Slippage of Nonsuperfluid Helium Films

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We measured the mechanical response of ^{3}He and ^{4}He films on an oscillating substrate using an ultrasonic technique. Up to the coverage at which the fluid state appears at absolute zero, a part of the nonsuperfluid ^{3}He and ^{4}He films underwent slipping relative to the substrate at low temperatures. From the temperature dependence of the slippage, it was found that a thermally activated process plays an important role in the frictional force of this system.

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Do adatoms move in concert with a lateral oscillating substrate under any conditions? This question is closely related to the atomic-scale mechanism of sliding friction, which is an important current topic of research [1]. Krim and her colleagues [2,3] measured the mechanical response of noble-gas monolayer films on a noble-metal substrate at 77 K using the quartz-crystal microbalance (QCM) technique. They found that the adsorbate slips with the "slip time" on the order of 10^{-9} s.

Simulation using the modified Frenkel-Kontorova model showed that the nature of the sliding friction depends strongly on the interatomic force between the adsorbate and the substrate [4]. Since the interatomic force of a helium atom is very weak, one may expect helium films to slip easily on a substrate. Although it is well known that the superfluid fraction of ^{4}He films decouples from an oscillating substrate, it is unclear whether nonsuperfluid helium films slip on the substrate [5]. It is therefore of great interest to determine which conditions, i.e., coverage, temperature, etc., may allow the slippage of nonsuperfluid helium films.

Thus motivated, we investigated the mechanical response of both superfluid and nonsuperfluid helium films on a two-dimensional porous material, hectorite, and found that nonsuperfluid helium films slip from the oscillating substrate under certain conditions. In the present experiments, the ultrasonic technique was adopted. Precise measurements of the sound velocity and attenuation with a high carrier frequency enabled us to study the mechanical response of one- and two-atom thick films.

The ultrasonic technique was used earlier for detection of the superfluidity of ^{4}He films on a porous material, Vycor glass [6]. Decoupling of the superfluid fraction from the oscillation causes an increase in sound velocity since the sound velocity of the porous material is related to the effective density ρ_{sub} and the elastic stiffness c, as $v_0 = \sqrt{c/\rho_{\text{sub}}}$. The increase in sound velocity is attributed to the vanishing of the frictional force acting on the superfluid fraction, and is proportional to this fraction, as $\Delta v/v_0 = (1 - \chi_s)\sigma_s S/2\rho_{\text{sub}}$, where σ_s is the areal density of the superfluid component, S is the surface area per unit volume, and χ_s is the fraction of the superfluid which remains locked to the substrate.

Here, let us suppose that nonsuperfluid helium films adsorbed on a two-dimensional porous material are *not* rigidly locked to the substrate, and have a weak frictional force. At the atomic scale, the frictional force acting on the adsorbate per unit area F_f depends on the sliding speed, and is expressed as

$$F_f = -\frac{\sigma}{\tau} V, \tag{1}$$

where σ is the areal density of the adsorbate and V is the sliding speed. The slip time τ is the relaxation time which characterizes the rate of momentum transfer between the adsorbate and the substrate. The frictional force and the inertia of the adsorbate change the sound velocity and the attenuation of the porous material. We obtain the decrease in sound velocity as

$$\frac{\Delta v}{v_0} = -\frac{\Delta\rho}{2\rho_{\text{sub}}}\left\{\chi + \frac{1}{1+\omega^2\tau^2}(1-\chi)\right\}, \tag{2}$$

where $\Delta\rho$ $(= \sigma S)$ is the effective density of the adsorbate, ω is the angular frequency of ultrasound, and χ is the fraction of the adsorbate which remains locked to the substrate. The excess attenuation caused by the adsorbate motion arises as

$$\Delta\alpha = \frac{\Delta\rho\,\omega}{2\rho_{\text{sub}}v_0}\frac{\omega\tau}{1+\omega^2\tau^2}(1-\chi). \tag{3}$$

Similar expressions of the acoustic impedance were used

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for a study on the interfacial friction using QCM [7]. As the normal direction of the substrate surface in the porous material is distributed uniformly, χ becomes 1/3. In the derivation of Eqs. (2) and (3), we assume that the elastic stiffness is unchanged [8].

The mechanical response of the adsorbate is determined by the value of $\omega\tau$. If $\omega\tau \ll 1$, the adsorbate appears to be locked to the oscillating substrate, and the decrease in sound velocity is proportional to the mass of the adsorbate. As $\omega\tau$ increases, the adsorbate slips, and then an attenuation peak and a rapid increase in sound velocity are observed at $\omega\tau \sim 1$. When $\omega\tau \gg 1$, the frictional force vanishes and the adsorbate undergoes decoupling from the oscillation.

