecules [8] and guaranties that the molecular shape is not destroyed during simulation.

Then, we put N lubricant molecules between the two (bottom and top) substrates. To reduce the number of parameters, we model each substrate by a closely packed monolayer constructed of the same molecules and pinned to the corresponding (top or bottom) substrate. In the 2D model, the closely packed monolayer corresponds to a chain of length M with the periodic boundary condition along the x direction as shown in Fig. 1. Thus, the lubricant and the substrates are deformable. The two outmost atoms

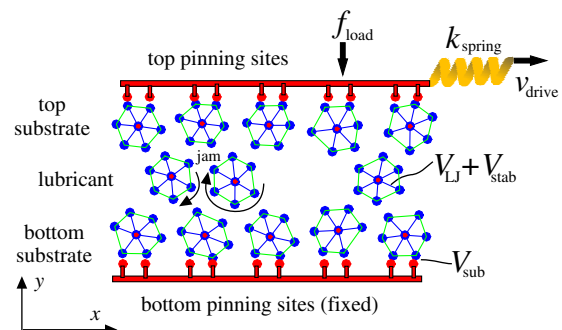


FIG. 1 (color online). The model.

of each substrate's molecule are coupled to the "pinning sites" (shown by circles attached to the substrates in Fig. 1) with the help of stiff springs of the elastic constant K_s , $V_{\text{sub}}(r) = \frac{1}{2}K_s r^2$. These pinning sites (PS's) themselves constitute a rigid lattice which models the removed part of semi-infinite substrate. The bottom PS lattice is fixed at $x = y = 0$, while the top one may move in two dimensions x and y . A load force f_{load} is applied to the top PS lattice (in what follows all forces are presented in units "force per one pinned molecule"). Also, the top PS lattice is connected by a weak spring of elastic constant k_{spring} with a base which moves with a constant velocity v_{drive} . The spring force, which corresponds to the frictional force, is monitored during simulation. For the elastic constants we choose $K_m = K_s = 100$ and $k_{\text{spring}} = 10^{-3}$. We checked that the molecular dynamics results remain qualitatively unchanged if these constants are changed at least 2 times to higher or lower values. Thus, our model is a 2D variant of a typical experimental device used in tribology. When the static friction force is nonzero, experiments always show a transition from stick-slip motion to smooth sliding with an increase of the driving velocity [9]. In our model, when the driving velocity increases, the system exhibits a transition from stick-slip to smooth motion (see Fig. 2), where slips (in the stick-slip regime, left panel of Fig. 2) as well as smooth motion (right panel of Fig. 2) correspond to rolling of lubricant molecules.

Because a 2D model cannot reproduce even qualitatively the true phonon spectrum of a 3D system, we use a Langevin equation of motion with Gaussian random force corresponding to a given temperature T , and the damping force $f_{\eta,x} = -m\eta(y)\dot{x} - m\eta(Y-y)(\dot{x} - \dot{X})$, where x, y are the atomic coordinates and X, Y are the coordinates of

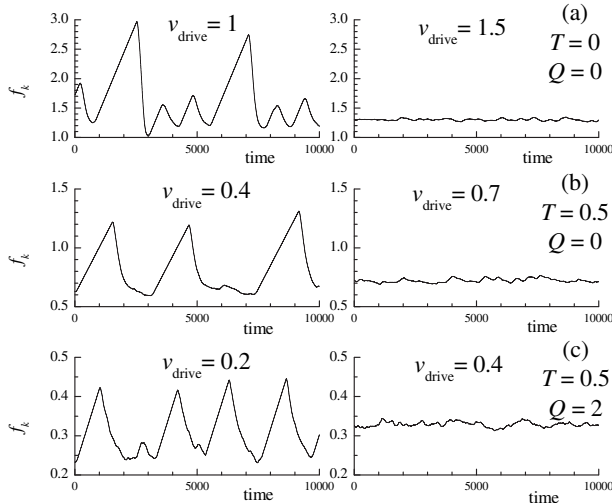


FIG. 2. Spring force versus time for two driving velocities: a lower velocity (left panel, stick-slip motion due to rolling/jamming mechanism) and a higher velocity (right panel, smooth-rolling regime). Driving velocities are given in the legends. (a) $T = 0$ and $Q = 0$, (b) $T = 0.5$ and $Q = 0$, (c) $T = 0.5$ and $Q = 2$. $\theta = 2/3$ ($N = 20$ and $M = 30$), $f_{\text{load}} = 2$.

the top PS lattice (the force $f_{\eta,y}$ is defined in the same way). The viscous damping coefficient decreases exponentially with the distance from the corresponding PS lattice, $\eta(y) = \eta_0[1 - \tanh(y/2R_m)]$, where we typically used $\eta_0 = 1$.

