Transverse Thermal Depinning and Nonlinear Sliding Friction of an Adsorbed Monolayer

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We study the response of an adsorbed monolayer under a driving force as a model of sliding friction phenomena between two crystalline surfaces with a boundary lubrication layer. Using Langevin-dynamics simulation, we determine the nonlinear response in the direction transverse to a high symmetry direction along which the layer is already sliding. We find that below a finite transition temperature there exist a critical depinning force and hysteresis effects in the transverse response in the dynamical state when the adlayer is sliding smoothly along the longitudinal direction.

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Driven lattice systems interacting with pinning potentials have attracted growing interest recently due to the variety of possible nonequilibrium dynamic phases. This problem is relevant for many systems including adsorbed layers in tribology [1–8], charge-density waves [9], and vortex lattices [10–12] in type-II superconductors. In particular, in the boundary lubrication problem for interfaces [3], the macroscopic sliding friction is controlled by the response of the tightly bound lubricant layer between the surfaces to the external driving force. It has been shown that there are many dynamical phases, and the transitions between these phases can lead to such macroscopic behavior as the stick and slip motion. The essence of the physics in the boundary lubrication has been reasonably well understood with the study of a zeroth order model consisting of an overlayer of interacting atoms adsorbed in a periodic potential subject to an external driving force [5,8]. However, an interesting aspect of sliding friction which has not been addressed so far is how the nonlinear response of the overlayer is affected by the symmetry properties of the underlying periodic pinning. Initially, the overlayer can be driven to slide along a high symmetry direction of the surface. In this sliding state, the response to an additional force in the transverse direction is of interest. It should provide important information on the nature of the sliding state. In addition, the transverse force can also generate novel nonequilibrium dynamic phases of the overlayer. In this paper, we report the result from a study of the transverse sliding friction of a monolayer initially driven along the high symmetry direction of the periodic pinning potential. We find evidence of a transverse depinning transition at a zero temperature T_p. The nonequilibrium dynamic phase below T_p shows a critical depinning force in the transverse response even when the overlayer is in a state sliding smoothly along the longitudinal direction.

We consider a model of interacting adatoms in a periodic potential subject to an external driving force [3,5]. The dynamics is described by the Langevin equation

$$m\ddot{\mathbf{r}}_i + m\eta\dot{\mathbf{r}}_i = -\frac{\partial U}{\partial \mathbf{r}_i} - \frac{\partial V}{\partial \mathbf{r}_i} + \mathbf{f}_i + \mathbf{F}, \qquad (1)$$

where $\mathbf{r}_i$ is the adsorbate position, $U = \sum_i u(r_i)$ is the periodic substrate pinning potential, $V = \sum_{i \neq j} v(|\mathbf{r}_i - \mathbf{r}_j)$ is the interacting potential between adatoms, $\mathbf{F}$ is the uniform external force acting on each adatom, and $\mathbf{f}_i$ is an uncorrelated stochastic force, with zero average, and variance related to the microscopic friction parameter η, the mass of the particles m and the temperature T by the fluctuation dissipation relation $\langle f_i^\alpha(t) f_i^\alpha(t')\rangle = 2\eta m k T \delta(t - t')$, where α represents the vector components. We choose a periodic pinning potential with square symmetry

$$u(\mathbf{r}) = U_0[2 - \cos(2\pi x/a) - \cos(2\pi y/a)], \qquad (2)$$

where a is the lattice constant of the substrate, and an interacting Lennard-Jones pair potential

