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Ellipsometric Study of Superfluid Onset in Thin Liquid ^{4}Helium Films

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We have developed a modulated null ellipsometer capable of measuring single layers of adsorbed ^{4}He films at 1.4 K. The small optical index of liquid helium, the extreme sensitivity to temperature gradients, and the requirement of sub-monolayer stability over many hours presents significant experimental challenges, which will be briefly discussed. The main goal of our experiments is to independently measure the superfluid and normal coverage in thin adsorbed ^{4}He films. This is a particularly important issue for helium films on intermediate strength substrates such as rubidium and thin cesium, where previous measurements indicate that prewetting and the Kosterlitz-Thouless transition interact strongly, and the K-T transition appears to have non-universal features. Independent determination of the superfluid and normal fraction can be accomplished by using the ellipsometer in conjunction with a quartz crystal microbalance (QCM). QCM measurements rely on viscous coupling of the fluid layers, and therefore respond only to the normal component of a ^{4}He film. In contrast, the ellipsometer is sensitive to the total thickness, independent of the state (superfluid or normal) of the film. By combining the QCM and ellipsometric measurements we can determine the total coverage, the normal fluid component and thus the superfluid fraction.

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1. INTRODUCTION

Ellipsometry is a technique which exploits the change in polarization of light reflected from a surface to measure a film thickness or index of refraction. The relevant parameters are the wavelength of the incident light, the angle of incidence, the indices of refraction of the reflecting surfaces and their thickness. If these parameters are known, Fresnel's equations can be solved to predict the reflected polarization for a given incident polarization. Ellipsometry is a popular, precise, non-invasive method of thin film measurement. Previous ellipsometric measurements of ^{4}He films[1−4] have taken place at saturation and have sought to measure the thickness of relatively thick films at a given distance above the bath. These experiments were able to measure the change in hundreds of layers of helium over distances on the scale of centimeters. More recent[5,6] ellipsometric measurements have focused on the adsorption of other gases; Xe, Kr, Ar, and N_2, onto substrates like silicon and graphite. These experiments have yielded monolayer resolution and the isotherms show strongly bound individual layers. The index of refraction of the liquid phase of these gases in the visible spectrum is about n=1.3 as compared to helium which has n= 1.026. This difference in the optical index results in a reflected signal that is approximately 100 times smaller for helium than other rare gases.

The low value of n and the resulting small ellipsometric signal makes measuring a helium adsorption isotherm with ellipsometry quite challenging. The conventional tool for this type of measurement is a quartz crystal microbalance(QCM). The high-Q oscillator is capable of resolving fractions of a monolayer of helium by tracking small shifts in its resonant frequency. An important limitation of the QCM is that it responds only to the portion of adsorbed fluid that is viscously clamped to the surface, i.e. the normal fraction. As a thin helium film undergoes a Kosterlitz-Thouless transition and goes superfluid, it decouples from the surface and the resonant frequency of the QCM rises. Previous experiments[7,8] on intermediate binding strength substrates of rubidium and thin cesium, indicate deviations from a standard K-T transition. The superfluid transitions are hysteretic and more importantly the size and temperature dependence of the transitions do not agree with K-T theory. Isotherms of this system taken with a QCM are difficult to interpret because the superfluid onset occurs almost concurrently with the wetting thin/thick transition. The ellipsometer was initially developed to resolve these issues. By measuring the total coverage of the film with the ellipsometer and the normal fraction with the QCM we should be able to uncouple the wetting and superfluid transitions.

2. EXPERIMENT

The modulated null ellipsometer we designed is a common[9] polarizer-compensator-sample-analyzer system, as shown in Figure 1. It uses a HeNe laser as its light source and a photomultiplier tube(PMT) as its detector. The input optics are arranged such that the incoming elliptically polarized