Below we present the simulation results only for $L = 6$, i.e., for hexagonal shape of the molecules.

Results.—First let us consider a single lubricant molecule between the substrates. As the molecule is moved over the bottom substrate from left to right, the leftmost bond is broken while that to the right remains intact and acts as a "pivot" over which the molecule rolls (the same type of molecular motion was predicted by *ab initio* calculations [7]). The pivoting of the molecule leads to a coupling of rotation and translation.

According to simulation results presented in Fig. 3, for loads $f_{\text{load}} \geq 0.5$ the Amontons law operates in the smooth-rolling regime. The dependence of the kinetic friction coefficient on the driving velocity can be fitted approximately by a linear law $\mu \approx 0.15 + 0.27v_{\text{drive}}$. Note that the rolling is not frictionless; i.e., the static friction is nonzero, $\mu_s \geq 0.15$. In order to rotate a lubricant molecule over the substrate, one has to break at least one interatomic bond. Let V'_0 be the total energy of these bonds ($V'_0 \sim V_0$). Thus, one has to apply the driving force $f'_0 \sim 2V'_0/R_0$ in order to start rolling, where the factor 2 is due to two substrates. Then, the Amontons law works owing to proportionality $V'_0 \propto f'_{\text{load}}$ (here f'_{load} is the local load force), which follows from the relationship $\partial V_{\text{LJ}}/\partial y \sim \partial V_{\text{LJ}}/\partial x$.

When the concentration of lubricant molecules is nonzero, $\theta \equiv N/M > 0$, the kinetic friction in the smooth-rolling regime depends on θ as shown in Fig. 4. The frictional force per substrate molecule may be estimated as $f_k \approx f'_0 N/M = f'_0 \theta$ and, therefore, it should grow with θ provided f'_0 does not depend on f'_{load} . This indeed is the case for high concentrations $\theta > \theta_{\text{opt}} \sim 0.3$. However, the

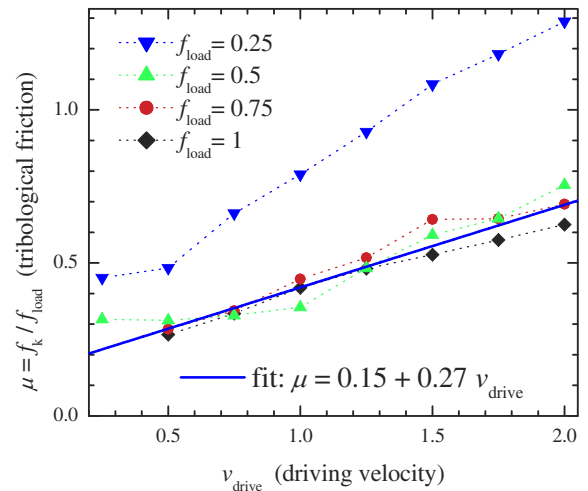


FIG. 3 (color online). The friction coefficient $\mu = f_k/f_{\text{load}}$ versus driving velocity for a single rolling lubricant molecule at different loads as given in the legend ($N = 1$ and $M = 12$).

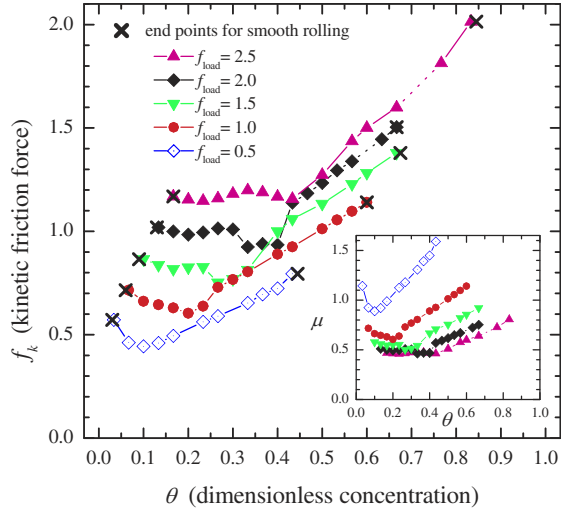


FIG. 4 (color online). Dependence of kinetic frictional force f_k in the smooth-rolling regime on the dimensionless concentration $\theta = N/M$ for different loads f_{load} as shown in the legend. Crosses indicate end points where smooth rolling is destroyed. Inset: tribological friction $\mu = f_k/f_{\text{load}}$ versus θ . $v_{\text{drive}} = 2.25$, $M = 30$, $T = Q = 0$.

