

## Theory of lubrication due to collective pinning

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(Received 14 September 2004; revised manuscript received 23 November 2004; published 13 May 2005)

In collective pinning theory, the problem of two three-dimensional solids in contact is at its critical dimension. This implies that when the disordered forces acting between the two solids at the interface are relatively strong, the force of static friction should be large, but at smaller values of these forces, the system switches over to a regime of weak static friction. It is argued that this provides a mechanism for the reduction of friction in boundary lubrication. Lubricant molecules reduce static friction by smoothing the roughness of the surface, thus allowing the force pushing the surfaces together to be supported by more points of contact, which can switch the interface from the strong- to weak-static-friction regime.

DOI: 10.1103/PhysRevE.71.056107

PACS number(s): 81.40.Pq, 68.35.Af, 68.35.Gy

### I. INTRODUCTION

Experimental work done using the surface force apparatus [1] shows that when liquid lubricants are squeezed between two surfaces, at pressures comparable to those that occur at the interface between two asperities in contact, the liquids exhibit a shear response characteristic of solids. Such a result has also been found in molecular dynamics simulations [2]. It is difficult to understand why such a film should shear significantly more easily than the bare solid surfaces that they are designed to lubricate. In this paper a possible mechanism is proposed for how such films can reduce friction between two solid surfaces.

In order to provide some background for this problem, let us first consider periodic surfaces in contact. Muser and Robbins [3], Muser [4], and Lancon [5] have shown using simulations that two perfect elastic crystalline solids in contact will exhibit no static friction if they do not interact chemically, even when the solids are pressed together with forces which are comparable to the forces at asperities in contact when rough surfaces are pressed together. This remains true until the force pressing the solids together exceeds a critical value. At that point, the interface undergoes a transition known as the Aubry transition [6] to a state in which the static friction becomes nonzero. In the Appendix, it is shown that whereas for two incommensurate periodic solids in contact distortions that occur as a result of the interactions of the solids across the interface between them are limited to a very thin region, for a disordered interface, the distortions are typically spread over long distances from the interface. Thus, whereas simulations on very thin solids used in Refs. [3,4] are justified for incommensurate periodic solid interfaces, they are not justified for disordered surfaces in contact. Several of these authors have found that when there are mobile molecules present at such an interface between periodic surfaces, there will always be static friction [7,8]. We have also studied both static and kinetic friction in the slow-sliding-speed limit based on this model in several previous publications [9,10]. The treatment in Ref. [10] is based on the idea that kinetic friction in the slow-sliding-speed limit results from multistability of the interface [11]. Most real interfaces, however, are not perfectly flat, and hence they are only in

contact at relatively small regions located on top of asperities which are in contact. It has been argued that this lack of flatness of the surfaces on the micron scale will likely result in static friction, even in the absence of mobile molecules, but the friction coefficient will likely be very small [9].

The surfaces in contact at these asperities, however, are not periodic, but are disordered. Muser [4] has shown in his simulations that when the periodicity of two incommensurate surfaces that were initially periodic is destroyed the interface can switch from one not exhibiting static friction to one which does. Collective pinning theory [12–14] applied to the geometry of the present problem predicts that as the forces between the two disordered surfaces increase, the interface will switch from a regime of small to one of large static friction.

When surfaces are pushed together with large forces, the forces between the surfaces are likely to be dominated by the hard-core repulsions of the surface atoms. This results in an increase of the components of the forces between atoms parallel to the interface on opposite surfaces as the force pushing the surfaces together is increased. Consider a pair of micron-scale asperities (one from each surface) which are being pushed together normal to the surface with a normal force or load  $F$ . If the area of contact of the interface between these asperities is  $A$  and a fraction  $c$  of the atoms at this interface are in contact, the load per atom in contact is of the order of  $F(a^2/cA)$ , where  $a$  is the mean interatomic spacing. Since the contacting atoms rarely lie one exactly above the second, there will be a component of force along the surface as well of the same order of magnitude, but in an arbitrary direction. Strong pinning, in the present context, means that all of these contacting atoms are displaced by a sufficient amount to minimize the potential due to the atoms from the asperity from the second surface, at the expense of the elastic forces holding the atoms in place. Then, if we attempt to shear these two asperities relative to each other, there will be a net force which opposes this shear motion, which is of the order of the above load per contacting atom multiplied by the number of atoms in contact, which gives  $F$ . In contrast, in the weak-pinning limit, the elastic forces are sufficiently strong to prevent these atoms from minimizing

the potential resulting from the second asperity. Hence, when we try to shear the interface, there will be a force opposing the shear which is of the order  $F(a^2/cA)^{1/2}$  (since  $cA/a^2$  is the number of atoms in contact) because the frictional forces which result from the hard-core repulsions of the atoms in contact do not add together coherently because they point in random directions. The net force of friction is a fluctuation in the resultant force, and fluctuations are proportional to the square root of the number of atoms in contact, which produce the force. Hence, it is given by the load per atom, given above, multiplied by the square root of the number of atoms in contact,  $cA/a^2$ .

If  $A$ , instead of representing the contact area of a micron-size asperity, represented the area of contact between two perfectly flat macroscopic surfaces (for which  $A$  is macroscopic), the force of static friction would be negligible. Since the area of contact of two contacting asperities, however, is not infinite, the static friction will never become zero but it can still become very small, if the interface can be switched from the strong- to weak-pinning limit. Likely types of roughness on a surface of an asperity in contact with a second asperity are regions at the surface where surface atoms are missing and step edges which are pressed into contact [15,16]. It is proposed in this article that the local forces at an interface between two asperities in contact could be reduced by filling in these holes in the surfaces with small molecules and that this is a possible mechanism for lubrication. By filling in these holes we spread the force pushing the asperities together over a larger area of contact, thus reducing the force per unit area. This can switch the interface from the strong-pinning regime, in which the static friction is large, to the weak-pinning regime, in which the static friction is small [12–14]. In the present article, this mechanism will be discussed in detail and another mechanism for lubrication will be proposed in which relatively large molecules of a monolayer of lubricant can be compressed, if they lie on relatively high regions on a surface, and thus spread the force pushing the solids together over a larger area of contact. Although the idea that increasing the amount of area of contact can reduce the friction seems at first sight to contradict the picture provided by Greenwood and Williamson [17] for explaining Amonton's law, in which it is assumed that the friction actually increases with increasing area of contact, this is in fact not the case. In the present treatment, the friction does in fact increase with increasing area of contact. When the area of contact gets large enough to switch the interface to the weak-pinning limit, however, the friction switches from being proportional to the area of contact to being proportional to its square root.

It is not being claimed that the mechanism for boundary lubrication proposed here explains how all lubricants reduce friction. All that is being proposed are a couple of mechanisms for reduction of friction by model lubricant molecules that are strongly attached to two surfaces which are in contact.

Section II will review collective pinning theory, for two three-dimensional elastic solids in contact. In Sec. III, a mechanism for friction due to small molecules is discussed. Section IV discusses a mechanism for lubrication due to large molecules.

