QCM Study on 2D Vortex in Superfluid $^4$He and $^3$He-$^4$He Mixture Films

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Abstract. Two-dimensional (2D) $^4$He fluid films show the Kosterlitz-Thouless (KT) transition where pairing and unpairing of the thermally excited 2D vortices play an important role. However, in the superfluid submonolayer, the vortex parameters (the diffusion constant $D$, the core diameter $a_0$) have been incompletely estimated for various conditions. Here, we undertake QCM measurements for pure $^4$He films (on gold and H$_2$ substrates) and $^3$He-$^4$He mixture films (on gold substrate), and accurately determine the parameter $D/a_0^2$ by the high frequency dependence of the superfluid onset from 20 to 180 MHz. By the comparison of the results of pure $^4$He film on gold and H$_2$ substrates, the vortex diffusion in our study has the largest value $D \sim \hbar/m$ in the quantum limit. The core diameter $a_0$ is estimated to be the same magnitude as the de Broglie wavelength at $T_{KT}$ between 0.1 and 0.9 K. In terms of $^3$He-$^4$He mixture films, we observe no effect of $^3$He on the vortex parameters up to the $^3$He coverage of 15.1 $\mu$mol/m$^2$.

1. Introduction

Two-dimensional (2D) $^4$He fluid films on various substrates show the Kosterlitz-Thouless (KT) superfluid transition [1] where pairing and unpairing of the thermally excited 2D vortices play a major role. Important vortex parameters (the diffusion constant $D$, the core diameter $a_0$) have been extensively studied on flat and porous substrates by various techniques [2, 3, 4]. Most of the researches have been done for the thicker films above the coverage with $T_{KT} = 1$ K. On the other hand, in the thinner coverage region, there is only a few systematic studies [5, 6, 7] on the vortex properties, and the important vortex parameters $D, a_0$, and even the combination of the two parameters $D/a_0^2$ are not well determined. In most of the experiments $D/a_0^2$ is estimated, since $D$ and $a_0$ are the difficult quantities to be estimated independently. In this paper, we report the accurate determination of the parameter $D/a_0^2$ in the superfluid submonolayer region by the frequency dependence of the superfluid onset from 20 to 180 MHz by a quartz crystal microbalance (QCM). To study on the 2D vortex under various conditions, the QCM measurements have been done for pure $^4$He films on H$_2$ substrate and $^3$He-$^4$He mixture films on gold substrate, and then both the experiments are compared to the pure $^4$He study on gold. H$_2$ substrate is a weak binding substrate which is prepared by preplating on gold. We can examine substrate dependences, the effects of varying the $^4$He-substrate potential strength and smoothing out the surface roughness. In terms of $^3$He-$^4$He mixture films, it is reported that the vortex core size increases nearly linearly with added $^3$He to a pure $^4$He film in the previous torsional oscillator (TO) study on porous alumina of 50 nm powder [8]. It is interesting of a question as to whether the $^3$He effect is reproduced on planar substrate.
2. Experimental
The QCM is a thin AT-cut quartz disc that oscillates in the shear mode when a voltage is applied across the electrodes. In this 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 0.4 m$^2$, and mounted on a mixing chamber of a dilution refrigerator. We used a 1.25 cm diameter commercial AT-cut quartz disk of which a 0.34 cm diameter gold electrode is deposited on each side. Using the overtone harmonic modes of the QCM with a fundamental frequency of 20 MHz, we measure the superfluid at the different five frequencies, 20, 60, 100, 140, and 180 MHz. The typical quality factors $Q$ on gold substrate are $5 \times 10^4$ (20 MHz), $3 \times 10^5$ (60 MHz), $3 \times 10^5$ (100 MHz), $1 \times 10^5$ (140 MHz), and $9 \times 10^4$ (180 MHz) at 1 K, respectively. We observe no heating problem in the present QCM measurement down to 70 mK. The coverage is determined from the frequency shift due to the adsorption by the conventional microbalance method for QCM. The data is acquired by the temperature scan of both warming and cooling at a constant excitation voltage $V_{ex}$. For the background correction, the temperature dependence of the empty data is subtracted from the obtained data. The typical $V_{ex}$ is 0.6 mV, of which the mechanical amplitude and the oscillation velocity $v$ at 60 MHz are estimated to be 0.51 pm and 31 $\mu$m/s respectively from the QCM output voltage [9]. In the present experiments, we observe no nonlinear dependence on $V_{ex}$ between 0.2 and 0.6 mV ($v = 15 \sim 55 \mu$m/s). Using the QCM, we can obtain information of the superfluid density and the dissipation from the frequency shift and the output voltage, respectively. From the frequency shift $\Delta(\omega/2\pi)$ due to the superfluid, the superfluid areal density $\sigma_s$ is given by