$$v(r_{ij}) = \epsilon[(r_0/r_{ij})^{12} - 2(r_0/r_{ij})^6], \qquad (3)$$

where ϵ is the well depth and r_0 is the particle separation at the minima in the pair potential. Periodic boundary conditions are used, with $r_0/a = 1.56$ and the adsorbate coverage set to $\theta = 1/2$. With this choice, the ground state of the overlayer is a commensurate pinned $c(2 \times 2)$ structure which has been extensively studied in connection with the boundary lubrication [3,5] problem and its longitudinal response to an applied force is well understood. Thus, this pinned structure provides a convenient starting point for the present study of transverse response. We vary the temperature T and the ratio ϵ/U_0 that is essentially a measure of the stiffness of the overlayer relative to the pinning potential. Dimensionless units are used where $a = 1$, $m = 1$, $U_0 = 1$ and we normalize the force by $2\pi U_0$ and velocities by $2\pi U_0/\eta$. We study the nonlinear response of the overlayer through dynamical simulations, using standard methods of Brownian molecular dynamics [13]. As in the previous work on the longitudinal response

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[5], small systems are used in order to allow for long equilibration times and reduce statistical errors. A system consisting of $L \times L$ substrate atoms with $L = 10-20$ was considered with the time variable discretized in units of $\delta t = (0.002-0.01)\tau$, where $\tau = (ma^2/U_0)^{1/2}$. Calculations were performed with typically $(1-4) \times 10^6$ time steps for each calculation of time-averaged quantities allowing equal time steps for equilibration.

As shown previously for an overlayer with the Lennard-Jones interacting potential [5] and also for a model with pure elastic interacting potential [8], the behavior of the drift velocity V_d as a function of F along the high symmetry direction x or y depends strongly on the initial equilibrium phase at $F = 0$. We first summarize the known behavior for the longitudinal response along the high symmetry x direction. When the overlayer at $F = 0$ is in an initial fluid or incommensurate state, then V_d will be nonzero for an arbitrarily small external force leading to a finite sliding friction $1/\bar{\eta}_L = V_d/F$ for arbitrarily small values of F. For our choice of parameters, the initial state has a commensurate $c(2 \times 2)$ structure below the melting temperature T_m. For this initial commensurate state, the drift velocity is essentially zero below a critical value of F, neglecting the contribution from creep motion due to thermal activation. The $F \times V_d$ characteristics show hysteresis behavior with unequal critical forces F_a and F_b for increasing or decreasing external force F_x, corresponding to the static and kinetic friction forces, respectively. This behavior is shown in Fig. 1 for a temperature $T = 0.2$ much below the melting transition temperature $T_m = 1.2$ of the commensurate overlayer at $F = 0$. In addition, at F_b there is a velocity gap V_b. For sliding velocities $V < V_b$, smooth sliding is not possible. As a consequence, the motion of the overlayer shows stick and slip behavior when driven by a spring moving with a velocity $V < V_b$, in agreement with experimental observations where smooth sliding is found only above a critical sliding velocity [1]. As first pointed out by Persson [5], for a sufficiently strong interacting system, the particle velocity distribution relative to the center of mass is Gaussian and one can define an effective temperature of the overlayer T^* [$kT^* = m(\langle v^2 \rangle - \langle v \rangle^2)/2$]. The irregular motion of the adatoms in the region $F_b < F_x < F_c$ leads to $T^* > T$, with the extra thermal dissipation, proportional to $T^* - T$, being needed to balance the heating effect of the external driving force to reach a steady state. The energy transfer to the overlayer is possible because the continuous translational symmetry is broken by the external pinning potential. In the limit $F \to \infty$, the pinning potential is negligible, and the overlayer slides rigidly with $T^* = T$. When the force is decreased, the sliding overlayer undergoes a dynamic melting transition into a sliding liquid phase at F_c, where T^* reaches a value above the melting transition temperature T_m. With a further decrease of the external force, the overlayer is finally pinned at the critical kinetic frictional force F_b to become the commensurate phase again.

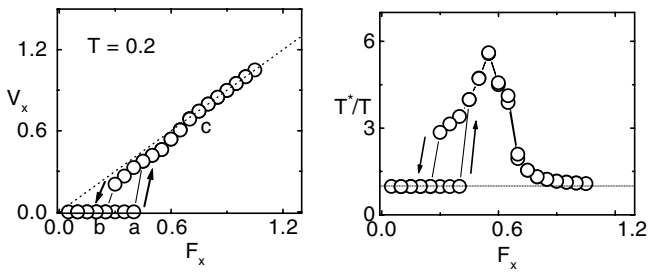

FIG. 1. Drift velocity V_x and effective temperature of the overlayer T^* as a function of the external force F_x along the x axis for $\epsilon = 1$ and $\eta = 1$. The direction of variation of F_x is indicated by arrows.

