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A low drift high resolution cryogenic null ellipsometer

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A cryogenic ellipsometer capable of measuring submonolayers of liquid helium has been developed. Methods for controlling drift and noise created by the cryostat windows and ellipsometer optics are discussed in detail. The cryostat vacuum windows were made from SF-57 glass due to its low stress-optic coefficient. Custom low-stress vacuum window mounts were designed and all windows were temperature controlled. Placing the compensator wave plate inside the cryostat was necessary to avoid room temperature fluctuation induced noise. These steps produced 12 h drifts in the measured polarization of less than 0.002°. A helium adsorption isotherm taken at 1.5 K on gold is presented to show the high resolution obtained once noise and drift associated with window birefringence is minimized. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1807591]

I. INTRODUCTION

Ellipsometry is a high precision method for surface characterization and thin film measurement. In this technique light prepared with a specific polarization is incident on a surface and the polarization state of the reflected light is analyzed. Often the goal is to measure the thickness of a film formed on an initially bare reflecting surface. In many cases, the surface and film under study must be isolated from the ambient conditions in the laboratory. Consequently the incident and reflected beams must pass through one or more windows. Each window, and in fact each optical element, has the potential to introduce stray polarization changes that can ultimately limit the resolution and accuracy of the ellipsometric technique. More generally, stray polarization shifts will degrade any polarization sensitive measurement and will at some point dominate the systematic errors and resolution in all experiments in this category. This paper discusses the steps we have taken to minimize the stress induced birefringence in the several windows in an optical cryostat and in a quarter wave plate and Faraday modulator used to prepare the polarization state of the incident beam in a cryogenic ellipsometer we have developed over the last two years.

The experimental goal which motivated the development of our ellipsometer is the measurement of the thickness of a liquid helium film as it grows from the vapor onto a variety of substrates, including bulk alkali metals and clean surfaces of single crystals. Our interest in these experimental systems stems from their rich phase behavior, which includes wetting and prewetting transitions, superfluid/normal transitions, and layering. The characteristic features of these transitions are thickness changes of approximately one monolayer, so our experiments require submonolayer resolution. Previous experiments[1,2] have utilized ellipsometry to study layer-by-layer film growth in cryogenic systems such as Xe, Kr, and N_2 at temperature above 40 K. Our experiments with liquid helium thin films involve additional experimental challenges, including much more stringent requirements for both stability and resolution of the ellipsometer signal. Helium has an index of refraction of approximately 1.028 while the index of Krypton is approximately 1.3. The ellipsometric signal per layer of film is therefore more than an order of magnitude smaller for helium than for Krypton and the other classical liquids. This resolution requirement is compounded by the sensitivity to long term drift of our experiment. Typical experimental runs in which we perform isotherms from vacuum to the saturated vapor pressure can take twelve or more hours. Any variation in the polarization shifts generated by the optical components over this time period will introduce a signal that is indistinguishable from a variation in the helium film thickness. Achieving submonolayer resolution for helium films thus required careful attention to minimizing stress and temperature variations in the cryostat windows and the ellipsometer components.

Although glass is nominally isotropic, nonuniform strains cause it to become birefringent. Glass windows in cryostats and vacuum systems are subject to temperature and pressure gradients,[3] thermal drifts due both to the ambient laboratory environment and deliberate temperature changes in the experiment, and large nonuniform clamping forces needed to form vacuum tight seals between the window and the rest of the apparatus. Even if external forces are minimized, most optical glass is birefringent due to strain frozen-in during the manufacturing process. If these effects were static and spatially homogeneous, the birefringence could, in principle, be measured and compensated for.[4,5] In practice, however, we found that the birefringence was spatially nonuniform, had long relaxation times, and was strongly affected by small temperature fluctuations, so treating the birefringence of the optics as a background signal which could be subtracted using a model was not feasible.

