Strongly Temperature Dependent Sliding Friction for a Superconducting Interface

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A sudden drop in mechanical friction between an adsorbed nitrogen monolayer and a lead substrate occurs when the lead passes through the superconducting transition temperature. We attribute this effect to a sudden drop at the superconducting transition temperature of the substrate Ohmic heating. The Ohmic heating is due to the electronic screening current that results from the sliding adsorbed film.

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A long-standing question in the field of nanotribology (wearless friction between well characterized surfaces) is whether frictional dissipation is dominated by the creation of phonons or the creation of electronic excitations. To study this question, an experiment using a quartz crystal microbalance (QCM) was performed in which a monolayer film of nitrogen was deposited on flat lead substrate microbalance electrodes [1]. The friction was deduced from the damping of the quartz microbalance oscillator. When the temperature of the lead was lowered below the superconducting transition temperature T_c, a rapid (nearly discontinuous) drop was observed in the monolayer friction. The drop in friction was interpreted as a reduction in the substrate electronic heating.

The electronic contribution to sliding friction experienced by a nonmetallic film sliding on a metallic surface is usually attributed to the creation of excitations of the metallic substrate electrons [2]. However, as the temperature is lowered below the substrate superconducting transition temperature, the normal (nonsuperconducting) fraction of electrons decreases gradually. The friction contribution due to the creation of electronic excitations might naively be thought to also decrease relatively gradually. On the basis of the above argument, the almost discontinuous experimental results in Ref. [1] would appear to be surprising. The solution to this puzzle lies in the nature of the electronic excitations in the superconducting substrate.

In addition to the finite frequency electron-hole pair creation mechanism of Ref. [2] for electronic friction, there exists the (virtually) zero frequency electron-hole pairs normally considered responsible for Ohmic heating in metals. Substrate Ohmic heating, induced by a sliding film, will give a sizable contribution if the atoms in the film are charged. A net charge may appear on an atom if electrons are donated to the metallic substrate. Such a charge redistribution results in a monolayer surface dipole moment. The surface dipole charge structure induces Ohmic heating resulting from the electric currents in the metallic substrate induced by the sliding monolayer film. The electronic charge redistribution follows the film atoms as they move over the substrate surface [3]. Unlike the mechanism of Ref. [2], this mechanism vanishes very quickly as electron pairs form in the superconductor, i.e., practically right at T_c. We propose this mechanism as the reason for the rapid drop in the friction observed in the experiment of Dayo and Krim.

Popov [4] proposed a mechanism for the sudden drop in the electronic friction, based on a phenomenological model in which the conduction electrons of the substrate are modeled as a single viscous fluid dragged along by the sliding film, whose viscosity does not change when the temperature drops below T_c. It is well known, however, that a correct phenomenological description of superconductivity requires that one use a two fluid model [5,6].

It is well established that the adsorption of a nonmetallic film results in an observed change [7] in the work function (i.e., electron chemical potential) of a metallic substrate. This observation has been interpreted in terms of transfer of electronic charge from the film to the metallic substrate, or equivalently as a surface dipole moment per unit area of the film atoms. This picture produces the electric fields needed for the mechanism in Ref. [3].

In Ref. [3], the force of friction acting on a charged atom whose center moves parallel to the substrate a distance Z above its surface is defined as the interaction of the atom with the electric field due to its image charge. The image electric field is

$$\mathbf{E}_{\text{image}} = -q\eta\left(i\frac{\partial}{\partial t}\right)\left\{\frac{(\mathbf{r}-\mathbf{R}_i)}{|\mathbf{r}-\mathbf{R}_i|^3}\right\}, \tag{1}$$

where the operator $\eta(i\partial/\partial t)$ is constructed by the substitution of $i(\partial/\partial t)$ for ω in the formula $\eta(\omega) = [\epsilon(\omega) - 1]/[\epsilon(\omega) + 1]$. The dielectric constant $\epsilon(\omega)$ is related to electrical conductivity $\sigma(\omega)$ via $\epsilon(\omega) = 1 + [4\pi i\sigma(\omega)/\omega]$. $\mathbf{R}_i$ is the location of the center of the electrical image of the charged atom. If $\mathbf{R} = (X, Y, Z)$ is the location of the center of the charged atom, then $\mathbf{R}_i = (X, Y, -Z)$. The conductivity of the substrate can be represented by a Drude model. For the superconducting state,

$$\sigma(\omega) = \sigma_n - \left(\frac{c^2}{i\Lambda^2\omega}\right), \tag{2}$$

where $\sigma_n = (\frac{n_n e^2\tau_s}{m})$ and $\frac{c^2}{\Lambda^2} = \frac{n_s e^2}{m}$ where Λ is the London penetration depth and n_n and n_s are the number

