Dynamical sticking of a solid ^{4}He film with superfluid overlayer

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(Received 14 February 2009; published 20 May 2009)

We observed a phenomenon in a quartz-crystal microbalance experiment for solid ^{4}He films with liquid overlayer on graphite. The solid atomic layers underneath the superfluid layer stop slipping from the oscillating substrate when the superfluid density grows up to a certain magnitude. This phenomenon can be explained by a mechanism in which the mass transport caused by the motion of edge dislocations in solid atomic layers is canceled by counterflow of the superfluid overlayer.

DOI: 10.1103/PhysRevB.79.172503 PACS number(s): 67.25.dp, 68.35.Af, 68.60.−p

An atomically thin film on a solid substrate exhibits various unique properties reflecting the geometry of the substrate and providing a number of attractive topics in physics for many years. One of them is the properties of phase transitions. Because of the low dimensionality, fluctuation effects play an important role and the long-range order is lost by thermal activation of topological defects.[1] The superfluidity of ^{4}He films is an example of topological long-range order and the superfluid density drops discontinuously to zero at the transition due to the unbinding of quantized vortex pairs.

Recently, nanotribology has been added to the subjects of study on the atomically thin film.[2–4] A He film on graphite is an ideal system for a study on the sliding friction. The surface of graphite is atomically flat and the film grows in layers up to more than five atoms thick.[5,6] We have measured the sliding friction of ^{4}He films on Grafoil (exfoliated graphite) using the quartz-crystal microbalance (QCM) technique and found that thin solid films, or two- and three-atom-thick films, undergo slipping from the oscillating substrate below a certain temperature, T_S. Some properties for thin solid films have already been reported.[7,8]

In this Brief Report, we report a phenomenon for thicker four-atom-thick films, which are composed of solid atomic layers and the *superfluid* overlayer. One might think that the dynamics of solid atomic layers would be decoupled from the overlayer when the latter becomes superfluid. What we have observed is, however, opposite to this naive guess. When the superfluid density grows up to a certain magnitude, the solid atomic layers stop slipping. The observed phenomenon can be explained by a dynamical mechanism through an interplay between the solid atomic layers and the superfluid overlayer.

The QCM technique enables us to measure a mass decoupling from an oscillating substrate and is applied to measurements of the sliding friction and the superfluidity of ^{4}He films. A change in resonance frequency, Δf, is expressed by

$$\frac{\Delta f}{f_R} = -\frac{\sigma_{\mathrm{eff}}}{M}, \qquad (1)$$

where f_R is the resonance frequency, M is the areal mass density of the quartz crystal, and σ_{eff} is the areal mass density coupled with the oscillating substrate. An increase in frequency means that the film undergoes decoupling from the oscillating substrate.

The experimental procedure has already been reported in detail.[7] In the present experiments, the resonator was a 5.0 MHz AT-cut quartz crystal. Grafoil was bonded on the Ag electrodes of the crystal after a 300-Å-thick Ag film was deposited. From the change in frequency from bare Ag to Grafoil/Ag electrodes, the average thickness of Grafoil on each electrode was estimated to be about 3.3 μm. Because the specific surface area of Grafoil is 15.4 $\mathrm{m^2/g}$, the effective surface area of the Grafoil/Ag electrode was about 56 times larger than that of the bare Ag electrode. The sensitivity for the mass loading of ^{4}He corresponds to 4.2 Hz/(atoms/nm^2). The crystal was mounted in the sample cell after heating in a vacuum at 130 °C for 5 h. During transport it was briefly (1 min or less) exposed to air. Then, the cell was evacuated and cooled to 4.2 K. To minimize the effect of desorption, Grafoil disks were put on the bottom of the cell. The resonance frequency was measured using a transmission circuit. In the circuit, the crystal was placed in series with a coaxial line connecting a signal generator and a homemade phase-sensitive detector or an rf lock-in amplifier.