Hectorite used for the substrate in the present experiments is a two-dimensional porous material made of layered silicate. It has plane spaces of ~17–20 Å between smectite layers of 9.6 Å thickness, which are supported by smectite fragments [9,10]. The surfaces of the smectite layer are atomically flat, and are composed of oxygen triangles of SiO_4 tetrahedra. Wada *et al.* [11] performed measurements on the heat capacity, the isosteric heat of adsorption and the torsional oscillator for both ^{3}He and ^{4}He films on hectorite, and discussed the motional state of the adsorbate. The first layer completion of ^{3}He and ^{4}He takes place at $n_{c3} = 0.10 \pm 0.01$ atoms/Å^2 and $n_{c4} = 0.110 \pm 0.002$ atoms/Å^2, respectively. Above $n_{d3} = 0.131 \pm 0.006$ atoms/Å^2 for ^{3}He the heat capacity at low temperatures becomes large due to the appearance of the Fermi liquid, and above $n_{d4} = 0.146 \pm 0.002$ atoms/Å^2 for ^{4}He the superfluidity is observed. That is, helium films up to n_{d3} and n_{d4} are solidlike, or "inert" at low temperatures. Although no distinctive feature of the phase transition between solid and fluid states is observed, helium films are thought to be fluidlike at high temperatures. As the coverage exceeds n_{d3} and n_{d4}, a part of the second layer remains fluid at absolute zero.

For ultrasonic measurements of helium films adsorbed on hectorite, a cylindrical pellet measuring ϕ12.4 mm × 7.9 mm was fabricated by pressing together hectorite powder and metal powder, and 10 MHz longitudinal $LiNbO_3$ transducers were then bonded to both ends of the pellet with silicone adhesive. The density of the pellet was 1.710 ± 0.008 g/cm^3 and the specific surface area was found to be 244 ± 2 m^2/g by the N_2 vapor pressure isotherm at 77 K. After fabrication, the pellet was dried and evacuated to remove water in mesopores at 150 °C for 3 h before it was mounted in the experimental cell. To calibrate the velocity decrease by the mass of the adsorbate, we measured this decrease due to nitrogen at 77 K. Nitrogen adsorbed on hectorite is bound strongly to the hectorite surface. The rate of velocity decrease was 34.7 ± 0.3%/(molecules/Å^2) for nitrogen [12]. Thus, taking the molecular weight into account, the rate for ^{3}He and ^{4}He is estimated to be 3.7%/(atoms/Å^2) and 5.0%/(atoms/Å^2), respectively.

We measured the sound velocity and attenuation of the pellet with ^{3}He and ^{4}He films down to 100 mK. In most of the present experiments, the amplitude of the transducer was about 0.1 Å, and the maximum oscillating speed corresponded to 4×10^{-4} m/s. However, in several coverages we confirmed that there was no large amplitude dependence up to about 6 Å.

We found that the adsorption of helium atoms causes a change in the sound velocity and attenuation of the pellet. Figure 1 shows the velocity shift relative to the bare pellet as a function of coverage at 150 mK and the temperature of the velocity minimum before the desorption effect appeared, which took place at 7 ~ 18 K. The minimum of the velocity shift decreased proportionally with increasing coverage, and showed a difference of molecular weight between ^{3}He and ^{4}He. The rate of velocity decrease was about 90% of the expected value from nitrogen. It is concluded that almost all helium adatoms at high temperatures move in concert with the oscillating substrate. On the other hand, when the temperature was lowered, the sound velocity increased and at 150 mK the velocity did *not* decrease due to the mass of the adsorbate. Although monolayer films show an interesting behavior, in this paper we confine ourselves to the coverages above 0.1 atoms/Å^2, or ^{3}He and ^{4}He bilayer films.

Figure 2 shows the sound velocity of ^{3}He and ^{4}He films above 0.1 atoms/Å^2 as a function of temperature. First, we mention briefly the observation of the decoupling caused by the superfluidity of ^{4}He films. As mentioned above, the torsional oscillator measurements revealed that ^{4}He films above n_{d4} show superfluidity at low temperatures [11]. At the corresponding coverages we observed a sharp bend of the temperature dependence in sound

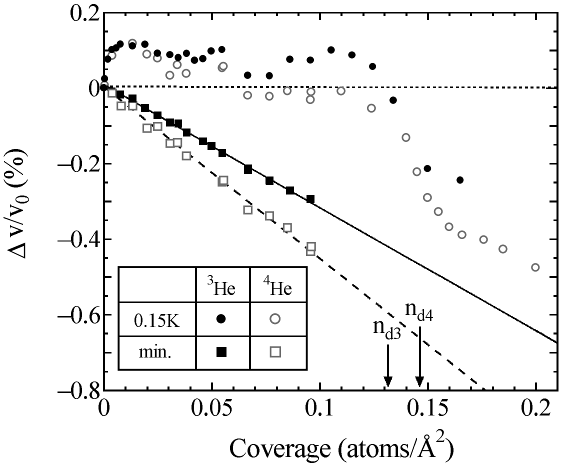

FIG. 1. Change in sound velocity from the pellet with no adsorbate as a function of coverage. Circles are the change in sound velocity at 150 mK. Squares represent the minimum which takes place at ~7–18 K. Open symbols correspond to ^{4}He, and filled symbols correspond to ^{3}He. The coverage dependence of the minimum is proportional to the mass of the adsorbate.