$$\sigma_s = -\frac{\ell \pi^2 Z_q \Delta(\omega/2\pi)}{\omega_0^2 (1 - \chi)}, \quad (1)$$

where $\omega_0$ is the initial angular frequency, $Z_q = 8.862 \times 10^6$ kg/m$^2$s the transverse acoustic impedance of the quartz, and $\ell = (1, 3, 5, 7, \text{or} 9)$ the harmonic acoustic mode number. $\chi$ is the fraction of the superfluid that couples to the substrate. We put $\chi = 0.1$, almost the same as that in the TO experiment on Mylar sheets [10]. The dissipation associated with the dynamic superfluid transition is measured from the change in the inverse $Q$ factor.

3. Analysis: Frequency Dependence of Dynamic KT Superfluid Transition
According to the dynamic KT theory [11], the superfluid onset depends on the measuring frequency $\omega/2\pi$. In oscillator measurements at the frequency $\omega/2\pi$, the superfluid density is observed when the vortex diffusion length $r_D = \sqrt{4D/\omega}$ in one period of oscillation is roughly equal to or less than the mean distance between free vortices, or the 2D phase coherence length $\xi_+ \approx a_0 \exp[(2\pi/b)/\sqrt{(T - T_{KT})/T_{KT}}]$ from the Kramers-Kronig relations in the dynamic KT theory, the dissipation peak is observed near the superfluid onset temperature. Thus, the dissipation peak temperature $T_p$ is calculated [5] as

$$\frac{T_p - T_{KT}}{T_{KT}} = \frac{4\pi^2}{b^2} \left( 1 + \ln \frac{14D}{a_0^2 \omega} \right)^{-2} \quad (2)$$

Figure 1 shows the frequency dependence of the superfluid dissipation peak temperature $T_p$ with the parameters, $T_{KT} = 0.561$ K, $D/a_0^2 = 2 \times 10^{10}$ s$^{-1}$, and $b = 7$. The typical frequency regions of TO ($0.1 \sim 3$ kHz) and QCM ($2 \sim 200$ MHz) are also indicated. $T_p$ in the TO region shows a little dependence on the frequency. The QCM is a much more sensitive device to the frequency dependence of the KT transition. In our study, the experimentally obtained frequency dependences of $T_p$ are fitted by eq. (2). Here, $D/a_0^2$ and $T_{KT}$ are the free fitting parameters and $b$ is the fixed parameter of 7$\pm$1 experimentally estimated from the coverage above $T_{KT} = 1$ K [5]. The experimental data of $T_p$ for pure $^4$He on gold substrate at $n_4 = 40.0$ $\mu$mol/m$^2$, $a_0 = 3.6\times 10^{-10}$ m, and $b_0 = 0.6$ nm, are fitted by eq. (2).
which is described in the section 4.1, is good agreement with eq. (2). This analysis has an advantage against analysis of the temperature dependence. The dissipation peak temperature $T_p$ is insensitive to broadening of the temperature dependence at around the superfluid transition due to inhomogeneity of the film density.

When $r_D \approx a_0$, eq. (2) diverges at the critical frequency $(14/2\pi)D/a_0^2$, and thus this equation is not valid near and above the critical frequency. At extremely high frequencies, the superfluid density will be observed far above $T_{KT}$ up to $T_{GL}$ in terms of the GL theory. In our previous paper [6] on gold substrate, we report that eq. (2) explains the superfluid transition at extremely high frequencies where the diffusion length $r_D$ is about 25% higher than the static KT transition temperature $T_{KT}$.

