Novel Behavior of ³He-⁴He Mixture Films on a Cesium Coated Substrate at Low Temperatures

K.S. Ketola* , T.A. Moreau, and R.B. Hallock

Laboratory for Low Temperature Physics, Department of Physics and Astronomy, University of Massachusetts, Amherst, MA 01003

We report quartz crystal measurements of the adsorption of ³He - ⁴He mixture films to gold and cesium substrates as a function of temperature and concentration. Measurements for higher concentrations than reported earlier show steps in the adsorption at low temperatures. Speculations as to the origin of these steps are presented.

1. INTRODUCTION

Cheng et al.[1] predicted and it was subsequently confirmed[2,3] that ⁴He does not wet cesium at low temperatures. In general, wetting occurs when $\sigma_{sl} + \sigma_{lv} \leq -\rho_0 \int V(z)dz$, where σ_{sl} and σ_{lv} are the solid-liquid and liquid-vapor surface tensions, respectively, z is the coordinate normal to the substrate surface, and ρ_0 is the bulk ⁴He density. Thus, in terms of the model potential proposed by Cheng et al.,[4] for $\sigma_{sl} \approx \sigma_{lv}$, wetting occurs whenever $(C_3D^2)^{1/3} \geq 3.3 \ \sigma_{lv}/\rho_0$, where D is the depth of the pair potential and C_3 is the coefficient of the van der Waals term. For ⁴He-Cs, C_3D^2 is quite small and the wetting inequality is not satisfied at low temperatures.

The addition of ³He to a ⁴He film results in a lowering of the surface tension at low temperatures since the ³He occupies a surface state on the ⁴He film. It was predicted by Pettersen and Saam[5] that this reduction in the surface tension would be adequate to satisfy the inequality and that the presence of the ³He would make the wetting of helium to cesium re-entrant: wetting would occur at low and high temperatures but would not occur at temperatures in between. Ketola and Hallock[6] confirmed this prediction.

*Current Address: Physics Department, Brown University, Providence, RI.

Here we extend their work to higher concentrations and report the observation of a new feature in the data.

2. APPARATUS

Two quartz crystal resonators with Au electrodes are mounted in an experimental chamber and one is coated *in situ* with Cs. We obtain the resonance frequency and a measure of changes in the total Q of the system by using a feedback technique similar to that developed by Lea et al.[7] A signal generator creates a high frequency carrier signal, f_0, which is frequency modulated and fed to one of the electrodes of a resonator. The AC signal at the other electrode of the resonator is amplified and converted to a DC signal proportional to the power of the AC signal by a diode detector. This low frequency signal from the diode detector output is measured using a lock-in amplifier. The output of the lock-in has a zero-crossing when the frequency of the signal generator equals the resonance frequency of the crystal. The feedback loop is closed by applying the output of the lock-in to the frequency modulation input on the signal generator. In this way, the frequency f_0, about which the high frequency signal is modulated, stays locked to the resonance frequency of the crystal. We also measure changes in the Q of the system by measuring the DC voltage which is at the output of the diode detector. We expect that the Q is proportional to the square root of this voltage, where Q is that resulting from the quality factors of the quartz, the circuit, and the helium film adsorbed to the crystal adding in parallel.

3. EXPERIMENTAL RESULTS AND DISCUSSION

Fig. 1 shows the frequency as a function of the temperature for the Au and the Cs crystals for the case a particular fixed number of ^{4}He atoms in the cell. The solid (open) symbols are for cooling (warming). The sensitivity of the crystals, due to their operation in the third harmonic, is approximately 1.8 Hz per layer of normal ^{4}He.