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densities of normal and superconducting electrons, respectively, and τ_s is the scattering time for the normal electrons. The time Fourier transforms of smooth functions of $\mathbf{R}_i(t)$ are expected to be peaked around a frequency of the order of the sliding velocity of the atom divided by the height h of the center of the atom above the substrate surface, an atomic length scale (i.e., about 10^{-8} cm) [1]. The only other frequencies in the problem in the superconducting state are (in Gaussian units) σ_n (which for $n_n \approx 10^{23}$ cm^{-3} and $\tau_s \approx 10^{-14}$ s^{-1} is of the order of 10^{17} s^{-1}) and c/Λ. (In the normal state, the only frequency is σ_n [3].) We can estimate Λ from Eq. (34.9) in Ashcroft and Mermin's book, using parameters for lead from the table on page 38 in that book [8]. We find that $c/\Lambda \gg V/h$ if $n_s/n_n \gg 0.24 \times 10^{-16}$. Thus, we are justified in expanding the operator $\eta(i\partial/\partial t)$ in powers of $i(\partial/\partial t)$ once the temperature is slightly below T_c. This is accomplished by expanding $\eta(\omega)$ in powers of ω and substituting $i\frac{\partial}{\partial t}$ for it. It gives us a series expansion of the friction in powers of the sliding velocity. For the superconductor, up to third order

$$\eta(\omega) \approx 1 + (1/2\pi)(\Lambda^2/c^2)\omega^2 + i(\sigma_n\Lambda^4/2\pi c^4)\omega^3. \tag{3}$$

The force of friction acting on the atom is equal to the product of the charge on the atom with the components of $\mathbf{E}_{\text{image}}$ parallel to the surface, evaluated at $\mathbf{r} = \mathbf{R}$. In order to find $\mathbf{E}_{\text{image}}$, let us operate with Eq. (3) with $i(\partial/\partial t)$ substituted for ω on the curly bracketed factor in Eq. (1), and then set $\mathbf{r} = \mathbf{R}$, using the fact that $\mathbf{R} - \mathbf{R}_i = 2h\hat{\mathbf{z}}$ and $\frac{\partial \mathbf{R}_i}{\partial t} = \mathbf{V}$, taken here to be along the surface and constant. [In the microbalance experiment the substrate and film are accelerating with respect to each other. Since the frequency of oscillation of the quartz crystal is about 10^6 Hz compared to the inverse slip time of 10^9 s^{-1}, however, the terms proportional to the acceleration that result from Eq. (1) can be considered negligibly small compared to the result that one obtains by assuming the velocity to be constant.] The second and third time derivatives of the expression inside the curly brackets in Eq. (1) are given by

$$(\partial^2/\partial t^2)\{\cdots\} = -\frac{4\mathbf{V}[\mathbf{r} - \mathbf{R}_i(t)] \cdot \mathbf{V} + 3[\mathbf{r} - \mathbf{R}_i(t)]v^2}{|\mathbf{r} - \mathbf{R}_i(t)|^5} - \frac{15[\mathbf{R} - \mathbf{R}_i(t)]\{[\mathbf{r} - \mathbf{R}_i(t)] \cdot \mathbf{V}\}^2}{|\mathbf{r} - \mathbf{R}_i(t)|^7}, \tag{4a}$$

$$(\partial^3/\partial t^3)\{\cdots\} = \frac{7V^2\mathbf{V}}{|\mathbf{r} - \mathbf{R}_i|^5}, \tag{4b}$$

plus terms with $\mathbf{r} - \mathbf{R}_i(t)$ or $[\mathbf{r} - \mathbf{R}_i(t)] \cdot \mathbf{V}$ in the numerator. When we set $\mathbf{r}$ equal to $\mathbf{R}(t)$ in Eq. (4a), the only nonzero terms are directed normal to the film, and hence will not contribute to the friction. When we set $\mathbf{r}$ equal to $\mathbf{R}(t)$ in the third time derivative of the curly bracketed expression in Eq. (1), only the term shown explicitly in Eq. (4b) is nonzero and directed parallel to the surface of the film. Substituting this expression into Eq. (1) gives

$$F = \left(\frac{5n_n\tau_s V}{16n_s h}\right)^2 \left\{\left(\frac{q^2 m}{2\pi n_n e^2 \tau_s}\right)\left(\frac{\mathbf{V}}{h^3}\right)\right\}. \tag{5}$$

Although the interpretation of QCM friction measurements in Ref. [1] is based on linear response theory [9], there is no *a priori* reason to believe that the friction between the substrate and film must be linear in the velocity. In fact, theoretical methods have been proposed to interpret QCM experiments when the friction is nonlinear [10] and there have been QCM friction measurements which give nonlinear friction [11]. The term in curly brackets in Eq. (5) is the expression for the force of friction for the normal state found in Ref. [3]. Since $\tau_s V/h \approx (10^{-13}\ \text{s}/10^{-8}\ \text{cm})(1\ \text{cm/s}) = 10^{-6}$, $\mathbf{F}$ in the superconducting state will be much smaller than the normal state force of friction as long as $n_n/n_s \ll 10^6$. This condition holds true even for T just slightly below T_c. The quantity n_s occurs in the denominator of Eq. (5) because our expansion of $\eta(\omega)$ is valid only for n_s not equal to zero. We see here, however, that the expansion can be valid for T only slightly below T_c (i.e., $n_s/n_n \gg 10^{-6}$). Alternatively the first bracketed expression in Eq. (5) can be written as $\frac{\sigma_n \Lambda^2 V}{c^2 h}$.