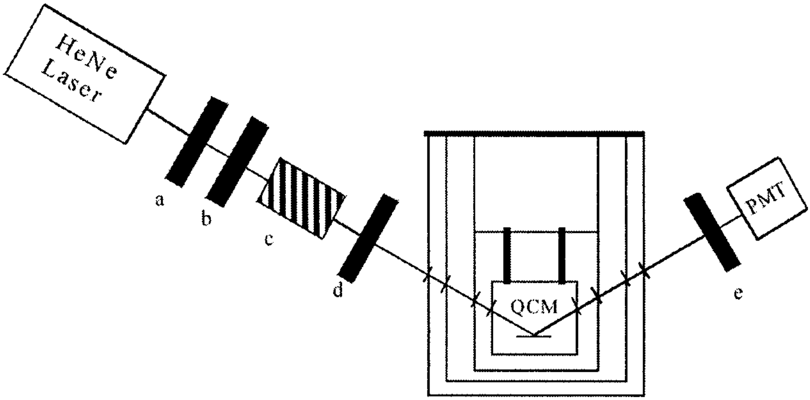

Fig. 1. Schematic of modulated null ellipsometer. The linearly polarized 632 nm light from the laser is turned circular by the first $\lambda/4$ plate(a). The first polarizer(b) does the coarse polarization adjustment and then the Faraday modulator(c) makes fine adjustments to the polarization, as well as adding a small modulation to the signal. A second $\lambda/4$ plate(d) makes the beam elliptically polarized and it then enters the optical cryostat to reflect off the QCM. The reflected linearly polarized light is then nulled with the second crossed polarizer(e) and measured with a photomultiplier tube.

beam is converted to linearly polarized light upon reflection. The first $\lambda/4$ plate directly after the laser is intended to take the linearly polarized light from the laser and make it circularly polarized light from which we can select the polarization we need without changing the intensity. An elliptically polarized beam is easily formed with a Glan-Thompson polarizer and first order $\lambda/4$ plate allowing us a coarse control of both the eccentricity and orientation of the ellipse. The Faraday modulator after the polarizer consists of a piece of SF-57 glass inside a magnet coil. The high Verdet constant of the glass allows fine adjustment, with a precision greater than .001° of the beam polarization by simply changing the current in the coil. The QCM sits at the center of our optical cryostat which has fixed windows at 60°. The gold electrode on the QCM, which will eventually be covered with evaporated cesium or rubidium, acts as the reflecting surface. The measured PMT signal can be nulled by adjusting the 2 polarizers and $\lambda/4$ plate. A small AC current is run through the coil to modulate the signal about the null. The PMT signal is read with a lock-in amplifier. A feedback loop takes the lock-in output as the error and adds a DC correction into the modulator to keep the measured signal at a minimum. This DC correction current is proportional to the angle of polarization and is the measurement signal. We calculated the faraday modulator would have to rotate the polarization about 10 millidegrees for each monolayer of helium adsorbed on a gold surface. This was done by solving Fresnel's equations for a given thickness of helium on bulk gold assuming ideal reflecting surfaces and neglecting the windows. Once the feedback loop stabilized, pure He gas was adiabatically added to the experimental cell. As the pressure and consequently the chemical potential begin to rise, a film adsorbs onto the substrate and begins to thicken. The QCM resonant frequency, and the correction current in the Faraday modulator required to keep the signal nulled, are recorded along with the current pressure in the cell. Ideally we would like to perform an isotherm above the λ-point, on bare gold, to calibrate the ellipsometer with the known QCM measurement. An isotherm below $T_\lambda$would then show the ellipsometer's ability to measure only the total coverage, regardless of the superfluid transition. The last step would be to perform these isotherms on rubidium and thin cesium to decouple the wetting and superfluid transitions observed previously.

3. RESULTS AND DISCUSSION

The resolution and stability of the ellipsometer is directly related to how well the light can be nulled. Birefringence, due to residual or induced stress, in the optics and windows was the most difficult issue to resolve experimentally. In travelling through the cryostat, the beam must pass eight windows: four vacuum windows and four radiation shield windows. Custom low stress mounts were designed for the vacuum windows which were made of super-precision annealed SF-57 glass, chosen for its low stress-optic coefficient. The radiation shield windows are IR absorbing KG-1 glass, which again had to be precision annealed. Glan-Thompson prism polarizers were chosen over polaroid sheets to gain an order of magnitude in the extinction ratio. Temperature controlling the faraday modulator and $\lambda/4$ plate to a few millidegrees also became necessary to attain sub-monolayer resolution. Lastly an OD3 filter was placed directly in front of the laser in order to avoid heating the gold and helium in the experiment. The 3 mW HeNe laser beam is attenuated to a 1 μW incident beam on the QCM. The majority of this is reflected off the gold so the sample is only capable of absorbing power on the order of a few nWs. Figure 2 shows the results of a recent isotherm at T= 1.48 K. The goal was to match the QCM's resolution but be able to measure both the normal and superfluid components of the film together with the ellipsometer. The graph shows the two measurements are