The Au crystal frequency decreases (Fig. 1a) as the temperature is reduced from 2.0 K and there is a KT transition at approximately 1.4 K which results in the frequency rapidly increasing by about 2 Hz due to the decoupling of superfluid mass from the substrate. As the temperature is lowered further, the frequency rapidly drops as the condensation temperature, T_N (≈ 1.3 K), is approached for the fixed number of atoms in the cell and the film thickness grows rapidly. For $T < T_N$, the vapor is saturated and the film thickness remains constant at a value which is determined by the height of the Au crystal above the bottom of the cell. The Au frequency increases
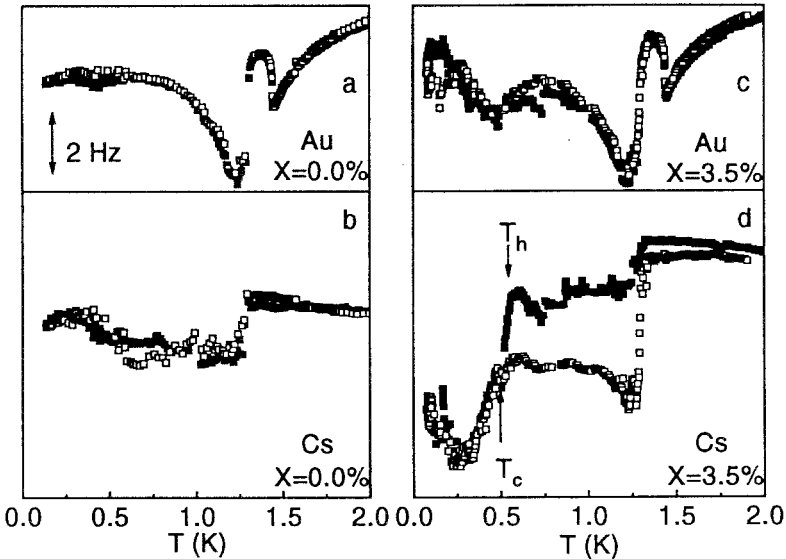
Fig. 1. Quartz crystal frequency *vs.* T(K) for (a) Pure ^{4}He - Au, (b) Pure ^{4}He - Cs, (c) 3.5 % ^{3}He - Au, (d) 3.5 % ^{3}He - Cs.

as the temperature is lowered below T_N because the superfluid fraction increases as T decreases. There is no hysteresis apparent on warming.

The behavior of the Cs frequency is very different (Fig. 1b). There is no growth of the film upon cooling from 2 K down to $\approx T_N$. There is no sign of a KT transition at any temperature. As T_N is reached, the Cs frequency drops by about 2 Hz. We interpret this to be due to a film of ^{4}He adsorbing to non-cesiated less sensitive parts of the quartz crystal and perhaps to the (possibly somewhat rough) Cs surface at and below T_N. Since the Cs frequency does not change much for T $\leq$ 1.25 K, we infer that any ^{4}He film adsorbed to the Cs must be very thin, certainly too thin to show superfluidity. For this case of pure ^{4}He, we observe no hysteresis of the Cs frequency as a function of temperature.

We have measured the Au and Cs frequencies as a function of temperature for a number of different ^{3}He concentrations in the range X=0.001 - 0.750 where X=N_3/(N_3+N_4) is the concentration in the cell. We measure the frequency as the cell is cooled from 2K down to ≈ 50 mK and then as the temperature is increased back up to 2K. In Fig. 1c-1d, we show the data for X=0.035 (3.5%). The effect of adding ^{3}He to the cell on the Au frequency can be seen for this concentration (Fig. 1c) only at low temperatures. Near 700 mK the Au frequency begins to drop as the temperature is lowered due

to ^{3}He adsorbing out of the vapor into the ^{4}He film. We interpret the increase observed for $T \leq 450\,mK$ as likely due to some of the ^{3}He moving from the bulk states to the liquid-vapor surface states in the ^{4}He film and the beginning of phase separation. Presumably, the coupling between the ^{3}He atoms and the shear motion of the crystal is less when the ^{3}He is at or near to the free surface of the ^{4}He film since the film thickness is large enough to be comparable to the viscous penetration length. We observe no hysteresis when warming the Au crystal.