## II. COLLECTIVE PINNING THEORY FOR SOLIDS IN CONTACT AT A DISORDERED INTERFACE

For completeness the transition from strong to weak pinning for two three-dimensional elastic solids in contact at a disordered interface using scaling arguments, which was first presented in Ref. [9], will be discussed. We expect that qualitatively correct results for this problem can be obtained by studying the simpler problem of a three-dimensional elastic solid in contact with a rigid disordered substrate. This problem was also studied in Ref. [18] in the context of a macroscopic solid in contact with a substrate at randomly distributed asperities using perturbation theory. In the present work, we will apply this model to a pair of asperities from two surfaces in contact. Here the disorder occurs over the area of contact of the two asperities. In collective pinning theory [12–14], there is competition between a disordered potential and an elastic medium which interacts with this potential. In the strong-pinning limit, the elastic solid is able to distort enough to essentially minimize its interaction with the disordered potential. In the weak-pinning limit, the solid has little distortion over a volume, known as a Larkin domain, whose linear size, the Larkin length, is determined by minimizing the sum of the interaction with the disordered potential and the elastic distortion energy, provided the system's dimension is below the critical dimension for the problem. The three-dimensional solid interacting with a two-dimensional disordered substrate, however, is at its critical dimension [9].

Following a generalization of the discussion in Ref. [9], let us consider a homogeneous elastic solid interacting with a two-dimensional rigid disordered substrate, located at  $z=0$ . The energy of this system can be written as [19]

$$E = \int d^3r \left[ K \sum_{\alpha,\beta} \left( \frac{\partial u_\alpha}{\partial x_\beta} \right)^2 + K' \left( \sum_\alpha \frac{\partial u_\alpha}{\partial x_\alpha} \right)^2 - V(\mathbf{r} + \mathbf{u}(\mathbf{r})) \delta(z) \right], \quad (1)$$

where  $\alpha$  and  $\beta$  run over the components  $x$ ,  $y$ , and  $z$ ,  $u_\alpha(\mathbf{r})$  denotes the  $\alpha$ th component of the displacement field at the point  $\mathbf{r}$  in the elastic medium,  $K$  and  $K'$  are the elastic moduli (i.e., the Lamé coefficients [19]), and  $V(\mathbf{r})$  denotes the substrate potential per unit area. We look for an approximate solution of the form  $u_\alpha = u_\alpha(x/L, y/L, z/L')$ , where  $u$  varies by an amount of the order of the range of a potential well of the substrate potential when  $x$  and  $y$  vary over a distance of  $L$  or  $z$  varies over a distance of order  $L'$ . These are the Larkin lengths along and perpendicular to the surface. We substitute this expression for  $u_\alpha$  in Eq. (1) and approximate the integral of the first two terms in the integrand of Eq. (1) over a single Larkin domain by the product of the average over a Larkin domain of first two terms in the integrand of Eq. (1) and the volume of a Larkin domain,  $L^2L'$ , and then multiply by the number of domains,  $A/L^2$ , where  $A$  is the area of the interface. Minimizing with respect to  $L'$ , we obtain

$$L' = L \left( \frac{\Gamma'}{\Gamma} \right)^{1/2}, \quad (2)$$

where  $\Gamma = \sum_{\alpha,\beta=x,y} K \langle (\partial u_\alpha / \partial x_\beta)^2 \rangle + K' \sum_{\alpha=x,y} \langle (\partial u_\alpha / \partial x_\alpha)^2 \rangle$ , and  $\Gamma' = K \langle (\partial u_z / \partial z)^2 \rangle + K' \sum_\alpha \langle (\partial u_\alpha / \partial z)^2 \rangle$ , where  $(x', y', z')$

$= (x/L, y/L, z/L')$ , and  $\langle \dots \rangle$  signifies an average over a Larkin domain. Since the derivatives of  $u$  are all of the order of atomic distances,  $L' \approx L$ . Assuming that  $V(\mathbf{r})$  is completely random and  $\mathbf{u}(\mathbf{r})$  varies by a negligible amount as  $\mathbf{r}$  runs over a domain of volume  $L^2 L'$ , the integral of  $V(\mathbf{r})$  over this domain is of the form  $V_0 c^{1/2} L/a$ , where  $V_0$  is the root-mean-square (rms) value of the potential of interaction between a surface atom and the substrate and  $c$  is the fraction of the surface atoms which are in contact with the substrate. In arriving at this result we assumed that the variation of  $\mathbf{u}(\mathbf{r})$ , when  $\mathbf{r}$  varies over a distance small compared to  $L$ , is negligibly small compared to the length scales on the substrate. Since the substrate is random, the integral over  $V(\mathbf{r})$  is proportional to the square root of the number of surface atoms in contact with the substrate, which is of the order of  $c(L/a)^2$  where  $a$  is a mean atomic spacing or potential well size. Let the force per unit apparent area pushing the surfaces together be denoted by  $P$ . Since the number of surface atoms in contact with atoms from the second surface is of the order of  $cA/a^2$ , the mean force between two atoms in contact from each of the two surfaces is given by  $PA$  divided by this quantity or  $Pa^2/c$ . Hence, since  $V(\mathbf{r})$  varies on a length scale  $a$ ,  $V_0 \approx Pa^3/c$ . Then substituting Eq. (2) in Eq. (1), we obtain

$$E = [2(\Gamma\Gamma')^{1/2} - Pa^2/c^{1/2}]A/L \quad (3)$$

for the energy, which is minimized for infinite  $L$  if  $2(\Gamma\Gamma')^{1/2} > Pa^2/c^{1/2}$  and for  $L=0$  (which in practice means that  $L$  is as small as the smallest length scale in the problem rather than zero) if  $2(\Gamma\Gamma')^{1/2} < Pa^2/c^{1/2}$ . Thus, it is clear that as  $c$  decreases, the interface can switch from weak pinning (if it was already in the weak-pinning regime) to strong pinning. In the latter regime, by the arguments given in the last paragraph, the surfaces will be pinned together; i.e., there will be static friction. Because the interface area between two asperities in contact is only of micron size, there will be a transition from low to high, rather than from zero to nonzero static friction (as would occur for an infinite interface). In the next two sections ways will be proposed to increase the effective value of  $c$  (i.e., increase the number of points of contact of the surfaces) by using proposed model lubricant molecules and, by doing so, reducing the friction.

This problem can also be considered using perturbation theory in the weak-pinning limit [18]. To do this, following Ref. [18], one calculates  $\mathbf{u}(\mathbf{r})$  which results from the random forces found from  $V(\mathbf{r})$  and from it calculates  $\langle |\mathbf{u}(\mathbf{R}) - \mathbf{u}(0)|^2 \rangle$  using the standard expression for the elasticity Green's function [19]. Here,  $\langle \dots \rangle$  signifies an average over the random substrate forces.  $R$  is considered to be equal to the Larkin length when this quantity is comparable to the square of the range of a substrate potential well, as this represents the distance over which the surface of the solid can be considered as rigid from the point of view of the random substrate potential. Following arguments similar to those in Ref. [18], we find a Larkin length that is an exponential function of the ratio of Young's modulus divided by  $V_0$ , which can easily be quite large compared to any reasonable size solid interface when this ratio is reasonably large, as it is in the weak-pinning regime. Thus, even though the varia-

tional method that I used above gives an infinite Larkin length in the weak-pinning limit, whereas the perturbation theory method of Ref. [18] gives a finite Larkin length, since the Larkin length found in Ref. [18] is extremely large (i.e., an exponential function of a fairly large number) in the weak-pinning limit, the two methods can be considered to give qualitatively the same result.