Figure 1 shows the temperature dependence of the resonance frequency for several ^{4}He samples of different areal densities. All data were taken during cooling with the oscillation amplitude being fixed at 0.4 nm. At any temperature, the frequency does not greatly change from no coverage to 10 atoms/nm^2 and decreases suddenly at around 11 atoms/nm^2. As the areal density increases further, it decreases gradually again. Between 14 and 20 atoms/nm^2, a characteristic temperature T_S appears and the frequency does not change much below T_S, i.e., ^{4}He films undergo slipping from the oscillating substrate below this temperature. In our previous paper, we analyzed these results using the multilayer model and found that the second atomic layer of solid (2ALS) slips below T_S relative to the first atomic layer (1ALS).[8] T_S shifts lower until the third layer promotion of 20.4 atoms/nm^2. Above 22 atoms/nm^2, the frequency increase below T_S becomes remarkable. Above 34 atoms/nm^2, the decoupling is clearly observed at T_C, which is in good

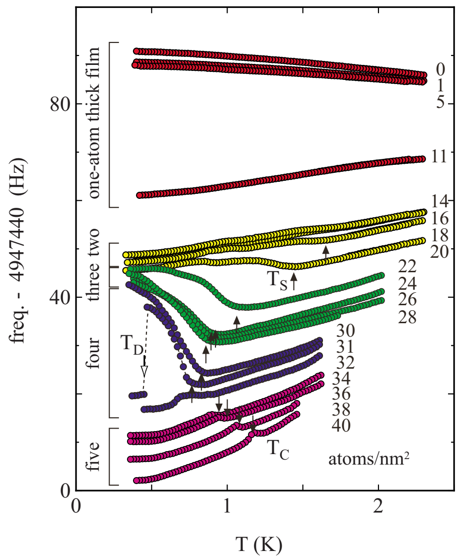

FIG. 1. (Color online) Variation in the resonance frequency for various ^{4}He areal densities as a function of temperature. Numbers represent areal densities of ^{4}He in the unit of atoms/nm^2. Arrows indicate T_S, T_C, and T_D.

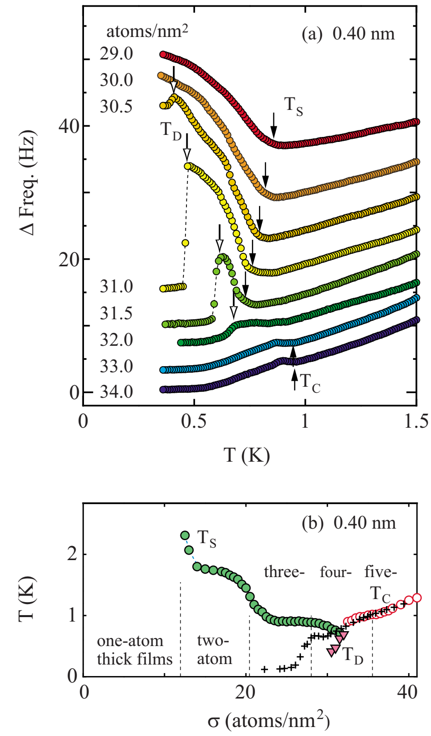

FIG. 2. (Color online) (a) Variation in the resonance frequency for various ^{4}He areal densities of four-atom thick film as a function of temperature. For clarity, the sets of data are shifted vertically. Figures represent areal densities of ^{4}He in the unit of atoms/nm^2. Arrows indicate T_S, T_C, and T_D. (b) T_S (open circles), T_C (open squares), and T_D (solid triangles) vs areal density. Dashed lines indicate the layer promotions of ^{4}He films. Crosses are the superfluid onset temperature obtained from torsional oscillator measurements (Ref. 6).

agreement with the superfluid onset temperature obtained from torsional oscillator measurements, as shown in Fig. 2(b).[6]

We observed a phenomenon in the resonance frequency below the areal density at which the superfluid onset is clearly observed. Typically, at 31 atoms/nm^2 in Fig. 1, an increase in the frequency below T_S is terminated abruptly at a lower temperature T_D. Figure 2(a) shows a detailed areal-density dependence of a four-atom thick film in the crossover region from slippage to superfluidity. Below 30.0 atoms/nm^2, the frequency increases largely below T_S. On the other hand, a small stepwise decoupling due to superfluidity is observed at T_C above 33.0 atoms/nm^2. Between these two areal densities, the frequency increases below T_S in the same manner as the lower areal density and drops at T_D down to the extrapolated curve from high temperatures. As the areal density increases, T_D shifts toward higher values. It is impressive that, as shown in Fig. 2(b), T_C appears at the areal density where T_S and T_D merge and thus the slipping region disappears. T_D is always located in the region at which the superfluid fraction exists according to the torsional oscillator measurements. This demonstrates that the superfluidity of the overlayer is crucial to the suppression of the slippage. In fact, in the case of ^{3}He film, we never observed such an abrupt suppression up to five-atom-thick film.[9]

We examined the effect of the oscillation amplitude on the resonance frequency. Figure 3 shows the temperature dependence of the resonance frequency at 31 atoms/nm^2 for various oscillation amplitudes.[10] At 0.77 nm amplitude, the frequency increases monotonously below T_S=0.75 K and a suppression is not observed. At 0.52 nm, an increase in frequency below T_S is terminated at $T_D \sim 0.5$ K. As the oscillation amplitude decreases further, T_D approaches T_C. Finally, T_S and T_D are smeared out and the superfluidity appears at T_C=0.75 K. This means that a large oscillation amplitude inhibits the mass sticking.