Rather than correct for the effects of birefringence, we attempted to reduce it to negligible levels and to minimize drift and fluctuations by temperature regulation of the optics. Some of the important techniques and procedures to accom-

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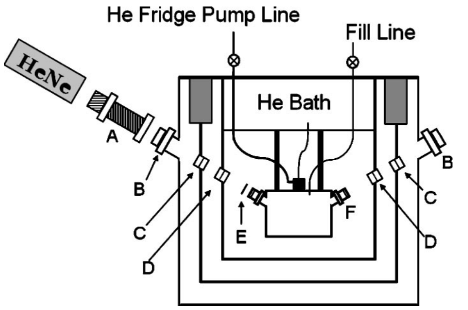

FIG. 1. Experimental apparatus. The Faraday modulator and polarizer (a) are locked together with their axes aligned. The beam passes through eight windows: the outer vacuum wall windows (b), the liquid nitrogen radiation shield windows (c), the liquid helium radiation shield windows (d), and the experimental cell windows (f). The compensator $\lambda/4$ plate (e) is attached to the low temperature cell just outside the window. The outer vacuum can have a diameter of 24 in., while the diameter of the low temperature cell is 6 in..

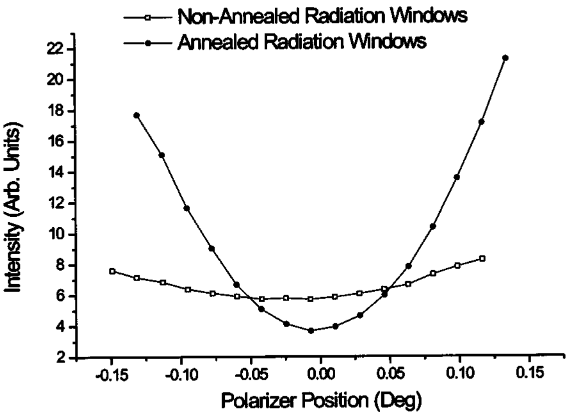

FIG. 2. Intensity of the reflected ellipsometer beam as a function of polarizer angle showing the effects of annealing the IR blocking radiation shield windows. The data were taken by rotating the input polarizer about the null point and measuring the intensity. The null using the annealed windows has a lower minimum and higher curvature, which corresponds to greater angular resolution in locating the null point. A shallow bright null is indicative of the birefringence in the windows which depolarizes the beam.

plish this were: (1) use of SF-57 glass[6] for windows where possible.[7] SF-57 has a particularly low stress-optic coefficient which measures how birefringent a medium becomes for a given stress; (2) super-precision annealing[8] of all windows to eliminate residual stress frozen into the glass; (3) custom designed low stress mounts for all of the windows[9,10] similar to those described by Mollenauer *et al.*;[11] (4) millidegree temperature control of all of the room temperature windows and installation of the compensator wave plate in the low temperature portion of the apparatus to reduce thermal drift; and (5) careful alignment of the residual birefringence axis of the modulator with the axis of the polarizers. Section II below describes the details of the apparatus and Sec. III shows the results of ellipsometric isotherms with helium and documents submonolayer resolution.

II. APPARATUS

Our optical cryostat, shown schematically in Fig. 1, was designed for ellipsometry on low temperature substrates. The experimental cell hangs from three thin walled stainless steel legs which thermally isolate the cell from the 4.2 K bath. The cell is equipped with an evaporative refrigerator on the top flange which allows the cell to reach temperatures as low as 1.3 K. A key requirement for reaching this base temperature is reducing the radiative heat load on the cell. For this we use two radiation shields; an inner one in thermal contact with the liquid helium bath and an outer shield in contact with the liquid nitrogen jacket. Although these shields are essential for reducing the heat load on the cell they add four additional windows that the ellipsometric beam must travel through, each of which is a potential sources of depolarization.