### III. LUBRICATION BY SMALL OR NARROW CHAIN LUBRICANT MOLECULES

Asperities on the surface of a solid can occur on many length scales, and in fact, for self-affine surfaces, the asperities look the same when viewed on all scales, until we get down to atomic length scales [16]. At this scale, the roughness must reflect the atomic arrangement of the solid. At the area of contact of two asperities, there is likely to be atomic-scale roughness, consisting of regions along the surface at which a small section of the top layer of atoms is missing. This type of roughness is illustrated by a sketch shown in Fig. 1(a). There is also likely to be atomic-level roughness due to steps on asperities which are distorted as they are pressed into contact. This is illustrated in Figs. 1(b) and 1(c). Figure 1(b) is a sketch of two asperities before they are placed in contact. The step structure illustrated in this figure corresponds to what is likely to occur if a crystal plane of each of the two surfaces in contact is parallel to the surface. The step structure shown is what one must have in order for there to be hills and valleys (i.e., asperities) on the surfaces. Figure 1(b) illustrates what is found in scanning tunneling microscope (STM) studies of surfaces [15]. As is illustrated in Fig. 1(c), if the asperity sides are in contact, the interface which must be sheared in order to initiate sliding between the asperities also has high regions at which the asperity surfaces are in contact separated by holes, whose depths are comparable to atomic spacings. Figure 1(c) was obtained from Fig. 1(b) by distorting the two asperities uniformly as they are placed in contact. This is qualitatively what one expects to occur if we assume that the asperity distortions are described by continuum elastic theory [19]. If the surface is not parallel to a crystal axis, there will also be steps on the peak of an asperity similar to the steps on the side of an asperity that are illustrated in Figs. 1(b) and 1(c). The mean upward slope of the asperity-asperity interface shown in Fig. 1(c) will not contribute to static friction because in practice it is highly unlikely that pairs of asperities in contact will be lined up so perfectly that they will be forced to slide directly over each other. Even if they did, it was shown by Caroli and Noziere [11] that a random distribution of the relatively short and fat asperities that occur on most surfaces will not exhibit static friction (if this atomic-level step structure is not considered). Rather, the important factor which determines whether or not there will be static friction is whether or not this interface is pinned by the roughness due to the steps. For the type of roughness illustrated in Fig. 1(c), the contact area will be a small fraction of the total asperity-asperity interface, making it likely that this interface will be in the strong-pinning limit. (What I am saying is that in the absence of such atomic-level roughness, the upward slopes of the asper-

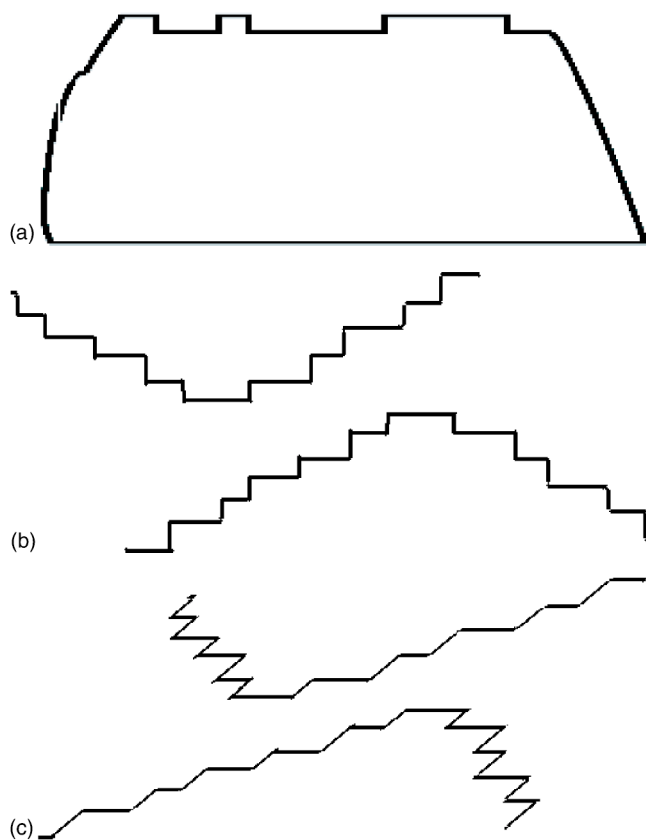


FIG. 1. Rough sketches are given illustrating the types of atomic-level disorder considered in this article. (a) The top sketch illustrates disorder due to an incomplete top atomic layer on an asperity. (There will, of course be step structure on the sides of the asperity as well, but it is not shown here because I wish to illustrate roughness due to an incomplete top layer of the asperity.) (b) The middle sketch illustrates the stepped structure of the sides of two asperities before they are placed in contact, and (c) the lower figure illustrates what the two asperities in (b) are likely to look like after they are placed in contact. Note that this figure is only meant to illustrate the concept of having atomic-level roughness of the asperities resulting from the step structure, and the steps are much larger compared to the asperity size than would be the case for real asperities.

ity interfaces will not lead to static frictions, as proved by the arguments given in Ref. [11].)

Consider the type of atomic-level roughness illustrated in Fig. 1(a). Now let us imagine placing two of these surfaces in contact and pressing them together with a load per unit apparent area of the interface  $P$ . If the load is sufficiently high, we may safely assume that the force between a pair of surface atoms from the two surfaces which are in contact is dominated by the hard-core interaction. Then, a good estimate of the magnitude of the mean force acting between a pair of surface atoms from the two surfaces that are in contact is  $P/n$ , where  $n$  is the number density per unit area of surface atoms which are in contact. This is an estimate of the component of force normal to the interface, but since such atom pairs are rarely lined up so that one is exactly on top of the other, there will be components of force of comparable magnitude along the interface as well, in random directions,

as discussed in the Introduction. Let us now imagine adding atoms of the same material to each of the outer layers until they each form a complete monolayer. Our model is then identical to the model studied in Refs. [3–5]. In this model it was reported in these references that even for loads per unit area as high as a GPa, there was no static friction between the surfaces. As the concentration of surface atoms is reduced from a complete monolayer on each surface by removing atoms, there will certainly be a concentration at which the interface switches over from the weak-pinning limit, in which the elastic energy dominates over the substrate potential (which in our case represents the second surface), to the strong-pinning regime, in which the opposite is true.

Let us now place molecules of a lubricant on these surfaces, which have the property that they attach themselves strongly to the surfaces. This is the property that lubricant molecules must have in order to be good lubricants [16]. Then, let us assume that the attractive force between a lubricant molecule and a surface atom is much greater than the attraction between two lubricant molecules and the temperature. These can be either single atoms or chains. If they are chains, we require that they be very flexible and consider how the individual monomers position themselves on the interface. Simulations done for two flat surfaces in contact [2] show that under GPa pressures such a lubricant will get squeezed out until, at the highest pressures, we are left with a bilayer. We will now illustrate this proposed mechanism for lubrication for the model for atomic level roughness illustrated in Fig. 1(a). In our case, where the surface is not smooth, we expect there to be a bilayer coating the steps (i.e., on the places on the outer layer at which there are surface atoms). The regions at which there are no top sub-monolayer atoms present will also get filled in with lubricant molecules. The reason for this is that as the surfaces are squeezed together, some of the lubricant molecules that are driven from the steps will be pushed into these regions. Some of them become trapped in the one atomic layer deep “valleys” in the outer surfaces of the solids. We can see from Fig. 1 that the second layer of lubricant molecules that we propose to be present inside the valleys can easily be trapped there by lubricant molecules which are adsorbed on the tops of islands of top surface atoms (since they are assumed to be strongly attached to them). These molecules partially support the load, and hence, since the load is now supported over a larger area of contact, the system might be switched to the weak pinning regime, resulting in a significant reduction of friction.

