Slippage of Nonsuperfluid $^3$He-$^4$He Mixture Film on Gold

M. Hieda, T. Oda, T. Matsushita, and N. Wada
Department of Physics, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8602, Japan
E-mail: hieda@cc.nagoya-u.ac.jp

Abstract. We report the result of a QCM experiment at 100 MHz for a coverage of $^3$He-$^4$He mixture film ($n_3 = 0.75$ and $n_4 = 2.90$ bulk-density layers) on a planar gold substrate. This study is done by taking the temperature scan of both warming and cooling at various constant oscillation velocities $v$ from 19 to 59 $\mu$m/s. At minimum $v = 19$ $\mu$m/s, a frequency shift of about 5 Hz due to the superfluid transition is observed below the onset temperature $T_o = 0.16$ K. Surprisingly, at $v = 35$ $\mu$m/s, the observation is dramatically changed and an extra large frequency shift with a hysteresis loop between warming and cooling is found. Upon increasing $v$ further, the extra shift evolves into much larger shifts. These observations suggest that a part of $^3$He on underlying $^4$He layer slips by a pinning-depinning mechanism above $v = 35$ $\mu$m/s. At maximum $v = 59$ $\mu$m/s, the decoupling ratio of slippage reaches 90% of the whole $^3$He population.

1. Introduction
In oscillator studies for atomically thin nonsuperfluid helium films, anomalous frequency shifts are observed in some experiments on graphite [1, 2], hectorite [3], and porous gold [4]. This anomalous behavior is often interpreted by a slippage of the nonsuperfluid films from oscillating substrate relating to a nanoscale friction of physisorbed films. In this interpretation, the interfacial viscous friction $F = -\left(\sigma/\tau\right)V$, where $\sigma$ is the areal density, $\tau$ the slip time, and $V$ the sliding velocity, acts at the film-substrate boundary, and the slip state is determined by $\omega\tau$, where $\omega$ is angular frequency of measurement. The slip time $\tau$ represents the time required for the sliding velocity to decay $1/e$ of its initial value. When $\omega\tau \ll 1$, a physisorbed film shows no slippage. When $\omega\tau$ increases and approaches to 1, the film starts to slip. When $\omega\tau \gg 1$, the film completely decouples from the oscillating substrate. The slippage behaviors have been reported for pure $^4$He or $^3$He films. We report here a result of our quartz crystal microbalance (QCM) experiment at 100 MHz for a coverage of $^3$He-$^4$He mixture film on a flat gold substrate. It could be interesting as $^3$He-$^4$He mixture films form a unique structure for adsorption.

$^3$He-$^4$He mixture films have been studied for a long time to explore $^3$He effects on the nature of 2D superfluidity [5]. However, the microscopic configuration of $^3$He-$^4$He mixture films is incompletely understood. 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. So far, at $T = 0$, a possible structure of the film is proposed to be a simple layer model, $^3$He/superfluid $^4$He/solid-like $^4$He/substrate, by torsional oscillator (TO) studies on Mylar [6] and porous gold [7]. In this study, the coverages of $^3$He, superfluid $^4$He, and solid-like $^4$He are $n_3 = 0.75$, $n_{4,\text{fluid}} = 0.15$, and $n_{4,\text{solid}} = 2.75$ bulk-density layers respectively. At finite temperatures, it must be complicated...
by the possibility of mixing of $^3$He and $^4$He. At the coverage of this study, there is no signature of the mixing in the previous TO study [7] and our QCM study [8] as the overall $T$ dependence of superfluid density $\rho_s$ of the $^3$He-$^4$He mixture film shows no difference from pure $^4$He.

2. Experimental
The QCM is a mass sensor using the piezoelectric property of quartz. Applying an AC voltage across two electrodes of a quartz disc, the disc oscillates with a thickness shear mode and detects nanogram order of mass on the electrodes. Therefore the QCM has been not only used for a conventional thickness monitor in a vacuum chamber of an evaporator but also for fundamental researches, adsorption, superfluid film, nanofriction, wetting and others.

