Temperature-Dependent Pinning or Depinning of a ^{3}He Overlayer in a ^{3}He-^{4}He Mixture Film

Takuya Oda and Mitsunori Hieda*

Department of Physics, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8602, Japan

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We report the results of a quartz crystal microbalance experiment at 100 MHz for a ^{3}He-^{4}He mixture film on a planar gold substrate. The results reveal temperature-dependent pinning or depinning of ^{3}He overlayers above a critical oscillation velocity and indicate that the appearance of a macroscopic condensed state in the underlying ^{4}He layer possibly affects the interfacial friction.

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To explore the nature of two-dimensional (2D) superfluidity arising from the Kosterlitz-Thouless (KT) transition [1], quantum fluid helium films have long been studied using shear oscillator techniques. Surprisingly, even in nonsuperfluid helium films, an unexpected anomalous frequency shift was also observed for various substrates in some oscillator experiments [2–7]. This anomalous behavior is frequently attributed to surface slippage of nonsuperfluid films that arise from insufficient nanoscale friction between the physisorbed films and the oscillating substrate [8–13]. According to this interpretation, the interfacial viscous friction $F = -(\sigma/\tau)V$ acts at the film-substrate boundary, and the slip state is determined by $\omega\tau$, where σ is the areal density, τ is the slip time, V is the sliding velocity, and ω is the angular frequency of measurement. τ is $1/e$ decay time for V. If $\omega\tau \ll 1$ (equilibrium condition), the physisorbed film does not slip. When $\omega\tau \gg 1$ (nonequilibrium condition), the film completely decouples from the oscillating substrate. The most striking example of decoupling is for a superfluid ^{4}He film with no viscosity. Neither of these extremes has any frictional energy dissipation. The film is slipping for all other values of $\omega\tau$, and the maximum dissipation arises at $\omega\tau \sim 1$.

To date, surface slippage of quantum fluids has been studied only for pure ^{4}He and ^{3}He, whereas films of a ^{3}He-^{4}He mixture remains to be studied. Through a quantum mechanism, the mixture film forms a unique layer structure. The different zero-point energies of ^{3}He and ^{4}He tend to separate the two isotopes in the van der Waals field perpendicular to the substrate, with ^{3}He residing on top of the ^{4}He film. At $T = 0$, third sound [14,15], nuclear magnetic resonance (NMR) [16,17], and torsional oscillator (TO) studies [18,19] suggest that the film consists of a simple isotopic layered structure such as a “superfluid sandwich” or of the trilayer structure of the ^{3}He, superfluid ^{4}He, and localized ^{4}He substrate, as shown in Fig. 1(a). The dissociation effect between ^{3}He and ^{4}He layers at finite temperature is still incompletely understood [20], but for $T \leq 0.5$ K, a third sound study [14] reported that nearly completely isotopic layering is retained. A quartz crystal microbalance (QCM) measurement reported unexpected decoupling of the ^{3}He overlayer at 0.25 K in mixture films on hydrogen, a weak binding substrate [21]. Thus, this system suggests that a previously unknown slippage occurs at the boundary between a ^{3}He overlayer and an underlying ^{4}He layer.

In this Letter, we report the results of 100 MHz QCM measurements of the ^{3}He overlayer in a ^{3}He-^{4}He mixture film on a planar gold substrate. The measurements were performed at various constant oscillating velocities v from 19 to 59 μm/s and reveal that slippage occurs between the ^{3}He overlayer and the underlying ^{4}He layer. For $v = 35$ μm/s, we observe a temperature-dependent pinning or depinning transition of ^{3}He on the underlying ^{4}He layer. The transition starts at temperatures slightly above the superfluid onset temperature T_o and follows a temperature hysteresis loop. The estimated critical force of the ^{3}He overlayer for depinning $F_c = 4.5 \times 10^{-4}$ N/m^2 is 10^3 times smaller than that of the classical physisorbed film, a 0.7 layer Kr film [11]. This result is possibly related to the