Before proposing the mechanism of the sticking phenomenon, we first discuss the slippage of solid atomic layers. As mentioned above, 2ALS slips on a periodic potential provided by 1ALS below T_S while 1ALS sticks to the substrate.[8] Quantities to characterize the slippage are the yield stress σ_y and the shear stress due to oscillation $F=\rho_2 m A \omega^2$, where ρ_2 is the areal number density of 2ALS, m is the mass of the ^{4}He atom, and A is the oscillation amplitude at angular frequency ω. $F > \sigma_y$ is a necessary condition for 2ALS to slip on 1ALS. In the present experiments, $F=10^{-1}-10^{-2}$ Pa. It was found that 2ALS does not slip on 1ALS *uniformly* regardless of the commensurability. When the commensurability is perfect, σ_y is expected to roughly coincide with the shear modulus of the bulk crystal, which is approximately 1×10^7 Pa.[11] In contrast, when 2ALS is uniform and incommensurate to 1ALS, σ_y can be zero. We stress, however, that this does not mean that slippage occurs at an infinitesimally small F. The finiteness of the spreading pressure $P_\parallel$ implies an edge of the graphite platelet pushing back the layer to spread out. $P_\parallel$ is estimated as on the order of 10^6 Pa.[12] The condition of uniform slippage is thus $F > \max(P_\parallel, \sigma_y)$, which is obviously not satisfied.

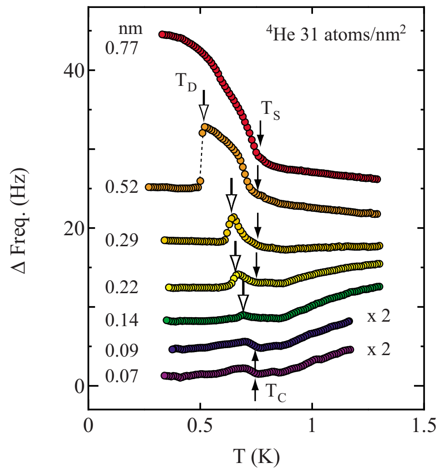

FIG. 3. (Color online) Variation in the resonance frequency at the areal density of 31 atoms/nm^2 for various oscillation amplitudes. Numbers represent the oscillation amplitude of the crystal in the unit of nm. Arrows indicate T_S, T_D, and T_C. For clarity, the data sets are shifted vertically and the two data sets at the bottom are expanded twice on the vertical scale.

It should be noted that 2ALS can slip *nonuniformly* under a more relaxed condition. It is well known that in the case of a metal the stress on an edge dislocation to move (the Peierls stress) can be on the order of 10^{-7} times the shear modulus.[13] Besides, the motion of edge dislocations generates only a localized mass transport and then $F > \sigma_y$ is enough to realize the slippage of 2ALS regardless of the value of $P_{\|}$.

We propose a possible mechanism of the sticking phenomenon by considering an interplay between 2ALS and a superfluid overlayer. Grafoil is composed of atomically flat crystalline platelets of typically $\ell = 45$ nm in diameter.[14] Connectivity of the platelets can be considered poor because χ factor, which is the fraction of the superfluid locked to the oscillating substrate, is very large.[15] We thus model the whole system as an ensemble of ^{4}He film on an isolated single platelet of graphite. As for the deformation of 2ALS on the platelet, only the in-plane deformation is allowed because of a large adsorption pressure and it is expressed by a two-dimensional displacement vector. Then, 2ALS is described by the two-dimensional Frenkel-Kontorova model. If the areal number density mismatches between 2ALS and 1ALS, an edge dislocation is spontaneously created.[16] In fact, at the third layer promotion, the number density of 2ALS is 0.71 times smaller than that of 1ALS. Then, 2ALS slips easily on 1ALS due to the motion of edge dislocations. The slippage generates a densified and rarefied region pair of 2ALS as seen in Fig. 4.