I have performed a Monte Carlo calculation to demonstrate that the lubricant molecules when compressed between two surfaces will get squeezed into atomic-depth holes in the surfaces. A lubricant, consisting of 245 spherically symmetric molecules interacting with a Lennard-Jones potential, is placed between the two surfaces, and the surfaces are then moved together so as to compress the lubricant down to a bilayer. The lubricant molecules interact with each of the surfaces with a Steele potential [20], which has the form

$$V(\mathbf{r}) = V_0(z) + V_1(z) \sum_{\mathbf{G}} e^{i\mathbf{G}\cdot\mathbf{r}},$$

where  $\mathbf{G}$  is a reciprocal-lattice vector for the surface. This potential, however, possesses a defect which must be dealt

with. Namely, when a molecule is sufficiently strongly pressed into one of the surfaces (because of the high pressures) the amplitude of the corrugation term  $V_1(z)$  can dominate over  $V_0(z)$ . Since the second term can be negative and since both  $V_1(z)$  and  $V_0(z)$  diverge as  $z$  approaches zero, the total potential  $V(\mathbf{r})$  can become unstable. This defect of the Steele potential is compensated for by making  $V_1(z)$  level off before it exceeds  $V_0(z)$ . The energy parameter  $\epsilon$  in the Lennard-Jones interaction  $-4\epsilon[(\sigma/r)^6 - (\sigma/r)^{12}]$  between the lubricant molecules was chosen to be a tenth of the energy parameter  $\epsilon_{gs}$  in the Steele potential [20]. The potential minima are taken to lie on a triangular lattice of lattice spacing  $a=2.88 \text{ \AA}$ . The bottom surface contains a hexagonally shaped hole of semimajor and semiminor axes 7.6 and 6.6  $\text{\AA}$ , respectively. The surfaces are circular with radii equal to  $6a\sqrt{3}/2$  where  $a$  is taken to be 2.885  $\text{\AA}$ . If  $V_s(x, y, z)$  represents the potential due to this surface for values of  $x$  and  $y$  outside the hole (where the  $z$  axis is normal to the interface),  $V_s(x, y, z+z_0)$  is taken to be the potential inside the hole. Here  $z_0$  is taken to be equal  $a(2/3)^{1/2}$ , the depth of a hole, resulting from removing atoms in the surface layer of a hexagonal-close-packed (hcp) lattice with its  $c$  axis normal to the surface or a face-centered-cubic lattice with its (111) surface parallel to the surface. The calculation is started with the lubricant molecules placed in an hcp lattice five atomic layers thick with lattice constant  $a$  between the two surfaces and centered over the hole. The initial separation of the surfaces is 15  $\text{\AA}$  which is just enough for the initial crystal of lubricant molecules to fit without being compressed. The surface is kept at that separation for  $2 \times 10^6$  iterations. The separation is then reduced by  $0.5 \times 10^{-7} \text{ \AA}$  for every Monte Carlo iteration until the separation reaches 9.84  $\text{\AA}$ . The interface pressure at this separation is  $3.27 \times 10^{11} \text{ dyn/cm}^2$ , which is larger than the pressure at the area of contact of two asperities used in Ref. [9]. We stop reducing the separation at this point. For smaller separations the total potential energy of the lubricant becomes large and positive, indicating that the film is becoming highly compressed. Results of these calculations for  $k_B T = 20\epsilon$  (where  $k_B$  is Boltzmann's constant and  $T$  is the absolute temperature) are shown in Fig. 2. The radius of the shaded spheres used to represent a lubricant molecule was chosen to be approximately equal to the radius of a lubricant molecule used in the simulation in  $b$  and  $c$  but not in  $a$  (for clarity). Lines were drawn to show the approximate locations of the surfaces and the hole in the lower surface. As can be seen, the film gets compressed into a bilayer outside of the hole and the hole gets filled with a high concentration of lubricant molecules one monolayer thick, which could support load over the region in which the hole occurs. The interface between the two layers of lubricant will be an interface which will shear quite easily compared to an interface between unlubricated surfaces, as the only interaction acting across this interface is the interaction between pairs of lubricant molecules, which was assumed to be much weaker than the interaction between the two bare surfaces.

In contrast to the valleys in the outer surfaces assumed to occur at the interface between two asperities, lubricant molecules are not expected to get trapped in the space at the sides of the micron-scale asperities, because the depth of

such regions is much too great to allow the attractive force between surface atoms and lubricant molecules to reach the lubricant molecules in most of this region. Thus, most of the molecules in this region are free to flow through the space around the sides of the asperities. As a result, most of the lubricant found here remains liquid. Only molecules which are trapped at the interface between a pair of asperities which are in contact will be under high enough pressure to exhibit the solidlike properties found in Ref. [1]. The mechanism for boundary lubrication suggested here should be applicable at pressures that occur at the contact area between two asperities, which can reduce the lubricant concentration down to a monolayer or less coating each surface. The model for atomic-level roughness illustrated in Figs. 1(b) and 1(c) should also result in finite-atomic-size depth holes in the surfaces of two asperities in contact because of the irregular shape of the steplike terraces resulting in kinks in the steps [15] making up the slopes of the sides of the asperities on the surfaces. Lubricant molecules will likely get trapped between pairs of kinks, in much the same way as they do in holes in the top layer [as illustrated in Fig. 1(a)]. Even if the layer of lubricant molecules which fills in the holes in the region of contact of two asperities is slightly higher or lower than the walls of the hole, the mechanism will still work because the high loads that occur at the interface between two asperities are sufficient to equalize these heights by compressing either the lubricant or surface atoms. For example, for a Lennard-Jones energy parameter of about 20 K, they can be compressed by at least 12%. This is easily shown by setting the repulsive part of the force due to the Lennard-Jones interaction equal to the force per surface atom due to the load supported by the pair of asperities in contact. The amount of compression could even be larger than this because until the compression occurs, the load is supported over a smaller area.

The idea of lubricant molecules filling in valleys in the top layer of each surface and thus making the top surface more smooth at first sight seems like the familiar idea of reducing friction by making the surfaces smoother, but here we have provided a mechanism for how such "smoothing" results in low friction. The idea that by doing so the interface switches from the strong-pinning regime, in which there is large static friction, to the weak-pinning regime, in which there is little static friction, provides a mechanism for how such "smoothing" of the surface with lubricant molecules can lead to low friction.

#### IV. LUBRICATION BY LARGE MOLECULES

In the last section, it was argued that a thin layer of molecules of size comparable to the depth of the holes in the surfaces in contact of the two solids could reduce the static friction by spreading the force pushing the surfaces together over a larger number of randomly placed points of contact (by filling in the holes in the surface). This can switch the interface from the strong- to weak-pinning limit, resulting in a large reduction in the friction. Here, I will propose another mechanism for reducing the friction. Let us coat the surface with a thin film of a more readily compressible material con-