total load is spread over all lubricant molecules, so that a local force per molecule is $f'_{\text{load}} = f_{\text{load}}M/N = f_{\text{load}}/\theta$. At low concentrations $\theta < \theta_{\text{opt}}$ the local load is so high that f'_0 strongly depends on f'_{load} , e.g., $f'_0 \propto f'_{\text{load}}$ or even stronger. This is why f_k does not depend on θ or even may decrease with θ at low concentrations. This leads to the existence of an optimal concentration $\theta_{\text{opt}} \sim 0.3$ which provides the minimal rolling friction.

At very low concentrations the smooth rolling is destroyed and changes to stick-slip or creep motion because of strong increase of the local load f'_{load} and, therefore, the local driving f'_0 . The range of concentrations where the smooth rolling exists is indicated by crosses in Fig. 4. The most important result of the present work is that the smooth rolling is also destroyed at high concentrations because of jamming of the lubricant. Jamming, or the “traffic-jam” effect in driven systems is well known (e.g., see [10–12] and references therein). In the case of rolling friction the jamming is, however, much more dramatic than in the case of sliding friction with conventional lubricants. When two nearest neighboring (NN) rolling molecules come in close contact, they hinder mutual rolling, because the two sides of colliding molecules roll in opposite directions as shown in Fig. 1. As a result both molecules stop rolling. Then the jam grows in size and totally destroys the smooth-rolling regime. Thus, the smooth rolling cannot exist at high concentrations of the lubricant, e.g., for the closely packed layer $\theta = 1$.

In order to extend the range of operation of the smooth-rolling regime, we have to avoid jams. According to Fig. 4, the simplest way to do so is to reduce the lubricant con-

centration, because jams emerge at concentrations higher than some critical value [10,11]. Another way is to use working temperatures higher than intermolecular interaction, when the rolling of NN molecules is not hindered. As can be seen from Fig. 2(b), the increase of temperature leads to a strong decrease of the critical velocity v_c of the transition from stick slip to smooth rolling. Figure 5(a) also demonstrates the decrease of kinetic friction when the temperature increases. Finally, a third way to avoid jams is to keep particles equidistant so that their collisions are suppressed [10]. For example, one may introduce an additional repulsion $V_{\text{rep}}(r) = Q^2/r$ between the central atoms of the molecules, which may correspond, e.g., to electrostatic repulsion of charged molecules. The dependence of the kinetic friction f_k on the amplitude of repulsion Q is presented in Fig. 5(b). Figure 2(c) also shows that the increase of Q leads to a strong decrease of the critical velocity v_c , thus extending the range of operation of the smooth-rolling regime. Note, however, that the repulsion of lubricant molecules may enhance escaping of the lubricant from the contact zone. Besides, when the bottom and top substrates are identical, the charge Q will be zero due to symmetry reasons.

The simulation results presented above were obtained within the 2D model, where the lubricant molecules move along the 1D “channel.” In a more realistic 3D tribological model the lubricant forms a 2D layer. One may expect that the jamming effects will be suppressed in the 2D driven system, because the molecules can go around the jams. However, the study of 2D driven models such as the discrete lattice-gas-like model [11] or the continuous Frenkel-Kontorova type model [12] has shown that the situation is just opposite: the mobility of 2D driven systems is typically lower than that of the 1D system because of spreading of jams in the direction perpendicular to the driving.