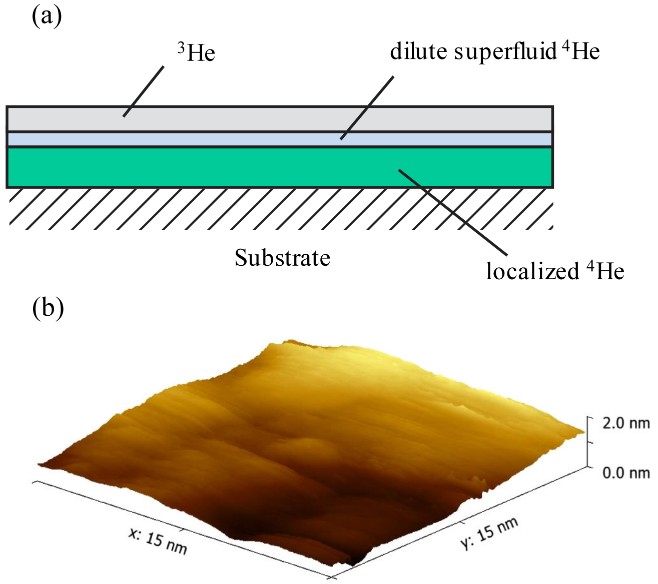

FIG. 1 (color online). (a) Possible isotopic layered structure of ^{3}He-^{4}He mixture films. In this study, the bulk-density-layer coverages of ^{3}He, superfluid ^{4}He, and localized ^{4}He are $n_3 = 0.75$, $n_{4,\mathrm{fluid}} = 0.11$, and $n_{4,\mathrm{local}} = 2.79$ layers, respectively. (b) STM image of gold substrate (15 nm × 15 nm × 2 nm).

appearance of a condensed state of underlying ^{4}He, which is analogous to the interplay between the superconductivity of the underlying substrate and the sliding friction [22–24].

For this low temperature study, we installed a commercial AT-cut quartz disc with a fundamental resonance at 20 MHz and gold electrodes in an experimental oxygen-free high-conductivity copper cell with a silver sinter of 0.4 m^2. The STM image of the gold substrate in Fig. 1(b), although relatively rather rough, was similar to a "smooth" rolling hill surface with a (111) texture, or a "textured" substrate, presented in the previous publications [9,13]. The surface area measured by nitrogen adsorption at 77 K, 0.10 ± 0.01 cm^2, was also closed to that of the geometrical flat plane 9.1×10^{-2} cm^2. Before cooling, the cell was evacuated at room temperature for one day using a turbo molecular pump through a 0.5 mm diameter CuNi capillary tube. The QCM experiment was performed at 100 MHz using the fifth harmonic mode (i.e., harmonic acoustic number $l = 5$) for a ^{3}He-^{4}He mixture film with a constant bulk-density-layer coverage of $n_3 = 0.75$ layers for the ^{3}He and n_4 $(= n_{4,\mathrm{fluid}} + n_{4,\mathrm{local}}) = 2.90$ layers for ^{4}He on the gold substrate. The bulk-density-layer coverages for the fluid and localized layers of ^{4}He were $n_{4,\mathrm{fluid}} = 0.11$ and $n_{4,\mathrm{local}} = 2.79$ layers, respectively. One bulk-density layer is defined as 12.9 and 10.6 μmol/m^2 for ^{4}He and ^{3}He, respectively [25]. The excitation voltage ranged from 0.2 to 0.7 mV. We found no heating problems in our previous study of 2D superfluidity in pure ^{4}He films at 60 MHz ($l = 3$) using the same QCM sample and experimental cell [26]. From the transmitted QCM voltage signal, the oscillation amplitude and velocity are estimated [27] to be 0.030–0.093 pm and 19–59 μm/s, respectively. The oscillation amplitude is much smaller than the atomic separation of both the adsorbed film and the substrate atoms. The frequency f and the change in the inverse Q factor ΔQ^{-1} are acquired through a heating or cooling temperature scan over 0.06–0.4 K at a constant oscillation velocity in the range of 19–59 μm/s. The measured Q factor at 100 MHz is 2×10^5 at 0.1 K. The resolutions for f and ΔQ^{-1} are 1 Hz and 1×10^{-8}, respectively, which is about 10 times worse than the best performance. One bulk-density layer of ^{3}He or ^{4}He causes a frequency shift of 28.7 or 46.6 Hz, respectively.