The behavior of the Cs crystal frequency as a function of temperature is quite different (Fig. 1d). Upon cooling, the behavior is similar to that of the pure ^{4}He case until the temperature reaches the vicinity of $500\,mK$, for this concentration. The Cs frequency rapidly drops upon cooling through the temperature labeled T_h. Once the temperature is cooled below T_c, the frequency continues to decrease, but the rate of change of the frequency with temperature is decreased. At a temperature near $250\,mK$ the Cs frequency levels off and then begins to increase as the temperature is decreased further. This behavior is qualitatively similar to the low temperature behavior of the Au frequency. Note that the Cs frequency minimum is at $250\,mK$, whereas the Au frequency minimum is at approximately $500\,mK$. Upon warming a hysteresis loop opens up at T_c and then closes when T_N is reached ($\approx$ $1.3\,K$). The behavior of the Cs frequency during warming between $1.0\,K$ and $1.3\,K$ shows curvature due to the increase of the normal fluid fraction as the temperature is increased which indicates that a thick ^{4}He film is present on the Cs surface. We believe that T_c, or possibly T_h, is a measure of the reentrant wetting temperature and so upon cooling, the Cs is nonwet above T_h. When the temperature is raised back above T_c, the Cs stays wet because the helium film is in a metastable state. When the temperature rises above T_N, helium goes into the vapor from all surfaces and the Cs surface becomes devoid of helium. The hysteresis loop is reproducible. Recently, Ross et al.[8] have carried out a similar experiment, obtained similar data, confirmed this general behavior, and extended the analysis to claim the possible presence of ^{3}He at the He-Cs interface. We did not conduct such an analysis due to uncertainty in σ_{sl} and the film concentration.

In Fig. 2a-2b, we show the Au and Cs frequencies as a function of temperature for cooling and warming for $X = 10.65\%$. The effect of the increase in ^{3}He in the cell on the Au frequency is to increase the depth of the frequency minimum which is at approximately $500\,mK$. We note from more extensive measurements that the temperature of this frequency minimum increases with the concentration X. Although the T_{min} vs. X coordinates do not match the bulk phase separation curve very well, for $X \gg 6\%$ the frequency minimum is likely associated with phase separation in the (saturated)
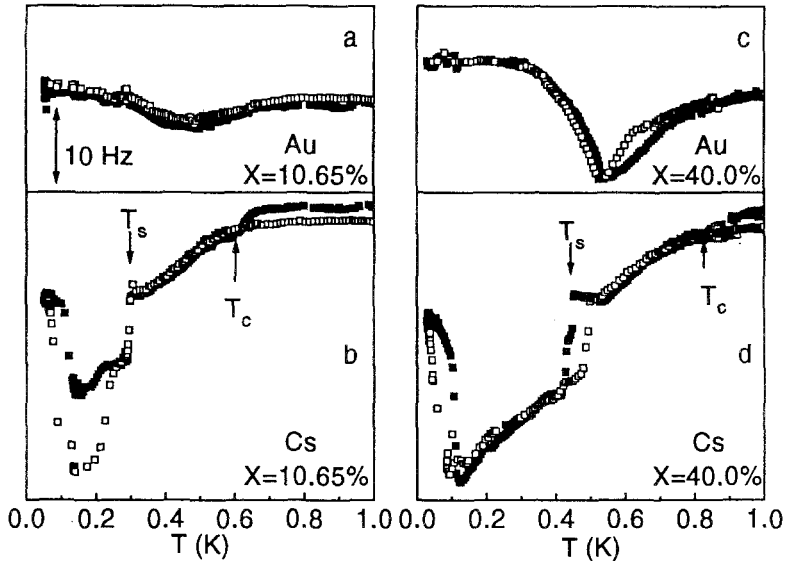
Fig. 2. Quartz crystal frequency *vs.* T(K) for (a) 10.65 % ³He - Au, (b) 10.65 % ³He - Cs, (c) 40.0 % ³He - Au, (d) 40.0 % ³He - Cs.

film. For the Cs case, the reentrant wetting occurs at a higher temperature than for the case of X = 3.5%, as is evident in Fig. 2b. Upon cooling below T_c , the frequency decreases and at a temperature labeled T_s there is a sharp downward shift in the frequency. Below T_s the frequency continues to decrease and then there is an increase in the frequency near 120 mK. There is some hysteresis upon warming from the minimum temperature to T_s.