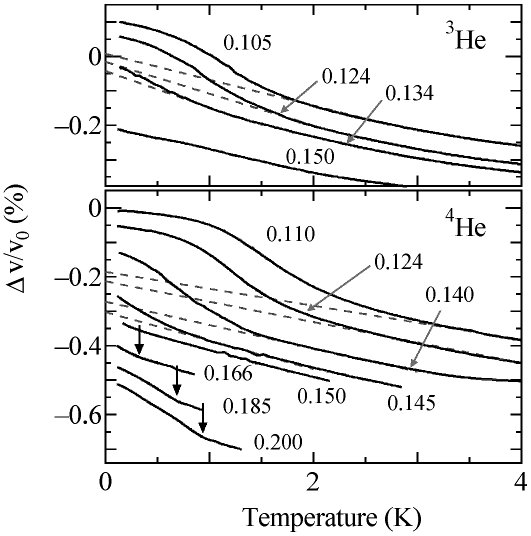

FIG. 2. Variation in the sound velocity of ^{3}He and ^{4}He bilayer films. Figures represent the areal density in the unit of atoms/Å^2. Arrows indicate the superfluid onset.

velocity, and the bend was recognized as the superfluid onset T_c.

The sound velocity deviation due to the superfluidity at 0.200 atoms/Å^2 is shown in the inset of Fig. 3. Here, we simply extrapolated the linear dependence above T_c and plotted the difference. The deviation at absolute zero corresponded to 0.08% of the sound velocity, and the decoupling rate of the superfluid fraction to the mass of the *fluid* state, $1 - \chi_s$, was evaluated to be 0.32. Although the value $1 - \chi_s$ is expected to be $2/3$ for a flat substrate, this depends on the morphology of substrate. The value of hectorite pellet should be compared with about 0.30 for Vycor glass, which can be read off the figure in Ref. [6]. We found that the decoupling rate of hectorite pellet is rather large, and it is interesting that this rate is about $1/3$ and close to that of Vycor glass.

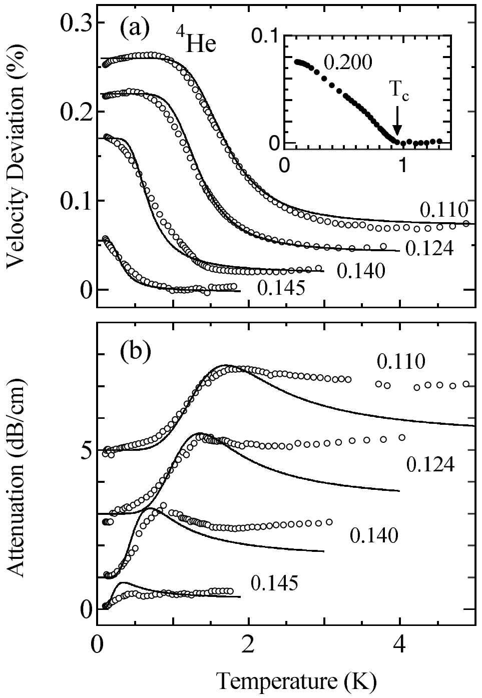

FIG. 3. (a) Sound velocity deviation at low temperatures and (b) the excess attenuation for nonsuperfluid ^{4}He films. Figures represent the areal density in the unit of atoms/Å^2. Solid curves are the fitted curves of Eqs. (2) and (3) with the "slip time" τ which obeys the Arrhenius relation. For clarity, the sets of data are shifted vertically. Inset: Sound velocity deviation due to the superfluidity.

In contrast to the decoupling of the superfluidity, it was found that the sound velocity of nonsuperfluid ^{4}He films *gradually* deviates upwards from the linear dependence extrapolated at high temperatures. Similar to ^{4}He, the sound velocity of ^{3}He increased gradually as the temperature was lowered. The deviation in sound velocity means that nonsuperfluid helium films slip from the substrate oscillation. It is interesting to compare the slipping rate of these films with the decoupling rate of the superfluid fraction. At 0.110 atoms/Å^2 for ^{4}He the sound velocity deviation at absolute zero was 0.19% of the sound velocity, and the slipping rate to the *total* mass, $1 - \chi$, corresponds to 0.35. This rate was much the same as the decoupling rate of the superfluid fraction.