In Ref. [2], the "slip time" τ of a film was measured, where τ is defined as the time required for the velocity of a sliding film to decay to $1/e$ of its initial value. τ^{-1} was found to decrease by 6.6×10^7 s^{-1} on dropping below T_c. We assume that the lead resistivity sample used in Ref. [1] and the lead substrate used in the microbalance both have a thickness of 1500 Å, and that the sample slabs were 3 mm × 3 mm squares [12]. The resistance for the lead sample given in the experiment was 0.04 Ω just above T_c. Thus the normal state resistivity was 6×10^{-7} Ω cm. Equivalently, the conductivity was 1.5×10^{18} s^{-1} in Gaussian units. On the basis of Ref. [2], the change in τ^{-1} at T_c is given by

$$\Delta(\tau)^{-1} = \left(\frac{Z^2 e^2}{16\pi m \sigma h^3}\right) \approx \frac{1^2(4.8 \times 10^{-10}\ \text{esu})^2}{16\pi(4.6 \times 10^{-23}\ \text{g})(1.5 \times 10^{18}\ \text{s}^{-1})(10^{-8}\ \text{cm})^3}, \tag{6}$$

for nitrogen sliding on lead. In Eq. (6), 4.6×10^{-23} g is the mass of a nitrogen molecule, and $h \approx 10^{-8}$ cm is half of a lattice constant for solid nitrogen, which we take as its distance above the substrate. Then we obtain $\Delta(\tau)^{-1} = 6.6 \times 10^7$ s^{-1}, which is consistent with the results of Ref. [1].

Since the picture presented here is based on a model in which the film atoms are charged, one might think that the resulting corrugation potential might be much larger than what one would obtain for Lennard-Jones potentials (i.e., for uncharged atoms) [13] since the Coulomb interactions are so much larger than Lennard-Jones interactions. This might invalidate the assumption in our model and in Ref. [13] that the substrate potential is small. Therefore, it is necessary to make some rough estimates of the corrugation potential (i.e., the dependence of the potential of interaction between a film atom on the surface and the ions inside the metal substrate on the position of the film atom on the surface) in order to determine whether our picture makes physical sense. We wish to determine whether the small values of the corrugation needed to explain experiments done with the QCM [13] can occur when the film atoms are charged. If the adsorbate atoms are charged, it is expected that the main contribution to the corrugation potential results from the screened Coulomb interaction between the adsorbate atom and the ions. As a rough approximation to this interaction, let us use the Thomas-Fermi [14] approximation to the interaction between an adsorbate atom and an ion in the metal, $(q_1 q_2 e^{-k_s r}/r)$, where q_1 and q_2 are the charges of the two atoms, and r is the distance between them. In these estimates, it is assumed that the adsorbate atom lies right on the surface and is "bathed" in conduction electrons. The inverse screening length $k_s \approx [4\pi e^2 g(\epsilon_F)]^{1/2}$, where $g(\epsilon_F)$ is the density of states at the Fermi level. Using the free electron density of states appropriate for lead [15], and choosing the effective mass of an electron in lead to be $m = 1.3 m_e$, we obtain $k_s \approx 2.2/\text{Å}$. For these estimates, the metal ions near the surface were taken to lie on a triangular lattice in which each ion has a charge $4e$ (for lead). We used a lattice constant 2.9 Å. The interaction between an adsorbate atom, located ≈4 Å above the center of the plane along which the centers of 900 substrate ions lie, and the substrate ions was then numerically summed over the positions of the ions in the lattice. This interaction was found for three positions of the adsorbate atom: (1) above the triangle formed by three near neighbor ions in the lattice, (2) above one of the ions, and (3) above the line joining two nearest neighbor ions. The potential of interaction was found to be, respectively, 17.6, 22.3, and 17.9 meV. Thus, we conclude that the corrugation potential is of the order of a few meV. This value is comparable to that used to successfully simulate QCM measurements of friction for rare gas atoms adsorbed on a noble metal substrate [13].

We conclude that the sudden drop in the sliding friction acting on a nitrogen film as it slides over a lead substrate, as the lead drops below its superconducting transition temperature, can be attributed to Ohmic heating induced in the metal by the sliding nitrogen film.

After this Letter was submitted, we became aware of a paper in preprint form by Novotny and Velicky [16], which proposes a similar mechanism for the results reported in Ref. [1].

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