The oscillation velocity range used in this study falls within the velocity region of the linear superfluid response in the dynamic KT theory [28]. The used velocities are much lower than the ~1 mm/s critical velocity of the nonlinear superfluid response that was reported in a TO study on Mylar [29]. This is also supported by the fact found in our previous QCM study [26] that the temperature variation in the superfluid density ρ_s and ΔQ^{-1} showed no amplitude dependence. Thus, the result reported herein is not connected with the anomalous nonlinear superfluid response.

Figure 2(a) shows f and ΔQ^{-1} as a function of the temperature at 100 MHz for various oscillating velocities v from 19 to 59 μm/s. At 100 MHz, the frequency shift due to the superfluid KT transition is not clearly observed, and the dissipation ΔQ^{-1} peak, which is due to the dissociation of vortex pairs, is also hidden by the noise. To prove the existence of the superfluid transition at this coverage, we also show the 60 MHz data (third harmonic $l = 3$) for $v = 19$ μm/s under the *in situ* condition (no change in the coverage, cooling maintained below 0.5 K), which has better resolution. At 60 MHz, both the frequency shift (which is equivalent to the superfluid density) and the dissipation peak are clearly observed at the superfluid onset temperature $T_o = 0.155 \pm 0.005$ K and at the dissipation peak temperature $T_{\mathrm{pk},60} = 0.136 \pm 0.001$ K, respectively. According to the dynamic KT theory [28], the dissipation peak (pk) temperature T_{pk} depends on the measuring frequency $f = \omega/2\pi$ as $(T_{\mathrm{pk}} - T_{\mathrm{KT}})/T_{\mathrm{KT}} = (4\pi^2/b^2) \times \{0.5\ln[14D/(a_0^2\omega)]\}^{-2}$ [26,30], where D is the vortex diffusion constant, a_0 is the vortex core diameter, and b is a nonuniversal constant. Using the previously reported values, $D/a_0^2 = 2.8 \times 10^9$ s^{-1}($a_0 = 2.4$ nm) and $b = 7$ for the superfluid coverage of this study [26,31], we calculated $T_{\mathrm{KT}} = 0.118$ K and then the superfluid onset temperature at 100 MHz $T_{\mathrm{pk},100} = 0.141$ K. There is no significant difference between the transition temperatures (T_{pk} and T_o) at 60 and 100 MHz that affects the discussion below.

The frequency-dependent superfluid onset temperature T_o represents the maximum detectable temperature of superfluidity for the measured f. Within an oscillation period $1/f$, the observed superfluid density equals the stiffness for the size of the vortex diffusion length $r_D(\omega) = \sqrt{14D/\omega}$. In the low frequency limit (i.e., $r_D \rightarrow \infty$), the superfluid density exhibits the universal jump at T_{KT}. At a finite frequency, the superfluid onset temperature T_o is greater than $T_{\mathrm{KT}} = 0.118$ K, at which the 2D phase coherence length $\xi_+(T) \approx a_0 \exp[(2\pi/b)/\sqrt{(T - T_{\mathrm{KT}})/T_{\mathrm{KT}}}]$ becomes equal to $r_D(\omega)$. At 100 MHz, the coherence length ξ_+ at T_o is about 13 nm, as shown in Fig. 2(b).

Surprisingly, at $v = 35$ μm/s in Fig. 2(a), the situation changes dramatically: we find an extra large frequency shift and ΔQ^{-1} below 0.26 K with a hysteresis loop differentiating heating and cooling. Further increases in v cause the observed extra signals to become much larger. To more qualitatively understand the dependence on the oscillation velocity, in Fig. 2(c), we plot the frequency shift at absolute zero Δf ($T = 0$ K) as a function of the oscillation velocity. The frequency shift Δf ($T = 0$ K) is divided into two regions by the critical velocity $v_c \approx 30$ μm/s. Below v_c, the frequency shift (obtained using the superfluid fraction coupling to the substrate oscillation $\chi = 0.1$ reported in our previous study [26]) is equal to the expected superfluid decoupling of ^{4}He (4.6 Hz). Above v_c, the excess shift that generates the hysteresis loop begins, and it systematically increases with increasing v. These observations suggest that part of the nonsuperfluid component in the ^{3}He-^{4}He mixture film slips because of a