In the sliding solid phase that exists at $F_x > F_c$ in Fig. 1, the motion of the overlayer essentially averages out the pinning potential. Therefore, for our choice of isotropic interacting Lennard-Jones potential, the overlayer should have a triangular structure although the pinning potential has a square symmetry. However, in the transverse direction the pinning potential remains essentially static in a reference frame comoving with the overlayer [12] and it is plausible that the overlayer may remain pinned in the transverse direction, depending on temperature and velocity, although it is sliding in an incommensurate state along the longitudinal x direction.

We have determined the nonlinear response of the overlayer in the transverse direction in the sliding state. Once the system has reached a steady state for a fixed $F_x > F_c$, an additional force F_y is applied, and the average drift velocity V_d is determined after the new steady state is reached. Alternatively, the transverse mobility $\mu_t = V_y/F_y$ can also be obtained by applying a total force (F_x, F_y) to an initial equilibrium state $(F_x = 0)$. The two methods give the same result, indicating a well-defined transverse depinning behavior [12] as shown in Fig. 2a. At temperatures below a critical value T_p, which is much smaller than the melting transition temperature T_m, the mobility μ_t vanishes, implying that the overlayer remains pinned along the transverse direction, even when it is sliding along the longitudinal x direction. Above T_p, the mobility μ_t has a finite value. In the temperature range $T_p < T < T_m$, there is no transverse critical force and the overlayer is an incommensurate sliding solid in all directions. As shown in Figs. 2b and 2c, below T_p the $F_y \times V_y$ characteristics show a critical transverse force F_{ya} and hysteresis behavior when the system is first equilibrated at lower temperatures. Note, however, that, for decreasing transverse force, the pinned state is only reached at $F_{yb} \sim 0$, unlike the behavior in the longitudinal direction in Fig. 1, suggesting that along the transverse direction the existence of a critical force F_{ya} may not necessarily lead to stick and slip motion. The behavior of the nonlinear response in the transverse direction correlates with the structure of the sliding state at $F_y = 0$.

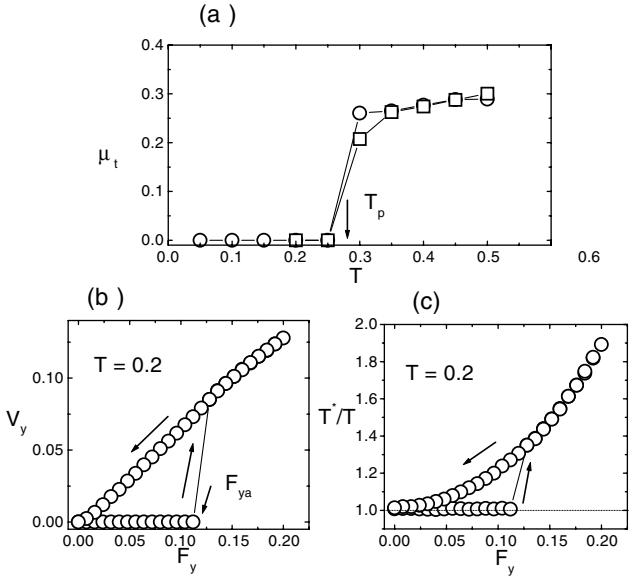

FIG. 2. (a) Transverse mobility μ_t as a function of temperature obtained by applying a total force (F_x, F_y) to an initial equilibrium state (squares) and by applying an additional F_y to the sliding state at $(F_x, 0)$ (circles). (b),(c) Transverse velocity V_y and effective temperature of the overlayer T^* as a function of an additional force along the y axis for fixed F_x. For large F_y, the behavior of T^* is similar to Fig. 1. Large arrows indicate the direction of force variation. The results are for $L = 10$, $F_x = 1.5$, and $\epsilon = 1$.