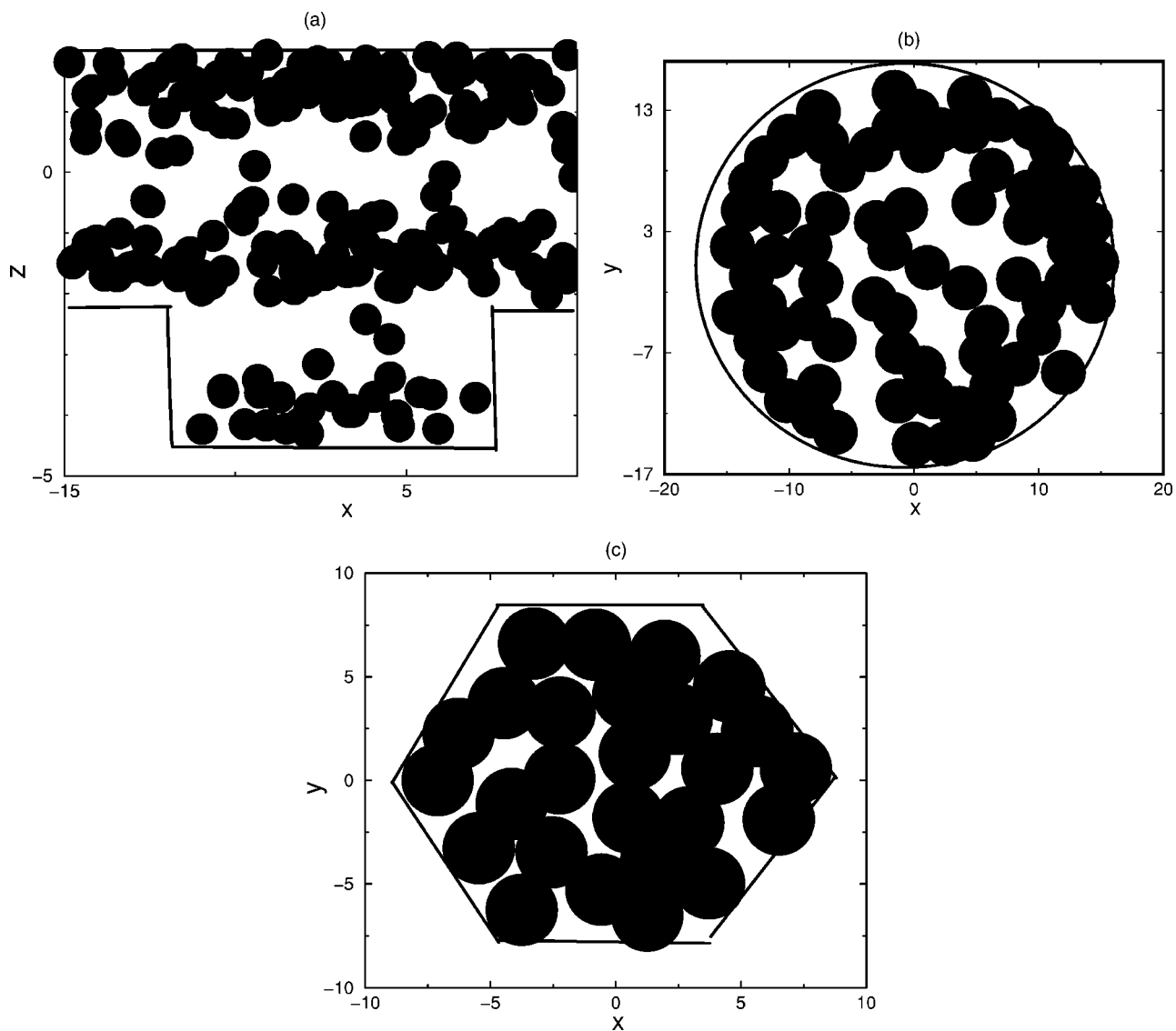


FIG. 2. In (a), there is a side view of the distribution of molecules between the two surfaces. In (b), the molecules in a slice of width equal to  $1 \text{ \AA}$  centered around  $z = -3.8 \text{ \AA}$  are shown, which are clearly located inside the hole. In (c), the molecules in a slice of width equal to  $1 \text{ \AA}$  centered around  $z = -1.5 \text{ \AA}$  are shown, which are clearly located in one of the layers outside of the hole. Both the  $x$  and  $y$  axes in (a), (b), and (c) are in units of  $\text{\AA}$ .

sisting of molecules which are noticeably larger than atomic dimensions. For example, let us consider a complete monolayer of elastic globular molecules of radius noticeably larger than an atomic spacing [for example, octamethylcyclotetrasiloxane (OMCTS) [21]] coating the surfaces. The surface of the solid has the type of atomic-level roughness discussed earlier and illustrated in Fig. 1—namely, holes in the surface of atomic-distance depths. The outer surface on the other side of the coating will clearly reflect this roughness, although some of it will get smoothed out because of the large ratio of the lubricant molecule’s size and the length scale of the surface roughness. This is illustrated schematically in Fig. 3. In fact, this will already significantly increase the area of contact compared to the bare surface, because the bare surface has a very small concentration of its surface atoms in contact (namely, those at those parts of the steps that were in contact before the surfaces were coated). When the outer

surface gets pressed against a second solid, which we will model here for simplicity by a flat substrate, the lubricant molecules on the high parts of the outer surface will get compressed. If they get compressed enough, parts of the outer surface which were not initially in contact will now come in contact. This will spread the force pressing the solids together over more points of contact, which could switch the system from the strong-pinning to the weak-pinning regime, resulting in a large reduction of the static friction.

Those lubricant molecules which are in contact are the ones located on high points on the surfaces. We will refer to tops of these higher-lubricant molecules, which are in contact, as miniasperities. The interface potential between two miniasperities varies on atomic-length scales, as does the interface between two micron-scale asperities. Since the miniasperities are much smaller, however, they are much stiffer, and hence, it is possible for appropriate parameters for them

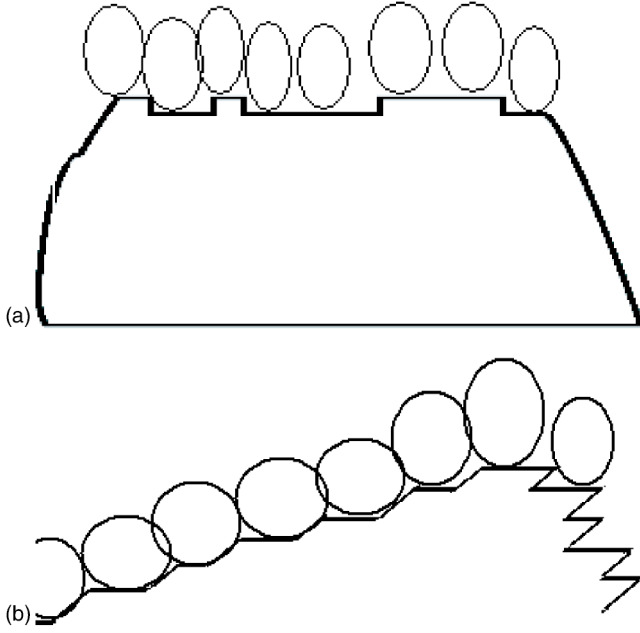


FIG. 3. The arrangement of larger lubricant molecules discussed in Sec. IV is illustrated schematically for the type of roughness shown in Figs. 1(a) and 1(c).

to be sufficiently stiff so that they cannot sink into their interface potential minimum. The condition for this to occur will be considered later in this section. In order for the interface to be in the weak-pinning regime it must be in this regime on several length scales. First of all, the elastic forces that hold the atoms at the area of contact of two miniasperities in place must be sufficiently strong so that each atom on one miniasperity does not drop into the minimum of the potential well provided by the other miniasperity. Second, the individual miniasperities must be sufficiently stiff so that they do not drop into a minimum of the net interface potential (i.e., the sum of the potentials provided by the atoms of the second miniasperity with which it is in contact). Third, if both of these conditions are met, the film as a whole must be in the weak-pinning regime, and fourth, the bulk solid must be sufficiently stiff so as to be in the weak-pinning regime.