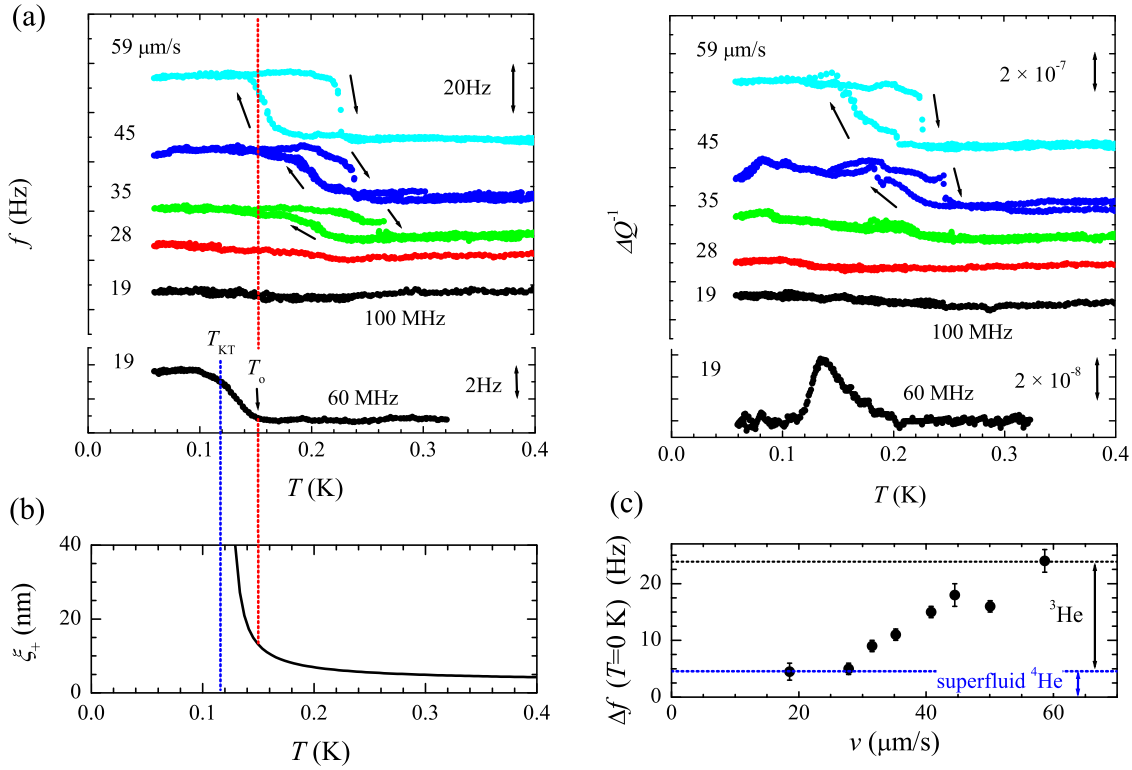

FIG. 2 (color online). (a) Frequency and change in inverse Q factor (energy dissipation) versus temperature at 100 MHz at various oscillating velocities from 19 to 59 μm/s. To clarify the superfluid onset temperature T_o, the higher resolution 60 MHz data at 19 μm/s are also shown. (b) Two-dimensional phase coherence length of superfluid ^{4}He versus temperature. The locations of T_{KT} and T_o are indicated by vertical dotted lines. (c) Frequency shift at 0 K versus substrate oscillation velocity. Dotted lines indicate the frequency shifts expected for superfluid ^{4}He and the decoupling of the ^{3}He overlayer.

temperature-dependent pinning or depinning mechanism. At the maximum velocity $v = 59$ μm/s, 10% of the entire mixture film decouples from the oscillation. Figure 3 shows the depinning (T_d) and pinning (T_p) temperatures, which were determined using the frequency data in Fig. 2(a). The temperatures T_d and T_p are always slightly greater than T_o in the v region that we measured. We currently have no clear explanation for the dependence of T_d and T_p on v.

The first question to address is which part of the mixture film in the origin of the slippage signal comes from. In our previous study [26] of pure ^{4}He films using the same experimental setup (quartz, experimental cell, 100 MHz frequency, and 0.6 mV driving voltage), we found no similar anomaly. In other words, neither the nonsuperfluid nor the localized layers of ^{4}He slipped. This indicates that the slippage must come from part of the ^{3}He overlayer on the underlying ^{4}He fluid layer. For perfect ^{3}He decoupling, the expected frequency shift is 19.3 Hz, with the natural assumption that for $\chi = 0.1$, 10% of the ^{3}He always couples to the substrate oscillation. At the maximum velocity $v = 59$ μm/s, all ^{3}He decouples from the oscillation, as shown in Fig. 2(c).

