

Effects of defects on the friction between film and substrate in a microbalance experiment

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The contribution of disorder to the force of friction between a solid film and the substrate in a microbalance friction experiment is calculated using perturbation theory. The disorder gives a much larger force of friction than a defect-free interface if the defect concentration is larger than one part in 10^5 . It is also independent of the sliding velocity. Implications for the interpretation of the experimental results are discussed.

It has been pointed out in several studies of friction between perfect crystalline interfaces that when the interfaces have incommensurate periods, the frictional force between them is many orders of magnitude smaller than for commensurate interfaces.¹⁻³ In the microbalance friction experiment,⁴ a thin film of silver or gold is evaporated onto a quartz crystal microbalance. In the traditional applications of a microbalance, the shift in the resonance frequency of the quartz oscillator as molecules are adsorbed onto the silver or gold film is used to determine the mass of the adsorbate. For the case of weakly adsorbed molecules, such as noble-gas atoms, there is some slippage which gives a measurable contribution to the damping of the oscillator caused by the friction between the adsorbed molecules and the silver or gold substrate. At high coverage, the adsorbate forms a solid film, which is generally incommensurate with the substrate. This is consistent with the model used in recent computer simulations of this experiment.⁵ One thing that was pointed out was that the calculation of the force of friction for an incommensurate interface that was done in Ref. 1 can account for the observed slip time (the time required for a film to come to rest after it has been set in sliding motion) if the phonon lifetime for Brillouin-zone boundary phonons (which is of the order of 10^{-3} s) is used in the calculation instead of the value of about 10^{-7} s which was used in Ref. 1. Nevertheless, one question remains. The main reason that the friction is so much smaller for incommensurate interfaces is because of the perfect crystalline translational symmetry of the surfaces in contact. In real situations, however, there are always defects in the interfaces which destroy this symmetry, which should result in a large increase in the force of friction over what it would be for a perfect incommensurate interface. In this brief report, I will use the perturbation theoretic methods introduced in Ref. 1 (which are valid above a critical sliding speed) to estimate the effects of defects.

In the method of Ref. 1, the rate of energy absorption by a solid acted on by a potential which is sliding by one surface of the crystal with a constant velocity was calculated. That potential was taken to be spatially periodic. Let us now consider replacing that potential by general potential (i.e., not necessarily periodic). We shall denote

the force due to this potential by $f(j'_1 a - vt, j'_2 a)$, where f is taken to point along the sliding velocity v , and $j'_1 a$ and $j'_2 a$ are the x and y coordinates of one of the atoms at the interface. Equations (4) and (6) of Ref. 1 are replaced by

$$x_{j_1 j_2 j_3} = -m^{-1} \sum_{j'_1 j'_2} \int dt' G_{j_1 j_2 j_3, j'_1 j'_2 j'_3}(t-t') f \times (j'_1 a - vt', j'_2 a), \quad (1)$$

$$F_{av} v = (mT)^{-1} \int_0^T dt \sum_{j_1 j_2} f(j_1 a - vt, j_2 a) \dot{x}_{j_1 j_2 N}. \quad (2)$$

Substituting Eq. (1) into Eq. (2) and using the Green's function given in Ref. 1, we obtain after performing some of the summations and integrals

$$F_{av} v = m^{-1} \sum_{\mathbf{k}} \frac{\gamma (k_x v)^2 |f(\mathbf{k})|^2}{[\omega_0^2(\mathbf{k}) - k_x^2 v^2]^2 + \gamma^2 k_x^2 v^2}, \quad (3)$$

where k_x is the component of \mathbf{k} along the sliding direction. If the potential is periodic, $f(\mathbf{k})$ is proportional to $\delta_{\mathbf{k}, \mathbf{Q}}$, where \mathbf{Q} is the wave vector of the potential. If \mathbf{Q} is incommensurate with the surface of the crystal, Eq. (3) reduces to the result that was obtained in Ref. 1 for the incommensurate case. If $f(\mathbf{k})$ is a smooth function of \mathbf{k} , however, Eq. (3) may be studied in the small γ limit for which it reduces to

$$F_{av} = (\pi/m) \sum_{\mathbf{k}} k_x |f(\mathbf{k})|^2 \delta[\omega_0^2(\mathbf{k}) - k_x^2 v^2]. \quad (4)$$