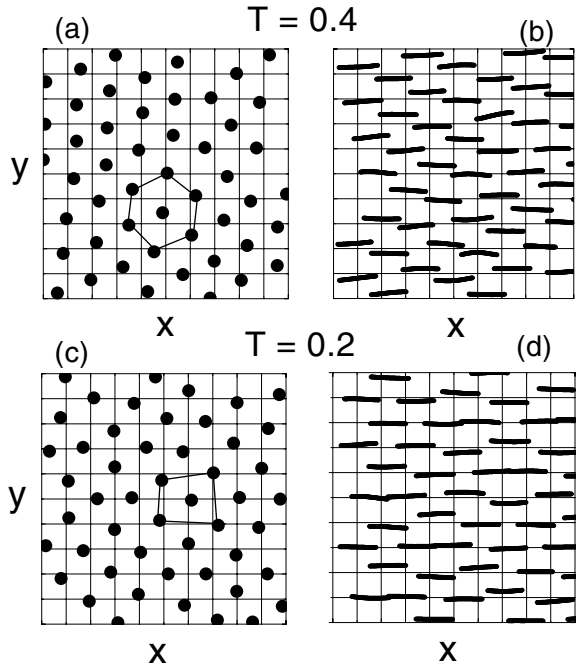

FIG. 3. Snapshot pictures [(a),(c)] and trajectories of the adsorbates [(b),(d)] at $F_y = 0$ for temperatures $T = 0.4$ and $T = 0.2$ above and below the depinning temperature T_p, respectively, for $F_x = 1.5$ and $\epsilon = 1$. The grid lines represent the periodic pinning potential.

Figures 3a–3d show the instantaneous configurations and the trajectories of the adatoms in the overlayer below and above T_p for $F_y = 0$. The trajectories are obtained by superposing successive configurations in the sliding state for a fixed time interval. It can be seen from Figs. 3c and 3d that, in the transverse pinned phase below T_p, the particles are essentially moving in one-dimensional channels along the principal x axis of the pinning potential and the particle coordination number is 4, leading to a square lattice structure. For $T > T_p$, the particle trajectories are free to fluctuate in the transverse direction (Fig. 3b) and an isotropic triangular lattice structure occurs, as shown in Fig. 3a. This is exactly the structure that the overlayer would have in the absence of a periodic pinning potential. The isotropic triangular structure has also been observed in previous calculations [5] but was considered the only possible phase in the sliding state at any finite temperature. Here we find instead that for temperatures below T_p a transverse pinned phase with a different structure is the stable configuration. ffThe dynamical phase transition separating the two sliding states has the characteristic features of an equilibrium first order transition. Besides the hysteresis effects mentioned earlier, we also observe supercooling effects in the sense that when the sliding state above T_p is cooled down to a temperature below T_p, at fixed F_x, the final sliding state retains its triangular structure in a metastable state, as shown in Fig. 4a. This indicates that a nucleation process with large energy barrier is involved. While the details change, the hysteresis and supercooling effects are observed up to the largest size (20×20) studied here. We also note that the physics underlying the transverse depinning transition is very similar to that of the longitudinal depinning transition which has been shown to be very general and model independent [5,8].

The results for other values of F_x and ϵ are qualitatively the same as long as $F_x > F_c$. The linear transverse mobility μ_t (in the limit $F_y \to 0$) as a function of the longitudinal driving force is shown in Fig. 4b for a temperature $T = 0.2$ below T_p. It is seen that μ_t remains zero for large F_x and the onset of finite μ_t for decreasing force occurs at $F_x = F_c$, where the sliding state goes from a solid phase into a fluid phase as would be expected from the behavior of the longitudinal response in Fig. 1. Thus there is a transverse depinning transition at all values of $F_x > F_c$. The dependence of the transverse depinning transition temperature T_p on the stiffness of the overlayer ϵ together with the melting transition temperature T_m at $F_x = 0$ are indicated in Fig. 4c. These transition temperatures are located from the onset of finite linear mobility μ_t and μ_L, respectively, for increasing temperatures.