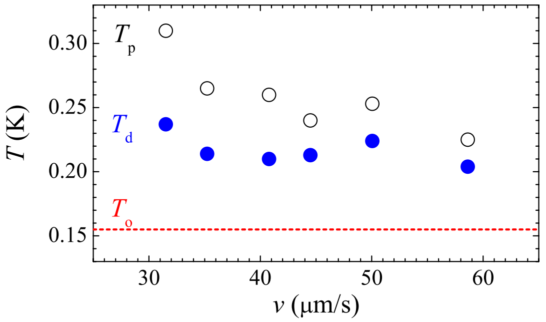

FIG. 3 (color online). Depinning (T_d) and pinning (T_p) temperatures versus oscillation velocity. Superfluid onset temperature T_o is also shown.

The depinning parameters, the critical amplitude A_c, velocity v_c, and force F_c, are summarized in Table I. The estimated F_c of the ^{3}He overlayer 4.5×10^{-4} N/m^2 is 10^3 times smaller than that of the classical physisorbed film, a 0.7 layer Kr film on gold [11]. This small F_c of ^{3}He is possibly related to the appearance of a condensed state of underlying ^{4}He, which is similar to the interplay between the superconductivity of the underlying substrate and the sliding friction [22–24].

The slip time is generally calculated [10] from the frequency shift δf and energy dissipation δQ^{-1}, which is due to the slippage of the nonsuperfluid with respect to the vacuum value, as $\tau = \delta Q^{-1}/(4\pi \delta f)$. In this study, to

TABLE I. Parameters for the depinning of ^{3}He in the ^{3}He-^{4}He mixture film and Kr on gold.

	Layer	f (MHz)	A_c (nm)	v_c (μm/s)	F_c (N/m^2)
^{3}He	0.75	100	4.8×10^{-5}	30	4.5×10^{-4}
Kr[a]	0.7[b]	6	0.45	1.7×10^4	0.41

[a] Parameters are estimated from data in Ref. [11].
[b] One monolayer of Kr is 11.0 μmol/m^2 (6.6 atoms/nm^2).

eliminate the superfluid signal, the data at 19 μm/s are treated as the background instead of the vacuum data and are subtracted. Figure 4 shows the slip time τ versus temperature at oscillation velocities of 28, 35, 45, and 59 μm/s. At 28 μm/s, which is below v_c, the slip time always satisfies $\omega\tau \ll 1$, which means that the mixture film is rigidly pinned to the gold substrate at all the temperatures measured. However, at 35, 45, and 59 μm/s, which are above v_c, the slip time exhibits a hysteresis loop between heating and cooling. When cooling and at 35 μm/s, τ abruptly increases at the depinning temperature T_d, which is slightly above the superfluid onset T_o, and is ~2 ns at 0.1 K. This sudden change in τ when passing through $\omega\tau = 1$ means that part of the ^{3}He overlayer decouples from the underlying ^{4}He layer. Conversely, when the temperature is increased above 0.06 K, τ increases to ~3 ns and then suddenly drops toward 0 ns at the pinning temperature T_p. At the higher velocities 45 and 59 μm/s, the hysteresis loop becomes larger. At the maximum, $\tau \approx 20$ ns, and thus the TO (~1 KHz) cannot access the equilibrium region, namely, $\omega\tau \gg 1$.