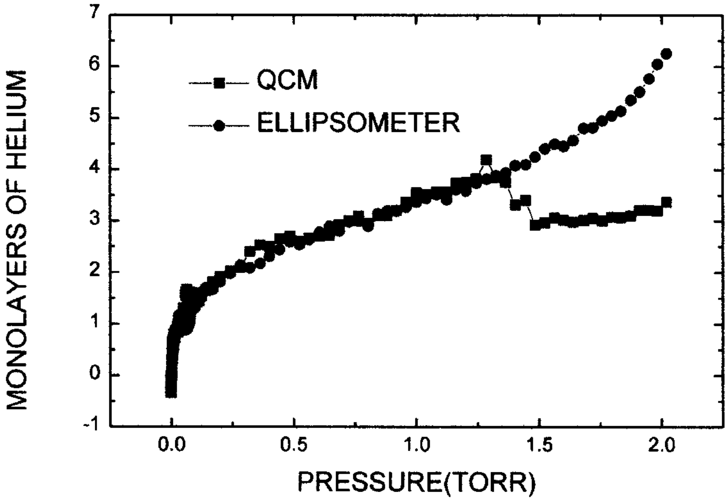

Fig. 2. Helium adsorption isotherm at 1.48K on gold and silicon. The QCM signal on gold shows the superfluid transition after about four layers. The ellipsometer signal taken at the same time but on a silicon substrate is invariant to the superfluid fraction and continues to measure just the total coverage. By comparing the two signals we can make a precise measurement of the superfluid fraction.

almost identical before the superfluid onset, with sub-monolayer resolution. Superfluidity brings the familiar decoupling of the helium from the QCM and characteristic drop in frequency, however the ellipsometer continues to measure the total film thickness. For this particular isotherm, both a QCM and a silicon wafer were placed in the cryostat on a rotator above the cesium evaporator. The QCM was intended to be a thickness monitor for the cesium when it was evaporated onto the silicon. The two curves shown in Figure 2 were not taken off of the same surface, but were taken simultaneously at the same chemical potential. Both gold and silicon are such strong surfaces that they should exhibit very similar wetting properties.

4. CONCLUSION

Our ellipsometer can detect single monolayers of liquid helium adsorbed onto a substrate. It is invariant to the superfluidity of the film and thus a very valuable tool. Used in parallel with a QCM we can now precisely determine the superfluid fraction of a film. Future experiments on intermediate binding substrates should provide critical data to explain our non K-T transitions previously observed.

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REFERENCES

1. A.C. Ham and L.C. Jackson, *Proc. R. Soc. Lon.* **Ser-A240(1221)**, 243-64 (1957).
2. O.T. Anderson, D.H. Liebenberg and J.R. Dillinger, *Phys. Rev.* **117**, 39-42 (1960).
3. D. Hemming, *Canadian Journal of Physics* **49**, 2621-29 (1971).
4. D.G. Blair and C.C. Matheson, *Physica B&C* **80**, 541-9 (1975).
5. J.W.O. Faul, U.G. Volkmann and K. Knorr, *Surface Science* **227**, 390-4 (1990).
6. H.S. Youn, X.F. Meng and G.B. Hess, *Phys. Rev. B* **48**, 14556-76 (1993).
7. J.A. Phillips, D. Ross, P. Taborek and J.E. Rutledge, *Phys. Rev. B* **58**, 3361-70 (1998).
8. J.E. Rutledge, J.A. Phillips and P. Taborek, *Physica B* **280**, 78-9 (1993).
9. C.C. Matheson, J.G. Wright, R. Gunderman and H. Norris, *Surface Science* **56**, 196-21 (1976).