As seen in the figure, the deviation at low temperatures appeared abruptly below 0.15 atoms/Å^2. This demonstrates clearly that a part of nonsuperfluid helium films changes its mechanical response at low temperatures from locking to slipping below this coverage.

Next, we consider the temperature dependence in the slippage of nonsuperfluid helium films. In comparison between the sound velocity and Eqs. (2) and (3), it was found that τ increases drastically as the temperature is lowered. Here, we assume that τ obeys the Arrhenius relation,

$$\tau = \tau_0 \exp(E/k_B T), \tag{4}$$

where E is the activation energy, and $1/\tau_0$ is the attempt frequency. The deviation of the sound velocity, which is the difference from the linear dependence at high temperatures, and the excess attenuation relative to the bare pellet are plotted in Fig. 3, together with the fitted curves. In the calculation, $\Delta\rho$ was also chosen as one of the fitting parameters, which corresponds to the effective density concerning the slippage. Although the observed attenuation is larger than the model fit at high temperatures, the temperature dependence of both the sound velocity and the attenuation was found to be rather well described as a whole. (We obtained $E \sim 4$ K for ^{3}He and $E \sim 5$ K for ^{4}He around n_{c3} and n_{c4}, and τ_0 was on the order of 10^{-9} s. The calculated activation energy E decreased with increasing coverage.) Thus, it is concluded that the thermally activated process plays an important role in the resistance to slippage of this system, and that the frictional force decreases rapidly as the temperature is lowered.

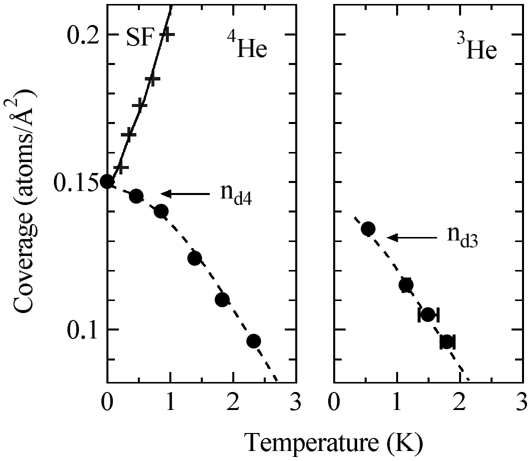

FIG. 4. Attenuation peak T^* and superfluid onset T_c as a function of coverage. The arrows indicate the coverages when the fluid state appears at absolute zero in the experiments of Wada *et al.* [11].

The decrease in the frictional force with decreasing temperature may contradict the naive idea of the pinning-depinning mechanism [1]: the adsorbate is pinned at low temperatures by the substrate potential, while the thermally activated motion occurs at high temperatures. However, this observation may reflect the known behavior that the frictional force of liquid films is larger than that of incommensurate-solid ones [2], i.e., the observed behavior may be related to the fact that helium films become fluid-like at high temperatures.

Figure 4 shows the attenuation peak T^* and the superfluid onset T_c as a function of coverage. The attenuation peak T^* decreased with increasing coverage, and tended towards absolute zero. It was found that T^* as a function of coverage agrees qualitatively between ^{3}He and ^{4}He. Here, we define the critical coverage n_{cr} as the coverage when the deviation in sound velocity and the attenuation peak vanish. It is worth noting that n_{cr} almost coincides with n_{d3} and n_{d4}. Furthermore, in the case of ^{4}He, we observed superfluidity just above n_{cr}.

Finally, we should comment on ultrasonic measurements of nonsuperfluid helium films on Vycor glass. Beamish and his colleagues observed a broad bump in the attenuation around 2 K [6]. This could not be attributed to a feature of the standard tunneling model and the origin was unknown. As compared with the present experiments, this attenuation is possibly associated with slippage of these films. We believe that slippage is observed for various substrates.

In summary, we measured the mechanical response of ^{3}He and ^{4}He films on the oscillating substrate, and found the conditions for slippage of nonsuperfluid helium films. Up to the coverage at which the fluid state appears at absolute zero, nonsuperfluid ^{3}He and ^{4}He films slipped from the oscillation at low temperatures, and this slippage was inhibited as the coverage increases further. In addition, it was found that from the temperature dependence of the slippage, a thermally activated process plays an important role in the frictional force of this system.

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[8] We observed the change in elastic stiffness below $\sim$0.01 atoms/Å^2. Up to this coverage, each adatom is localized at a registered site with a H^+ cation at low temperatures, which may cause the deformation of the substrate. In this paper, however, we confine ourselves to the films above 0.1 atoms/Å^2, or bilayer films. At these coverages, the binding energy is small, and shows no structure as a function of coverage. Thus, we can safely ignore the coverage dependence of elastic stiffness at bilayer films.

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