Recent work [10] on the dynamics of sliding two-dimensional vortex lattices on a periodic potential also shows evidence of a transverse pinning and critical Lorentz force in the sliding state. The form of interaction and the pinning potentials for the vortex lattice model are different from that for the overlayer model considered

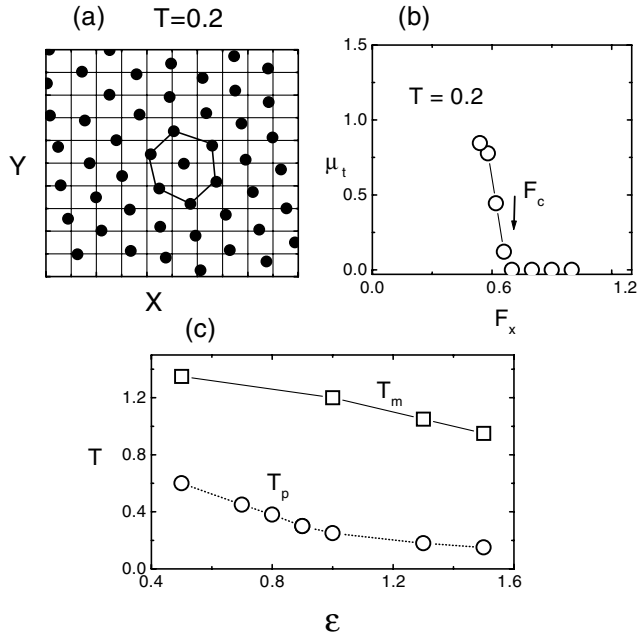

FIG. 4. (a) Snapshot picture of the final state at $T = 0.2$, below T_p, obtained from an initial sliding state at $T = 0.4$, above T_p, by lowering the temperature at fixed $F_x = 1.5$. (b) Transverse mobility μ_t as a function of the longitudinal force F_x at a temperature below the depinning transition temperature T_p. The results are for $\epsilon = 1$, $L = 10$. (c) Critical temperature T_p for transverse depinning in the sliding state for $F_x = 1.5$ and equilibrium ($F_x = 0$) melting transition temperature T_m as a function of the overlayer stiffness ϵ.

here, but this is not expected to be qualitatively important in the sliding state [14]. More importantly, the results for the vortex lattice were obtained at zero temperature and for an overdamped dynamics with zero vortex mass. Our results are obtained for a nonzero mass, finite damping coefficient η, and finite temperatures. We have shown that in our model the transverse pinned state is stable against thermal fluctuations up to a transverse depinning critical temperature $T_p < T_m$, as indicated in Fig. 4c. In view of the similarities of these systems and the current interest in the transverse critical current for vortex lattices in superconductors [12], it should also be interesting to investigate how a nonzero mass [15] will affect the sliding state in vortex lattices and if the presence of weak disorder will broaden the sharp transition found in this paper.

The existence of transverse static friction in the sliding state implies that, once two lubricated surfaces are sliding relative to each other in a particular direction commensurate with the underlying pinning potential, changing the direction of motion may require a nonzero additional force depending on the temperature and sliding velocity. Experimentally, the transverse critical force could, in principle, be detected from measurements of sliding friction with simultaneous transverse (in-plane) vibrations between two boundary lubricated sliding surfaces, using similar techniques as in recent experiments which introduce coupling to normal (out-of-plane) vibrations [2] and also in sliding colloidal layers confined between two surfaces [16]. Below the thermal depinning temperature the transverse vibrations should have very little effect on the longitudinal motion for sufficiently small vibration amplitude, while, above the transition, induced oscillations on the longitudinal friction are expected. We hope that this work will motivate novel experiments on sliding friction.

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