In this low temperature study, a commercial AT-cut quartz disc with a fundamental resonance at 20 MHz and with gold electrodes is installed in an OFHC experimental cell with silver sinter of surface area 0.4 m$^2$. Before cooling down, the cell is evacuated at room temperature for one day by a turbo molecular pump through a CuNi capillary 0.5 mm in diameter. The QCM experiment is done at 100 MHz using the 5th harmonic mode (the harmonic acoustic number $l = 5$) and at the constant coverage of $^3$He-$^4$He mixture film ($n_3 = 0.75$ and $n_4 = n_{4,\text{fluid}} + n_{4,\text{solid}} = 2.90$ bulk-density layers) on flat gold substrate. One bulk-density layer (one bulk liquid density at zero bar) is defined as 12.9 $\mu$mol/m$^2$ and 10.6 $\mu$mol/m$^2$ for $^4$He and $^3$He, respectively. The excitation voltage is used in the region of 0.2 - 0.7 mV. No heating problem is confirmed in the previous study of 2D superfluidity for pure $^4$He films at 60 MHz ($l = 3$) using the same QCM sample and experimental cell [9]. From the transmitted voltage signal of the QCM, the oscillating mechanical amplitude and velocity are estimated [10] to be 0.030 - 0.093 pm and 19 - 59 $\mu$m/s respectively. The data is acquired by taking the temperature scan of both warming and cooling in the range of 0.06 - 0.4 K at the constant oscillating velocity of 19 - 59 $\mu$m/s. The $Q$ factor at 100 MHz is measured to be $2 \times 10^4$ at 0.1 K. The resolution of frequency and the inverse $Q$ factor change $\Delta Q^{-1}$ are 1 Hz and $1 \times 10^{-8}$ respectively, which is about 10 times worse than the best performance for some reason. One bulk-density layer of $^3$He and $^4$He causes the frequency shifts of 28.7 and 46.6 Hz respectively.

We note that this study is performed within the velocity region of the linear superfluid response. The velocities of our measurement are much lower than the critical velocity, 1 mm/s, for the nonlinear response of a superfluid $^4$He film observed in the TO study on Mylar [11]. This is also verified in our previous QCM study [9] by the fact that the temperature dependences of $\rho_s$ and $\Delta Q^{-1}$ show no amplitude dependence. Our result in this paper is not influenced by any anomalous behavior of the nonlinear superfluid response.

3. Results and Discussion
Figure 1 shows the frequency and $\Delta Q^{-1}$ as a function of temperature at 100 MHz for various oscillating velocities. At the minimum $v = 19$ $\mu$m/s, the frequency shift about 5 Hz due to the superfluid KT transition is observed below the onset temperature $T_o \sim 0.16$ K. The associated dissipation peak of $\Delta Q^{-1}$ is hidden by the noise. For clarity, the 60 MHz data with the better resolution at the same coverage and excitation voltage is also shown, and $T_o = 0.155 \pm 0.005$ K. Surprisingly, at $v = 35$ $\mu$m/s, the observation is dramatically changed and an extra large frequency shift and $\Delta Q^{-1}$ with a hysteresis loop between warming and cooling is found. On increasing $v$ further, the extra shift evolves into a much larger shift than the expected value for the superfluid transition. These observations suggest that a part of the nonsuperfluid component in the $^3$He-$^4$He mixture film slips by a pinning-depinning mechanism above $v = 35$ $\mu$m/s. At the maximum $v = 59$ $\mu$m/s, 10% of the whole mixture film decouples from the oscillation.