For all concentrations x ≥ 8.0%, the cooling frequency data for the Cs crystal exhibit steps at a concentration dependent temperature, T_s. For X ≥ 25% (Fig. 2d shows data for X = 40%) there are *two* steps which are apparent on cooling. It should be noted that upon warming, there is hysteresis in the vicinity of the step. In particular, the frequency jumps up at a higher temperature than the temperature at which the frequency jumped down and there is typically only one step up. The rapid rise in the frequency at the lowest temperatures in the range 120-150 mK which is observable for most of the concentrations is not presently understood. Relatively long time constants are present for T ≤ 150 mK. The downward steps in the frequency might be due to a loading of ³He bound states at the Cs surface (so-called substrate states) which have been predicted to exist[9] but have not been unambiguously observed. For this to be the case, the potential seen by the ³He must be a function of X. One expects that the
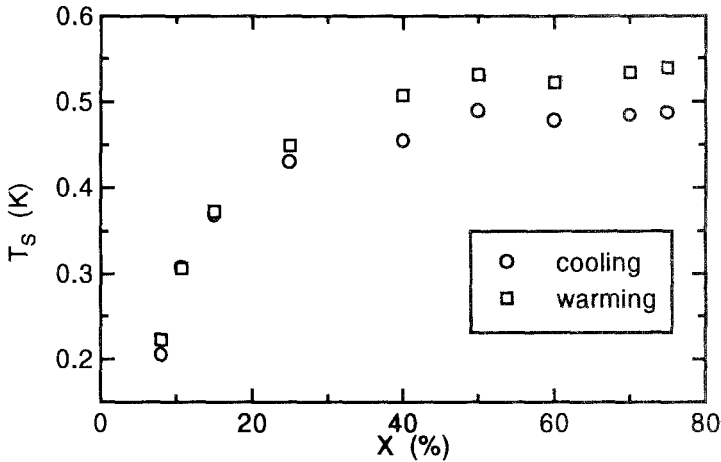
Fig. 3. T_s vs. concentration. The T_s, X coordinates are similar to the bulk phase separation curve for $X \leq 15$ %, but not for higher concentrations.

occupation of the predicted substrate state would be a thermally activated process, but the data show sharp features as a function of temperature. The occupation may accelerate once it commences as the temperature is lowered, as might be the case if the ^{3}He energetics were altered by finite occupation. The hysteretic behavior of T_s as a function of X is shown in Fig. 3.

ACKNOWLEDGEMENTS

REFERENCES

1. E. Cheng, M.W. Cole, W.F. Saam and J. Treiner, *Phys. Rev. Letters* **67**, 1007 (1991).
2. P.J. Nacher and J. Dupont-Roc, *Phys. Rev. Letters*, **67**, 2966 (1991).
3. K.S. Ketola, S. Wang and R.B. Hallock, *Phys. Rev. Letters*, **68**, 201 (1992).
4. E. Cheng, M.W. Cole, J. Dupont-Roc, W.F. Saam and J. Treiner, *Rev. Mod. Phys.* **65**, 557 (1993).
5. M.S. Pettersen and W.F. Saam, *J. Low Temp. Phys.* **90**, 159 (1993).
6. K.S. Ketola and R.B. Hallock, *Phys. Rev. Letters* **71**, 3295 (1993).
7. M.J. Lea, P. Fozooni and P.W. Retz, *J. Low Temp. Phys.* **54**, 303 (1984).
8. D. Ross, P. Taborek and J.E. Rutledge, preprint, 1995.
9. J. Treiner, *J. Low Temp. Phys.* **92**, 1 (1993).