When the liquid overlayer becomes superfluid, an interplay between 2ALS and the superfluid overlayer will appear. ^{4}He atoms of 2ALS dissolves in the overlayer at the densified region, while at the rarefied region ^{4}He atoms of the overlayer condense to 2ALS. Then, between the pair regions, the supercurrent of the overlayer transports ^{4}He atoms. This process cancels the mass transport of 2ALS due to slippage and the film substantially sticks to the substrate, which is illustrated schematically in Fig. 4. It is well known that a

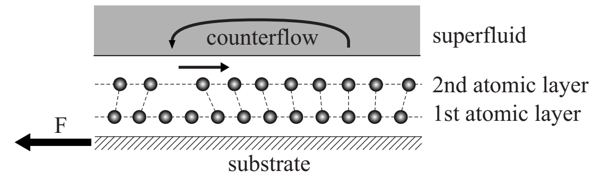

FIG. 4. Schematic drawing of the mass cancellation due to superfluid counterflow to the edge dislocation motion.

similar coupling is realized at the solid-liquid interface of ^{4}He. The periodic displacement of the interface propagates as a crystallization wave and the supercurrent in the liquid transports ^{4}He atoms.[17,18]

This mechanism can be formulated as follows.[19] For simplicity, we consider the one-dimensional case: the slippage of 2ALS occurs in the x direction in which the external oscillation is driven. The displacement of the nth atoms is expressed as $q_n - n\lambda$, where λ is the spatial periodicity of the corrugate potential of 1ALS. We here define the phase displacement $\phi_n = 2\pi(q_n - n\lambda)/\lambda$. Taking a continuum limit, an excess number density $\delta\rho$ due to the slippage in 2ALS is expressed by

$$\delta\rho = \frac{1}{2\pi}\frac{\lambda}{d}\frac{\partial\phi}{\partial x}. \quad (2)$$

The excess number density is released through the supercurrent of the overlayer, $\delta\dot{\rho} = -\rho_s \,\mathrm{div}\, \vec{v}_s$, where ρ_s is the superfluid number density and $\vec{v}_s$ is the superfluid velocity. This relation along with Eq. (2) leads to

$$\dot{\phi} = -2\pi\frac{d}{\lambda}\rho_s v_s. \quad (3)$$

This equation means that the effective mass of ϕ is significantly enhanced.[20] In addition, the total momentum density g is

$$g = \frac{1}{2\pi}\frac{\lambda}{d}\dot{\phi} + \rho_s v_s = 0, \quad (4)$$

i.e., the mass transport of 2ALS is canceled by counterflow of the superfluid overlayer. The whole layers thus dynamically stick to 1ALS.

Finally, we comment on some criteria of this dynamical sticking. One is on the superfluid critical velocity v_{sc}. The densified and rarefied regions of 2ALS must oscillate with the angular frequency ω, at most, in the distance ℓ. Then, the supercurrent should follow the oscillation and the response time is limited by v_{sc}. Then, the condition of $\omega < \pi v_{sc}/\ell$ is necessary. Another criterion is related to the momentum density. When 2ALS slips completely on 1ALS, the momentum areal density due to slippage becomes $\rho_2 m A\omega$ and should be canceled by the supercurrent. The momentum areal density in the superfluid is limited by the superfluid critical velocity and the superfluid density, and $\rho_2 m A\omega < \sigma_s v_{sc}$, where σ_s is the superfluid areal mass density. If we assume that the observed abrupt cessation of slippage at T_D is related to these criteria, v_{sc} should be 10^{-2}–10^{-1} m/s when appropriate values are substituted for the other quantities. This is a plausible value of the superfluid critical velocity near T_C.[21] We note that the latter criterion also shows that a large oscillation amplitude inhibits the dynamical sticking, which qualitatively explains the experimental observations.

In summary, we carried out a QCM experiment for ^{4}He films on Grafoil composed of solid atomic layers and a liquid overlayer and found that the slippage is suppressed when the overlayer becomes superfluid. This can be explained by the cancellation of the mass transport of solid atomic layers due to counterflow of the superfluid. A quantitative study of this scenario is being developed to compare with the experiment.

This work was partly supported by MEXT under KAKENHI Grants No. 20540351 and No. 17540313 of Japan. One of the authors (N.H.) acknowledges Grant-in-Aid for JSPS.

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