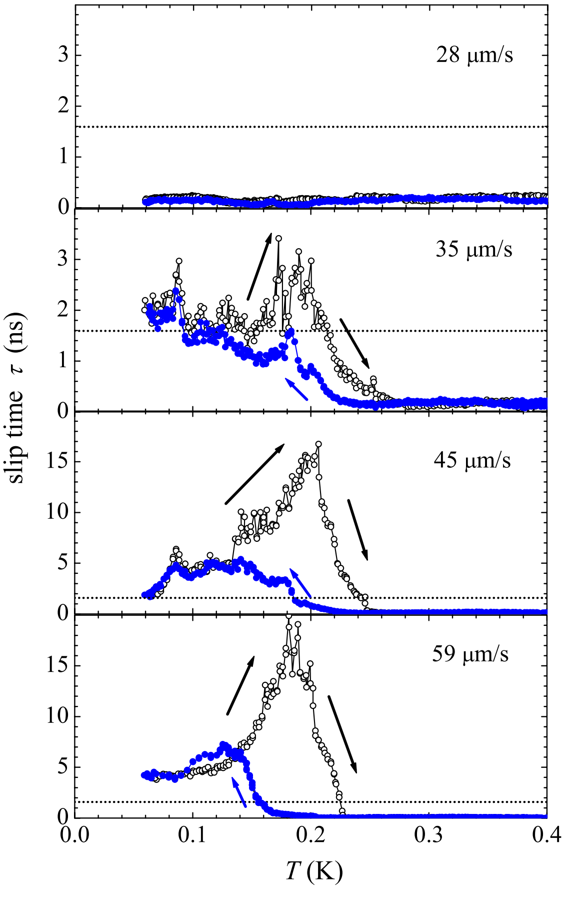

FIG. 4 (color online). Slip time τ versus temperature at oscillation velocities of 28, 35, 45, and 59 μm/s. The dotted lines show slip times corresponding to $\omega\tau = 1$.

Previous studies on classical physisorbed films (Kr and Ne) that involved amplitude scans at constant temperature suggest two factors for the origin of the pinning: substrate surface defects [11,13,32] and structural mismatch [12] between the adsorbate and the substrate surface. However, these effects do not explain our observation for two reasons: (1) substrate surface defects do not vary with temperature at this low temperature and (2) no sharp structural changes in the physisorbed films are observed on evaporated metal electrodes of commercially available QCM which are likely to have a polycrystalline (111) texture with a rough surface profile.

What mechanism causes this temperature-dependent pinning or depinning? A possible mechanism is the combined effect of (i) the bound state of ^{3}He in the mixture film and (ii) a precursor of the underlying superfluid ^{4}He above T_o. Finley *et al.* proposed effect (i) to explain their observation of ^{3}He decoupling in mixture films at $T < T_o$ [21]. ^{3}He has unique bound states in mixture films: the ground state occurs at the free surface of the underlying ^{4}He film and the excited states are inside the ^{4}He film. For films having thickness of several layers, these excited states have been predicted theoretically [33]. NMR studies of mixture films of comparable ^{3}He coverage on Nuclepore filters [17] show activation from the ground state to the first excited state with an energy gap of ~2 K. Thus, upon cooling, ^{3}He transitions from an excited state inside the ^{4}He film to the ground state at the free surface of superfluid ^{4}He, and ^{3}He floating on the superfluid ^{4}He must decouple from the oscillation. In practice, when the superfluid is very thin (i.e., submonolayer), momentum transfer probably occurs via pinning centers such as substrate defects or foreign molecules adsorbed on the substrate, and with low static friction, ^{3}He barely oscillates. Therefore, increasing the oscillation velocity v above the threshold v_c leads to the depinning transition.

To explain our observation that ^{3}He decouples slightly above T_o, effect (ii), the precursor of the superfluid ^{4}He film, must exist above T_o. Even above T_o, the phase coherence length $\xi_+(T)$ remains finite. In this study, ξ_+ at T_d is distributed over 5–8 nm, as shown in Fig. 2(b). Such nanoscale superfluidity is observed when the system size is reduced to the order of ξ_+, as demonstrated in porous media [34,35]. Therefore, we hypothesize that the ^{3}He overlayer forms many nanoscale patches of 5–8 nm in size and feels the nanoscale superfluidity of the underlying

^{4}He. The combination of effects (i) and (ii) may cause the depinning of ^{3}He above T_o.

In conclusion, through 100 MHz QCM measurements, we detected the slippage of a nonsuperfluid ^{3}He-^{4}He mixture film on a planar gold substrate at constant oscillating velocities $v = 19$ to 59 μm/s. Above the critical velocity $v_c \approx 30$ μm/s, the temperature-dependent pinning or depinning transition of the ^{3}He overlayer on the underlying ^{4}He occurs slightly above T_o. This study indicates that the appearance of a macroscopic condensed state in the underlying ^{4}He layer possibly affects the interfacial friction.

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*hieda@cc.nagoya-u.ac.jp

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