4. Results and Discussion

4.1. Pure $^4$He Film on Au and H$_2$

In this section, we introduce the results of pure $^4$He on Au and H$_2$ substrates. For preparation of H$_2$ substrate, it is preplated on gold substrate by admitting at $\sim 20$ K through an ortho-para converter and then cooling down. The thickness of H$_2$ is 45 $\mu$m/m$^2$, which is equivalent to 3.3 monolayers using the adsorption area of one H$_2$ atom, 0.121 nm$^2$, estimated by the frequency shift due to the adsorption data at 4 K. The potential strength between one $^4$He atom and substrate in the most common form is written by $V(z) = \frac{4C^2}{2D^2} \frac{1}{z^2} - \frac{C^2}{z^2}$, where $C$ and $D$ are the Lennard-Jones parameters. The well depth $D$ of gold and H$_2$ substrates is 93 and 28 K, respectively [12]. H$_2$ substrate is the weaker binding substrate. In addition, by preplating H$_2$, the surface roughness is expected to be smoothed out. By the comparison of bare gold and preplated H$_2$ substrates, we can investigate these effects on the vortex parameters.

Figure 2 shows the superfluid density $\sigma_s$ and the dissipation $\Delta Q^{-1}$ versus temperature at 20, 60, and 140 MHz on gold and H$_2$ substrates. For comparison, the $^4$He coverages $n_4$ with almost same $T_{KT} \sim 0.57$ K are 40.0 and 14.7 $\mu$m/m$^2$ on gold and H$_2$ substrates, respectively. On both the substrates, the superfluid onset is observed at remarkably higher temperature than the static KT transition temperature $T_{KT}$. The onset and dissipation peak temperatures increase with increasing frequency. At 140 MHz on gold substrate, the onset temperature at 0.7 K is about 25% higher than $T_{KT} = 0.561$ K. As reported in our previous papers [6, 13], the coverage dependence on both gold and H$_2$ substrates is explained by the KT universal line expected from $T_{KT} = \frac{h^2}{2mk_B} \sigma_s$, where $m$ is atomic mass of $^4$He, $h$ the Planck constant, $k_B$ the Boltzmann constant. In terms of the onset coverage, we observe a substrate dependence. When H$_2$ is preplated on gold substrate, the superfluid onset coverage reduces from $\sim 32$ $\mu$m/m$^2$ to $\sim 7$ $\mu$m/m$^2$. This corresponds to the reduction of the potential strength between one $^4$He atom and substrate. The previous study on porous gold by TO [14] reports that the onset
Figure 2. Superfluid density $\sigma_s$ and dissipation $\Delta Q^{-1}$ versus temperature at 20, 60, and 140 MHz for (a) pure $^4$He on Au, (b) pure $^4$He film on H$_2$, and (c) $^3$He-$^4$He mixture film on Au. Solid curves are fittings by eq. (2) with the free parameters $D/a_0^2$ and $T_{KT}$, and the fixed parameter $b = 7 \pm 1$. $T_{KT}$ is shown in the figures.

coverage increases monotonically with increasing the well depth $D$.

The observed frequency dependences on both gold and H$_2$ substrates are well-fitted by eq. (2), as shown by the solid curves in Fig. 2 (a) and (b). The parameters on gold substrate are plotted by solid circles in Fig. 3 (a). The parameter $D/a_0^2$ on the gold substrate slowly decreases from $3 \times 10^{10}$ to $10^{9}$ s$^{-1}$ with decreasing $T_{KT}$. $D/a_0^2$ on the weaker binding substrate, H$_2$ substrate, is also shown in Fig. 3 (a), and this value on H$_2$ substrate shows exactly the same value $10^9 \sim 10^{10}$ s$^{-1}$ as that on gold substrate. We find no substrate dependence of $D/a_0^2$ between the two substrates with the different potential strength and degree of the surface roughness. This suggests that the vortex diffuses on both substrates with a possible largest value of the diffusion constant $D$ at the quantum diffusion limit. This largest value of $D$ is estimated to be $\sim \hbar/m$ by a dimensional analysis [11]. In a previous rotational TO experiment on Mylar [2], the values $D$ between 0.5
possible structure of the mixture film is proposed to be a simple layer model, 3.3

4.2. 4He-3He Mixture Film on Au

4He-3He mixture films have been studied for a long time to explore 3He effects on the nature
of 2D superfluidity [18]. The different zero point energies of 3He and 4He 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 mixture film is proposed to be a simple layer model, 3He/superfluid
4He/solid-like 4He/substrate, even for the superfluid submonolayer 3He by the TO studies on
porous gold [14] and Mylar [19].