In order to determine whether the atoms at the interface between two miniasperities in contact are in the weak-pinning regime, we can apply the methods of the last section to a single miniasperity. We will for simplicity replace the miniasperity with which it is in contact by a substrate, consistent with the approach that we used in Sec. II. If the lubricant molecule contains enough atoms so that the continuum approximation is a good first approximation to the problem, this can be examined using the methods of the previous section. If the molecules are sufficiently large compared to atomic dimensions, we can apply the arguments used in Sec. II for an asperity to a miniasperity. The argument is as follows: For a load per unit apparent area  $P$ , the total load pushing the surfaces together is  $PA$ , where  $A$  is the total surface area. The number of miniasperities in contact is  $\sim c'A/\ell^2$ , where  $c'$  is the fraction of miniasperities which are in contact and  $\ell$  is the radius of a lubricant molecule. Then the net load per contacting miniasperity is the quotient of

these two quantities or  $P\ell^2/c'$ . Hence, the force per miniasperity atom is  $(P\ell^2/c')(a^2/A_c)$ , where  $A_c$  is the mean area of contact of a miniasperity with the substrate. Then, since the potential per atom varies over a distance of the order of  $a$ , the root mean potential per atom is of the order of the product of the force per atom and  $a$ . The integral of the potential  $V(\mathbf{r})$  over a Larkin domain of length  $L$  is the product of the root-mean-square potential per atom and the square root of the number of atoms in a Larkin domain,  $L^2/a^2$ , which gives  $(P\ell^2/c')(a^2/A_c)L$ . Multiplying by the number of Larkin domains in a miniasperity,  $A_c/L^2$ , we get  $(P\ell^2/c')(a^2/A_c) \times (A_c/L)$ , which replaces the second term in Eq. (3). The quantity  $A$  in the first term gets replaced by  $A_c$ , since we are applying the methods of Sec. II to a miniasperity, whose area of contact with the substrate is denoted by  $A_c$  rather than  $A$ . Then the condition for weak pinning becomes

$$(P\ell^2/c')(a^2/A_c) < 2(\Gamma\Gamma')^{1/2} \approx \bar{K}a^2, \quad (4)$$

where  $\bar{K}$  is a quantity of the order of  $K$  or  $K'$  in value. The far right-hand side of this expression follows from the expressions for  $\Gamma$  and  $\Gamma'$  under Eq. (2) and from the fact that  $\mathbf{u}$  varies by an amount of order  $a$  when  $(x', y', z')$  vary by an amount of order unity, which implies that the derivatives of the components of  $\mathbf{u}$  are of order  $a$ . From Eq. (4) we get the following condition for weak pinning at the miniasperity interface:

$$P < c'\bar{K}(A_c/\ell^2). \quad (5)$$

This will by itself reduce the friction by spreading the load that for the bare surfaces was supported by a few atoms that stuck out from each surface by a surface consisting of several atoms belonging to a lubricant molecule.

Next, we must examine whether each miniasperity will be in the weak-pinning limit. This will be true if under the above mean force parallel to the surface, the miniasperity is not able to sink into the minimum of its interface potential. The mean load on a pair of miniasperities in contact is of the order of  $P(A/c'N_m)$ , where  $N_m$  is the number of miniasperities along the interface between two asperities. Since the interface potential varies over a distance of order  $a$  [9], the mean interface potential for a miniasperity  $V'_0 \approx P(A/c'N_m)a$ . The condition for the miniasperity being in the weak-pinning regime is then

$$V'_0/a \ll \ell'\ell^2K(\Delta u/\ell'^2), \quad (6)$$

where  $\Delta u$  is the displacement of a miniasperity along the interface and is of the order of  $a$  and  $\ell'$  is the height of a miniasperity, which is the amount that a molecule stuck to a higher than average location on the bare surface is higher than a molecule stuck to the lowest point on the surface. Since  $A/N_m \approx \ell^2$ ,  $V'_0/a \approx P\ell^2/c'$ . Substituting in Eq. (6), we obtain

$$P \ll c'(a/\ell')K, \quad (7)$$

as the condition for the miniasperity to not sink into its net interface potential minimum. By our definition of a miniasperity,  $\ell' \approx a$ . Also,  $A_c$  is of the order of but less than  $\ell^2$ .

Thus, if Eq. (5) is satisfied, so will Eq. (7) be satisfied. When these conditions are satisfied, the miniasperity can be treated as a rigid area of contact. It is then necessary to study whether the monolayer lubricant film as a whole is in the weak-pinning regime.

Since the three-dimensional solid is now coated with a monolayer lubricant film which is likely to be more flexible than the material that the solid is made from, we must examine whether the coated solid is still able to exhibit a weak-pinning regime, characterized by a Larkin length which is as large as the interface. This would certainly not be the case for a two-dimensional solid film which is not attached to a stiffer three-dimensional solid [14]. We will for the moment assume the solid to be completely rigid (as it is being assumed to be rigid in comparison to the film). Then, the energy of the film is given by

$$E = \int_{\text{film}} d^3r \left[ K_1 \sum_{\alpha,\beta} \left\langle \left( \frac{\partial u_\alpha}{\partial x_\beta} \right)^2 \right\rangle + K'_1 \left\langle \left( \sum_\alpha \frac{\partial u_\alpha}{\partial x_\alpha} \right)^2 \right\rangle - V(\mathbf{r} + \mathbf{u}(\mathbf{r})) \delta(z) \right], \quad (8)$$

where  $K_1$  and  $K'_1$  are the elastic constants of the film. Inside the film, we choose the position dependence of  $\mathbf{u}$  to be  $u_\alpha(x', y', z')$ , where  $(x', y', z') = (x/L, y/L, z/L_f)$ , where  $L_f$  is the thickness of the film. Here,  $L_f$  represents the height of the lubricant molecules, which are assumed to be strongly attached to the solid. Substituting this assumed expression for  $u_\alpha$  in Eq. (8), we get

$$E = A \left[ K_1 \sum_{\alpha,\beta=x,y} \left\langle \left( \frac{\partial u_\alpha}{\partial x'_\beta} \right)^2 \right\rangle (L_f/L)^2 + K_1 \sum_\alpha \left\langle \left( \frac{\partial u_\alpha}{\partial z'} \right)^2 \right\rangle L_f^{-1} + K'_1 L_f \left\langle \sum_{\alpha=x,y} \left( \frac{\partial u_\alpha}{\partial x'_\alpha} L^{-1} + \frac{\partial u_z}{\partial z'} L_f^{-1} \right)^2 \right\rangle - Pa(A/cN_m)^{1/2}/L \right]. \quad (9)$$

In deriving Eq. (9), we let  $V'_0$  be the rms interaction of a single miniasperity with the substrate. Then the interaction energy of the substrate with a single Larkin domain is approximately equal to  $V'_0(c'N_m/A)^{1/2}L$ . The rms substrate potential energy of a single atom on the molecule  $V'_0$  is found by setting the mean force between a miniasperity and the substrate ( $V'_0/a$ ) [we divide by  $a$  because the distance scale of  $V(\mathbf{r})$  is of the order of  $a$ ] equal to the force pushing the surface and substrate together per miniasperity in contact,  $PA/(c'N_m)$ . As in Sec. II we are assuming hard-core interaction between individual atoms and the substrate, and therefore the latter force is comparable to the component parallel to the substrate of the force acting on a single miniasperity. This gives  $V'_0 \approx (PA/c'N_m)a$ , which when substituted in the above expression for the interaction energy of a Larkin domain with the substrate gives the last term in Eq. (9). Differentiating Eq. (9) with respect to  $L$ , we find a solution for the value of  $L$  that minimizes  $E$  (by setting this derivative with respect to  $L$  equal to zero) if

$$Pa(A/c'N_m)^{1/2} > 2K'_1 \left\langle \frac{\partial u_z}{\partial z'} \sum_{\alpha=x,y} \frac{\partial u_\alpha}{\partial x'_\alpha} \right\rangle$$

or  $P$  of the order of  $c'^{1/2}(a/\ell)K$ . The resulting Larkin length is given by

$$L = \frac{\left[ K_1 \sum_{\alpha,\beta=x,y} \left\langle \left( \frac{\partial u_\alpha}{\partial x'_\beta} \right)^2 \right\rangle + K'_1 \left\langle \sum_{\alpha,\beta=x,y} \left( \frac{\partial u_\alpha}{\partial x'_\alpha} \right)^2 \right\rangle \right] L_f}{Pa(A/c'N_m)^{1/2} - 2K'_1 \sum_{\alpha,\beta=x,y} \left\langle \frac{\partial u_\alpha}{\partial x'_\alpha} \frac{\partial u_z}{\partial z'} \right\rangle}. \quad (10)$$