To better understand this temperature-induced pinning-depinning transition, we estimate the slip time $\tau$. The slip time is calculated from the frequency $\delta f$ and dissipation $\delta Q^{-1}$ shifts due to the slippage of the nonsuperfluid with respect to the vacuum value as $\tau = \delta Q^{-1}/(4\pi \delta f)$ [12].
Figure 1. Evolution of frequency and $\Delta Q^{-1}$ versus temperature at 100 MHz with increasing oscillating velocity of QCM. For clarity, the 60 MHz data with the better resolution at the same coverage and excitation voltage is also shown.

Figure 2 shows the slip time $\tau$ versus temperature at $v = 59 \, \mu m/s$. We assume that $\tau \sim 0$ at the higher temperature side of the anomalous hysteresis since the mixture film rigidly locks to the gold substrate. The calculated $\tau$ shows a hysteresis loop. Decreasing temperature, $\tau$ abruptly jumps up at 0.16 K and is 20 ns at 0.1 K. Increasing temperature, $\tau$ slowly rises to 30 ns and then suddenly drop down to 0 ns at 0.20 K. The pinning-depinning occurs during the sudden change of $\tau$ passing through $\omega \tau = 1$.

The first question is which part of the film the slippage signal comes from. In our previous study [9] for pure $^4$He films using the same experimental situation (quartz, experimental cell, frequency (100 MHz), and driving voltage (0.6 mv)), no observation of the same anomaly is found. In other words, both the fluid and solid-like layers of $^4$He show no slippage. This indicates that the slippage signals mainly come from a part of $^3$He on $^4$He fluid layer. At the maximum $v = 59 \, \mu m/s$, $\delta f = 19 \, Hz$, which corresponds to 90 % of the whole $^3$He component.

The second question is what mechanism the temperature-induced pinning-depinning transition occurs by. In case of the classical physisorbed films (Kr and Ne), by keeping a constant temperature and varying the coverage and the amplitude, two factors are suggested as the origin of the pinning; defects of the substrate and a structural mismatch between surfaces of adsorbate and substrate [13, 14]. However these are not the intrinsic reasons for our observation of the temperature-induced pinning-depinning transition with the sudden change at $\sim 0.2 \, K$ because there is generally no sharp structural change for the physisorbed films on a substrate with poor quality like commercially available QCMs. Therefore we need to speculate further about the other mechanism of the pinning-depinning transition.

To suggest a possible scenario, we note two things observed in this study; i) a part of $^3$He on underlying $^4$He slips and ii) the pinning-depinning transition temperature is close to the superfluid onset $T_\alpha$. At higher temperatures, the momentum transfer to $^3$He from the oscillator occurs at the interface of $^3$He and nonsuperfluid $^4$He. At lower temperatures, no superfluid $^4$He is believed to transfer the momentum to $^3$He. Thus, when thickness of the superfluid is sufficiently thick, $^3$He floating on superfluid $^4$He must decouple from the oscillation. On the other hand, when the thickness of the superfluid is very thin, like a submonolayer, the momentum transfer probably occurs by some pinning centers, such as defects of the substrate, and $^3$He barely oscillates with a low static friction. Therefore, by increasing the amplitude, the pinning-depinning transition is observed beyond a threshold.
In general, reproducibility of the experimental studies on the slippage of physisorbed films is a frequent problem. This is probably because the slippage is highly sensitive to surface roughness and contamination of the substrate [13, 14]. Therefore it is natural to think that the results of this study also depends on the experimental condition and preparation. In particular, further studies of 2D superfluidity in $^3$He-$^4$He mixture films should pay attention to the possibility for the decoupling of $^3$He on the underlying $^4$He and use low amplitude of oscillators.

4. Conclusion

We report the slippage of the nonsuperfluid $^3$He-$^4$He mixture film on the flat gold by the QCM measurement at 100 MHz for various constant oscillating velocities $v$ from 19 to 59 $\mu$m/s. Above $v = 35$ $\mu$m/s, the temperature-induced pinning-depinning transition of $^3$He on underlying $^4$He is observed accompanied by a hysteresis loop. To describe the mechanism, the possible scenario related to the superfluid transition of $^4$He fluid layer is suggested.

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