In the previous TO study of 3He-4He mixture films on porous aluminia of 50 nm powder [8],
a strong broadening of the temperature dependence of \( \sigma_s \) is observed as 3He coverage is increased.
Analyzing of the broadening by a modified KT theory for the finite sizes yields the vortex core
size which increases nearly linearly with added 3He. When 3He of \( \sim 5 \mu\text{mol/m}^2 \) (\( \sim 0.5 \) bulk-
density layers) is added to pure 4He of the coverage with \( T_{KT} \sim 0.45 \) K, the vortex core size
grows up to \( \sim 5 \) nm from \( \sim 0.8 \) nm. One bulk-density layer (one bulk liquid density at zero bar)
is defined as 12.9 \( \mu\text{mol/m}^2 \) and 10.6 \( \mu\text{mol/m}^2 \) for 4He and 3He, respectively. The coverage of
the mixture film is frequently represented using the bulk-density layer.

To examine the 3He effect on the 2D vortex in the superfluidity on planar gold, we study
the mixture films at the coverages similar to the porous aluminia study [8], with keeping at the
constant 4He coverage \( n_4 = 42.3 \mu\text{mol/m}^2 \) with \( T_{KT} = 0.695 \) K and then adding 3He, \( n_3 = 0.7, 4.0, 9.8 \) and 15.1\( \mu\text{mol/m}^2 \) which corresponds to 0.07, 0.38, 0.92, and 1.42 bulk-density layers
respectively. A rapid reduction of the superfluid onset is observed with adding 3He, which is
agreement with the TO study on porous gold [14].
Figure 2(c) shows the superfluid density $\sigma_s$ and the dissipation $\Delta Q^{-1}$ versus temperature at 20, 60, and 140 MHz for the $^3$He-$^4$He mixture film at $n_3 = 0.7 \mu$mol/m$^2$ on gold substrate. The temperature dependences of $\sigma_s$ and $\Delta Q^{-1}$ are the same dependences as pure $^4$He and thus represent no extra broadening by $^3$He. The observed frequency dependence is well-fitted by eq. (2), as shown by the solid curve in Fig. 2(c). The parameters are plotted by solid squares in Fig. 3(a). $D/a_0^2$ of the mixture on the gold substrate slowly decreases from $3 \times 10^{10}$ to $10^9$ s$^{-1}$ with decreasing $T_{KT}$. This observation is equivalent to the results of pure $^4$He film. We find no observation of $^3$He effects on the parameter $D/a_0^2$ within the experimental errors. Our result at present on the planar substrate is contradictory to the previous reports on porous alumina [8]. This contradiction may suggest that the reported $^3$He effect on the vortex core size is an intrinsic behavior only in porous materials. It should be noted that, in our study, at very thick $^3$He coverages there are still possibility of the $^3$He effect, since the clear broadening of the temperature dependence of $\sigma_s$ is observed on Mylar at $n_3 = 136 \mu$mol/m$^2$ ($\sim$ 13 bulk-density layers) [19]. We are willing to extend our measurements to the thicker $^3$He region.

5. Conclusion

We accurately determine the parameter $D/a_0^2$ by the high frequency dependence of the superfluid onset from 20 to 180 MHz for pure $^4$He films (on gold and H$_2$ substrates) and $^3$He-$^4$He mixture films (on gold substrate) in the coverage of the superfluid submonolayer. By the comparison of the results of pure $^4$He film on gold and H$_2$ substrates, the vortex diffusion in our study has the largest value $D \sim h/m$ in the quantum limit. The core diameter $a_0$ is estimated to be the same magnitude as the de Broglie wavelength at $T_{KT}$ between 0.1 and 0.9 K. In terms of $^3$He-$^4$He mixture films, we find no effect of $^3$He on the vortex parameters up to the $^3$He coverage of 15.1 $\mu$mol/m$^2$.

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