The sum over \mathbf{k} can be converted to an integral, as was done in Ref. 1 and evaluated following the procedure outlined in the erratum to this reference. There are two contributions to F_{av} which can be computed in this way, the contribution due to the creation of excitations in the adsorbed film and the contribution due to the creation of excitations in the substrate. For the former contribution, a two-dimensional integral is evaluated in Eq. (4), giving a contribution to F_{av} of

$$(\pi^2 N / G m v^2) |f(\mathbf{G})|^2, \quad (5)$$

where N_p is the phonon velocity, N is the number of atoms in the film and \mathbf{G} is the smallest reciprocal-lattice vector. For the latter case, a three-dimensional integral is evaluated in Eq. (4), giving a contribution

$$(2\pi^2 N / G m v_p^2) |f(\mathbf{G})|^2 (v / v_p). \quad (6)$$

In Eq. (5), f represents the disordered force due to the substrate acting on the film, and in Eq. (6), it represents the disordered force due to the film acting on the substrate. Clearly for equal for these forces, the contribution due to the energy lost due to the creation of excitations in the film dominates over the contribution from the substrate since the sliding velocity v is always much less v_p . The reason for this is that the phonon density of states at low frequency is much smaller in a three-dimensional solid, such as the substrate, than in a two-dimensional solid, such as the film, implying that there are simply more phonons available at the "washboard frequency" in the film than in the substrate. In contrast, for a defect-free incommensurate interface, the contributions to F_{av} from both film and substrate are nearly equal. The contribution to F_{av} from the excitations in the film can be estimated from Eq. (5), using the following parameters: $G \approx 10^8 \text{ cm}^{-1}$, $m \approx 10^{-22} \text{ g}$, $v_p \approx 10^5 \text{ cm/s}$, and taking $|f(\mathbf{G})|$ to be approximately equal to λ_0 from Ref. 1, the amplitude of the sinusoidal force acting on the crystal, or about 10^{-6} dyn . We obtain for $F_{av}/N \approx 10^{-8} \text{ dyn}$. This gives a slip time of $\tau = mv / F_{av} \approx 10^{-14}$, which is much smaller than the experimentally observed slip time, which implies that the substrate is not completely disordered, as we have assumed here, but rather, it is only partly disordered. The most prominent surface defects, steps, occur over large distance scales (compared to a lattice constant), and hence, according to the results of Ref. 6, if their potential is sufficiently weak for one to use perturbation theory (as must be the case for those adsorbed films which slide easily⁴), their contribution to F_{av} will be negligible compared to the contribution from atomic scale defects, such as vacancies and interstitials. Let us consider the following model of a lattice containing vacancies: Consider a square lattice of lattice constant b , on which a fraction c of the sites, chosen at random, are occupied by atoms and hence exert a force on the film (i.e., the vacancy contribution is $1-c$). The force exerted on the film atom located at the site $(j_1 a, j_2 a)$ is given by

$$\sum_{l_1, l_2} c_{l_1, l_2} f(j_1 a - v t - l_1 b, j_2 a - l_2 b), \quad (7)$$

where c_{l_1, l_2} is 1 on an occupied site and 0 on a vacant one. Repeating the procedure used to derive Eq. (5), but averaging the expression for the power dissipated over c_{l_1, l_2} , using the fact that the average of c_{l_1, l_2, l'_1, l'_2} is equal to $c(1-c)$ if $l_1 = l'_1$ and $l_2 = l'_2$ and c^2 if they are not equal, gives a contribution to F_{av} with two terms. One of them is proportional to c^2 and has the form of the contribution for two perfect incommensurate crystalline interfaces, and the second is proportional to $c(1-c)$ and is of the form of Eq. (5).

The numerical values that we obtained for Eq. (5) combined with this result for the vacancy model implies that $c(1-c)$ must be of the order of 10^{-5} in order to obtain the observed slip time of 10^{-9} sec . Furthermore, since the disorder contribution [i.e., the term proportional to $c(1-c)$] is velocity independent, this term would not contribute a term proportional to v . Therefore, in order to determine how important disorder is in these experiments, it is necessary to determine the velocity dependence of the slip time observed in the microbalance experiments more precisely. If it were found that the inverse slip time was independent of the amplitude, one would be forced to conclude that either there is almost no atomic scale disorder or the friction is due almost entirely to electronic excitations because that contribution is proportional to the velocity.^{6,7} A possible explanation of the fact that the defect contribution to the friction measured in the microbalance experiment seems to be so small could be that the adsorbed films actually consist of small solid atomic clusters. It was shown in Ref. 8 that the contribution to sliding friction due to the creation of phonon excitations in very small solids might be almost zero. This could explain the fact that the defect contribution to the friction seems to be so small, because in the calculations presented in this article, almost all of the contribution to the friction due to defects came from the creation of excitations in the adsorbed film.

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