We see from Eq. (10) that if the first term in the denominator is of the same order but larger than the second term, a relatively small increase in  $c$  could make the Larkin length infinite and thus put the interface in the weak-pinning limit. Once the above inequality is not satisfied,  $E$  is minimized for infinite  $L$ . This transition from finite to infinite Larkin length for a thin film attached to a rigid solid could be tested by simulations done on relatively thin films, in contrast to a three-dimensional elastic solid interacting with a disordered substrate, which cannot (as discussed in the Appendix).

The miniasperities can be modeled with a Greenwood-Williamson-like model [17], in which there is a distribution of heights of the miniasperities, consisting of the tops of lubricant molecules attached to high points on the solid surface, which get compressed under load. In order for this mechanism to work, the height variations on the surface must be small enough so that by distorting by a relatively small percentage compared with the height of the lubricant molecules attached to high points on the surfaces, a significant additional fraction of the surfaces is put in contact. To treat the miniasperities using a Greenwood-Williamson model, we must choose a probability distribution for the heights of the miniasperities which is more appropriate for this situation than the distributions used in Ref. [17]. In the present case the height distribution arises from lubricant molecules placed at high and low points on the surface located on various points along the steps illustrated in Fig. 1. Thus, the highest miniasperities are likely to have a height of order  $a$ . For the type of disorder illustrated in Fig. 1(a), there will be two possible heights for the miniasperities, one for those resulting from lubricant molecules attached to the tops of the steps and one attached to a point not on a step. Hence, we should choose a probability distribution function  $\phi(z)$ , where  $z$  represents the height of a miniasperity, (using the same notation as Ref. [17]) such that  $\phi(z)$  becomes zero for  $z$  greater than  $a$ . Thus the height distribution for miniasperities is qualitatively different from that used in Ref. [17] to describe the micron-scale height variations of a surface. Let us for simplicity choose a distribution function

$$\phi(z) = (N_1/N_m) \delta(z) + (N_2/N_m) \delta(z - a) + (N_3/N_m) a^{-1} f(z), \quad (11)$$

where  $f(z) = 1$  and is nonzero only for  $0 < z < a$ , which satisfies the above requirements for the form of  $\phi(z)$ . Here,  $N_1$  and  $N_2$  are the numbers of lubricant molecules attached between steps and on steps, respectively, for the kind of rough-



ness depicted in Fig. 1(a) and  $N_3$  are the number of molecules attached to a region with the roughness depicted in Fig. 1(c), with the condition  $N_m = N_1 + N_2 + N_3$ . We should be able to get qualitatively correct results by applying the continuum elasticity theory result for the force per unit area,  $P$ , pushing the surfaces together (i.e., treating a lubricant molecule as an elastic sphere, as was done in Ref. [17] to a first approximation). Then, following Ref. [17],

$$c' = \int_d^\infty \phi(z) dz = (N_1/N_m) \delta_{d,0} + (N_2/N_m) + (N_3/N_m) \left( \frac{a-d}{a} \right), \quad (12)$$

where  $d$  is the distance of the substrate from a reference distance from the surface. The load per miniasperity, which using the standard Hertz expression [19] (which should be accurate enough if the molecule is sufficiently large) is of the order of  $P\ell'^2/c'$ , is given by

$$\begin{aligned} P\ell'^2/c' &= CN \int_d^\infty \phi(z) (z-d)^{3/2} dz \\ &= C \left[ N_1 a^{3/2} \delta_{d,0} + N_2 (a-d)^{3/2} + (2/5) N_3 \frac{(a-d)^{5/2}}{a} \right], \end{aligned} \quad (13)$$

where  $C$  is a constant equal to  $(4/3)R_c^{1/2}K'$ , where  $R_c$  is the radius of curvature of a miniasperity and  $K'$  is the compressional elastic constant in Eq. (1). Then, if  $P > (4/3)c'K'[(N_2/N_m) + (2/5)(N_3/N_m)](a/R_c)^{3/2}$ ,  $d$  becomes zero, signifying that the miniasperities all get completely compressed, which means that all of the lubricant molecules will be in contact. When  $P$  reaches this value  $c'$  switches from  $(N_2 + N_3)/N_m$  to 1. This increase in the value of  $c'$  could switch the interface from the strong- to weak-pinning regime if the condition for strong pinning given above Eq. (10) was initially satisfied. Since for a miniasperity  $R_c \approx \ell$ , we see that if the lubricant molecule's radius  $\ell$  is sufficiently large compared to  $a$ , this critical value of  $P$  could be smaller than the critical values given in Eqs. (5) and (7), for the individual interface atoms and miniasperities to be in the weak-pinning limit. In such a case, the film as a whole could be put in the weak-pinning limit by completely compressing all of the miniasperities, while interface atoms and the miniasperities are still in the weak-pinning regime.

In principle, the same arguments could be applied to films of self-assembled chain molecules. In this case, the miniasperities are made up of several molecules which are attached to a part of the surface at which its height is constant or nearly constant. These molecules might develop gauche defects, rather than distorting elastically, under high pressure, which could modify the picture presented here somewhat.

The discussion here differs from that in Sec. II in that whereas in Sec. II, we assumed that the solid is sufficiently rigid so that  $c'$  is a constant; here we assume that  $c'$  increases as the film is compressed. This implies that even if the lubricated surface would have been in the strong-pinning limit for the value of  $c'$  for zero load, as the film is compressed  $c'$  increases, resulting in the inequality above Eq.

(10) no longer being satisfied. At that point, there will no longer exist a finite value of  $L$  which minimizes the energy (i.e., a finite Larkin length). Instead, Eq. (8) is minimized when  $L$  approaches infinity.

Although in this discussion we have neglected the elastic distortion of the bulk solid and only considered the distortion of the lubricating film, a calculation which includes the distortion of the bulk solid gave the same result, provided the film was more flexible than the bulk solid.

It has been argued in this section that it is possible for the right choice of parameters for relatively large molecules to reduce friction by three possible mechanisms operating on three length scales. First, if the molecules are sufficiently stiff, they can spread the load over more atoms in contact, resulting in these atoms being put in a weak-pinning regime (i.e., a regime in which the force exerted on these atoms by atoms from the second surface is not able to overcome the elasticity and hence the interface forces on these atoms are in random directions). Second, the miniasperities can be in a weak-pinning regime, in which the interface forces on them are in random directions. Third, the lubricant film as a whole can be put in the weak-pinning regime, and fourth, the bulk solid could be put in the weak-pinning regime.