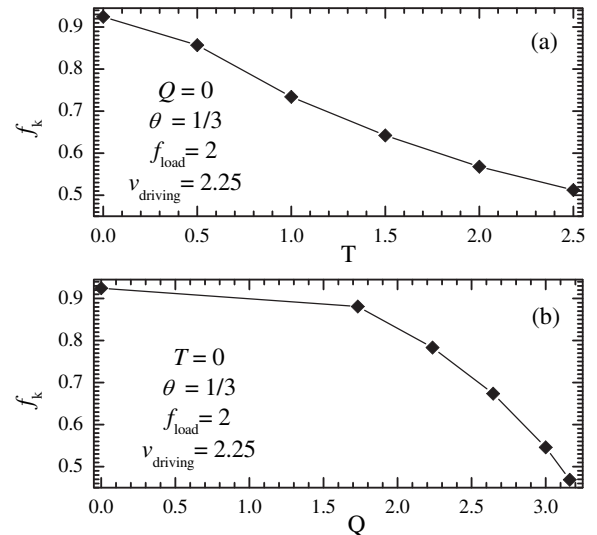


FIG. 5. Dependence of the kinetic friction force in the smooth-rolling regime (a) on temperature (at $Q = 0$) and (b) on Q (at $T = 0$). $v_{\text{drive}} = 2.25$, $\theta = 1/3$, $M = 30$, $f_{\text{load}} = 2$.

Discussion.—The simulation results of the present work are in qualitative agreement with known experimental and simulation results on fullerenes. The C_{60} molecules may form close-packed layers, e.g., on the graphite substrate [4,5]. A single C_{60} molecule confined between two solid substrates may begin to roll when a torque of order 1.0×10^{-19} N m is applied [13]. According to experimental results of Miura *et al.* [13], two layers of C_{60} molecules are stick slip but one (close-packed) monolayer may exhibit rolling.

The structure of C_{60} layers depends on temperature [14]. The C_{60} molecules take on a crystalline structure with spacial order at low temperatures, which undergoes the first-order orientational order-disorder phase transition at $T = T_m \approx 260$ K. In the high-temperature phase, $T > T_m$, the molecules exhibit free rotation so that all molecules are equivalent, while at $T < T_m$ the rotation is hindered and corresponds to jump reorientation between symmetrically equivalent orientations. This transition may give rise to an abrupt change in friction. For example, in the tip based experiment by Liang *et al.* [15], where the Si_3N_4 tip with an attached layer of C_{60} molecules was moved with a velocity $v \sim 0.1\text{--}2$ $\mu\text{m/s}$ over the C_{60} crystal, it was found that the friction could be described by a dependence $f_s \approx f_{s0} + \mu f_{\text{load}}$ with $\mu \approx 0.13$ and $f_{s0} \approx 2.1$ nN for $T > T_m$, but $f_{s0} \approx 3.5$ nN for $T < T_m$. These results were explained in the following way [15]: the C_{60} - C_{60} interaction energy has orientation-independent contribution ($\sim 1.5\text{--}1.7$ eV) and orientation-dependent contribution ($\sim 0.2\text{--}0.4$ eV). The latter contribution becomes unessential at $T > T_m$ which results in the abrupt decrease of friction.

One more way to avoid jams and improve the rolling of ball-shape molecules is to insert a “lubricant” *between* the fullerene molecules. For example, one may use fullerenes as additives to a conventional lubricant. However, the fullerenes should not form chemical bonds with the base lubricant, because otherwise the rolling of fullerenes will be hindered. Experiments by Rapoport *et al.* [16,17] showed that while the C_{60} fullerenes are ineffective as additives in oil lubricants, *inorganic* fullerenelike molecules of metal dichalcogenide MX_2 (where $M = \text{Mo}$ or W and $X = \text{S}$ or Se) can provide excellent tribological properties.

Finally, note that in our model the substrates and the lubricant are made of the same material. In such highly commensurate situations smooth sliding is impossible for the case of conventional lubricants, i.e., the lubricants made of nonrolling molecules. In such a system the friction is typically very large, $\mu \gtrsim 1$, and is accompanied by wear of the substrates. In the case of spherical lubricant molecules, as has been shown in the present work, smooth rolling with a much lower friction does exist. Therefore, we may predict that in the system where the substrates are more rigid than the lubricant and are incommensurate with

the latter, one has to expect a very low friction. However, the low friction may be achieved only for a lubricant concentration which is lower than some critical value, e.g., lower than the close-packed C_{60} layer. At the high concentration the rolling of NN molecules is hindered because of the jamming of lubricant molecules, and the smooth-rolling regime is destroyed. Besides, the increase of temperature or the introduction of an additional repulsion between the lubricant molecules may strongly improve the frictional properties of fullerenelike lubricants.

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