## V. CONCLUSION

It is proposed in this article that collective pinning theory provides a possible mechanism for boundary lubrication. Two possible ways for accomplishing this are discussed. One method is due to a thin layer of small molecules that attach themselves strongly to two surfaces making up an interface between two micron-size asperities in contact, filling in atomic-distance depth holes expected to occur on such surfaces. This results in the force pushing the two surfaces together being supported over a larger area of contact, which switches the interface from the strong-pinning (i.e., high-friction) to weak-pinning (i.e., low-friction) regimes. We then propose another mechanism using larger lubricant molecules which interact strongly with the surfaces, forming a bilayer coating of the interface (i.e., each surface is coated with a monolayer of lubricant). The resulting coated surfaces reflect the atomic-level roughness of the bare surfaces, but it is argued that compression of the lubricant molecules will smooth out this roughness, allowing the force pressing the two surfaces together to be distributed over a larger area of contact, which may switch the interface from the strong-pinning (i.e., high-friction) to the weak-pinning (i.e., low-friction) regime. This "smoothing" occurs on three length scales. If the mechanism proposed in Sec. III for small molecule lubricants switches the interface to the weak-pinning limit, the friction can be reduced by a factor  $(cA_c/a^2)^{-1/2}$ , since the forces acting on the atoms at a single asperity interface will now act in random directions, leading to a reduction by this factor, which is the inverse of the square root of the mean number of atoms in contact with atoms from the second surface. If the second mechanism results put the interface into the weak-pinning limit, the friction is reduced by a factor  $(cN_m)^{-1/2}$ . It is not being claimed that the mechanism for boundary lubrication proposed here explains how all lu-

bricants reduce friction. All that is being proposed are a couple of mechanisms for reduction of friction by model lubricant molecules that are strongly attached to two surfaces which are in contact.

While it might be difficult at this stage to test experimentally whether or not the mechanisms for reduction of friction explain boundary lubrication for lubricated surfaces in actual engineering applications, it should be possible to test the phenomenon of collective pinning theory reducing friction using the surface force apparatus [22,23]. In order to accomplish this, it would be necessary to produce surfaces in the surface force apparatus with the kind of atomic level roughness discussed in this manuscript and coat them with lubricant molecules having the properties discussed in this article.

### ACKNOWLEDGMENT

I wish to thank the U.S. Department of Energy (Grant No. DE-FG02-96ER45585).

### APPENDIX: COMPARISON OF THE DISPLACEMENT FIELD FOR PERIODIC AND DISORDERED SURFACES

References [3,4] studied two thin (a few atomic layers thick) crystalline solids in contact using molecular dynamics. We have seen in this paper, however, that for disordered surfaces, the distortions of the solids, due to their interaction at the interface, can extend well into the bulk of the solids. In fact, in Sec. II, it was found that in the weak-pinning regime, the distortion extends a distance of the order of the linear dimensions of the solid. We will show here that, in contrast, for periodic solids in contact, the distortions extend only a distance of the order of the periodicity of the surface. We will simplify the problem here by considering, in place of two elastic solids, an elastic solid interacting with a rigid periodic potential, which is incommensurate with it. Our results are not expected to be qualitatively different from what we would get if we considered two elastic solids in contact. Then, the displacement of the atom at site  $\mathbf{R}_j$  in the solid due to a periodic potential acting on one of its surfaces is given by

$$\mathbf{u}_j = \sum_{\ell} \mathbf{G}(\mathbf{R}_j - \mathbf{R}_{\ell}) \cdot \mathbf{f}(\mathbf{R}_{\ell}), \quad (\text{A1})$$

where the sum over  $\mathbf{R}_{\ell}$  runs over the surface at which the substrate force  $\mathbf{f}(\mathbf{R}_{\ell})$ , representing the second solid at the interface, acts. The tensor  $\mathbf{G}(\mathbf{R}_j - \mathbf{R}_{\ell})$  represents the elasticity Green's function for the elastic solid. It is given by

$$\mathbf{G}(\mathbf{R}_j - \mathbf{R}_{\ell}) = N^{-1} \sum_{\mathbf{k}, \alpha} \frac{e^{i\mathbf{k} \cdot (\mathbf{R}_j - \mathbf{R}_{\ell})} \hat{\mathbf{e}}_{\mathbf{k}, \alpha} \hat{\mathbf{e}}_{\mathbf{k}, \alpha}}{m \omega_{\alpha}^2(\mathbf{k})}, \quad (\text{A2})$$

where  $m$  is the atomic mass and  $\omega_{\alpha}(\mathbf{k})$  is the frequency of the  $\alpha$ th phonon of wave vector  $\mathbf{k}$ ,  $\hat{\mathbf{e}}_{\mathbf{k}, \alpha}$  is a unit vector in the polarization direction of the  $\alpha$ th phonon mode of wave vector  $\mathbf{k}$  [24], and  $N$  denotes the number of atoms in the crystal. For large distances from the surface we can approximate  $\omega^2$  by its small-wave-vector approximation  $c^2 k^2$ , where  $c$  is the sound velocity. Let us consider

$$\mathbf{f}(\mathbf{R}_{\ell}) = \sum_{\sigma} i \mathbf{Q}_{\sigma} v_0 e^{i \mathbf{Q}_{\sigma} \cdot \mathbf{R}_{\ell}},$$

where  $v_0$ , a constant, and  $\mathbf{Q}_{\sigma}$ , respectively, represent the amplitude and the  $\sigma^{\text{th}}$  of the smallest reciprocal lattice vectors of the periodic substrate potential. The  $Q$ 's will be taken to have the same magnitude  $Q$ . We will take  $\mathbf{R}_{\ell}$  to lie in the plane of the substrate. Then converting the sum over  $k$  in Eq. (A2) to an integral and assuming that  $\hat{\mathbf{e}}_{\mathbf{k}, \alpha}$  is not strongly dependent on  $\mathbf{k}$  in the small- $k$  limit, we obtain

$$\mathbf{u}_j \propto \sum_{\sigma} \int dk_z \frac{e^{ik_z z} e^{i \mathbf{Q}_{\sigma} \cdot \mathbf{r}_{\parallel}}}{k_z^2 + Q^2} \propto e^{-Qz} \sum_{\sigma} e^{i \mathbf{Q}_{\sigma} \cdot \mathbf{r}_{\parallel}}, \quad (\text{A3})$$

where  $\mathbf{r}_{\parallel}$  is the projection of  $\mathbf{r}$  along the surface. Thus we see that the displacement resulting from the substrate potential only penetrates a distance  $Q^{-1}$  into the solid, which implies that simulations done on periodic solids like those done in Refs. [3,4] are adequate to capture the correct physics of the problem.

The reason that studies using thin solids are not expected to be adequate for a disordered substrate can easily be understood using a simple argument. At long distances from the substrate, where  $\omega^2$  in the denominator of Eq. (A2) can be approximated by  $c^2 k^2$ , the elasticity Green's function of Eq. (A2) is approximately proportional to  $|\mathbf{R}_j - \mathbf{R}_{\ell}|^{-1}$ . Therefore, each component of Eq. (A1) can be thought of as an electrostatic potential due to a surface charge density represented by a component of the substrate force  $\mathbf{f}$ . For a random substrate, the sum of each component of  $\mathbf{f}(\mathbf{R}_{\ell})$  over  $\mathbf{R}_{\ell}$  over a region of any size on the surface will give a nonzero value (i.e., there will be a net force on that region due to fluctuations). In our electrostatic analogy, in which each component of the substrate force represents a surface charge density, this implies that each region has a net "charge." Thus the net "potential" contribution from that region [obtained by performing the summation over  $\ell$  in Eq. (A1) over that region] is proportional to the inverse distance from that region. Since the potential in our electrostatic analogy represents the distortion  $\mathbf{u}$  due to the substrate forces acting on that region, we see that it is essential to consider thick solids when we have disordered interfaces.

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