The radiation shields are made of gold plated copper. The beam passes through 1 in. windows made of KG-1 infrared absorbing glass, as shown Figs. 1(c) and 1(d). KG-1 is a short pass optical filter with a cutoff at 800 nm. These windows allow visible light to pass through to probe the sample while absorbing longer wavelength IR radiation. By thermally anchoring the windows to the shield and thus back to the baths, thermal radiation from room temperature and from the nitrogen temperature shield is minimized. The as-purchased KG-1 windows were strongly birefringent; strained regions could be easily observed by placing the windows between crossed polarizers. The strain was removed by an annealing procedure which consisted of heating a glass blank up to its melt temperature, 873 K, and then lowering the temperature by 0.02 K/h until 473 K. This procedure is called super-precision annealing. The windows were subsequently cut from the blank and polished. These mechanical operations did not introduce a new strain because the temperature was always far below the melt temperature. The improvement of the ellipsometer null due to annealing the IR blocking windows is illustrated in Fig. 2.

The radiation shields are not vacuum tight, so the IR blocking windows do not require a vacuum seal. In an attempt to minimize strain generated by thermal contraction, these windows sit in a loose recess and are held in place by gravity and a thin ring of vacuum grease to improve the thermal conduction to the metallic shield. In contrast, the windows in the room temperature outer vacuum wall of the cryostat and the windows in the low temperature cell must be rigorously vacuum tight, and must be able to withstand pressure differences of at least 1 atm.

The windows on the low temperature cell are made from 1 in. diameter $\times$ 3/8 in. thick super-precision annealed SF-57 glass, as shown in Fig. 3. This glass can be purchased directly from Schott Glass,[6] SF-57 glass was chosen for its exceptionally low stress-optic coefficient of $2\times10^{-8}\ \mathrm{mm}^2\ \mathrm{N}^{-1}$, which is approximately two orders of magnitude smaller than that of other conventional optical glasses such as BK-7. In order to avoid strain due to thermal contraction and compression seals, a copper flange was fabricated with a thin (0.004 in. wall thickness) tube to hold the SF-57 window. The robust base of the flange was sealed using an indium O ring into the body of the cell. Stresses in the bulk copper base cause deformations in the thin copper walls rather than squeezing the glass window. The glass is attached to the thin walls via a small shoulder in the tube wall and a thin bead of epoxy[12] to make a superfluid tight

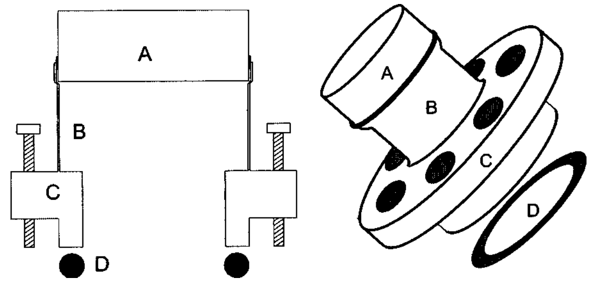

FIG. 3. Custom cell window. A precision annealed SF-57 window (a) epoxied into a 0.004 in. walled (b) holder. The copper base (c) is bolted onto the cell body with an indium O ring (d) to make a vacuum seal. The unusually thin walls were used to prevent strains in the base from affecting the window.

vacuum seal. The windows have been thermally cycled to 1.3 K and back to room temperature numerous times without developing leaks. The exterior of the cell is in vacuum while the inside pressure ranges from vacuum up to 1 atm. We have checked for possible pressure induced birefringence in the windows by measuring helium isotherms at 7 K, which is above the critical point of helium so no significant adsorption is expected. The ellipsometer signal was independent of pressure up to 1 atm, confirming that pressure induced birefringence is not a significant limitation to our measurements.

The final pair of windows on the outside of the cryostat are mounted into the vacuum vessel of the dewar using Conflat hardware. We found that commercially available Conflat viewports were unacceptable for our purposes: They suffered from large inhomogeneous frozen-in birefringence which depended sensitively on the temperature of the room and the torque used to tighten the bolts. We solved these problems by making a window using a 2 in. diameter, 5/8 in. thick superprecision annealed SF-57 disc mounted in a modified 3 3/8 in. Conflat blank as shown in Fig. 4. A stainless steel cup with an O ring groove was welded into the bored out Conflat blank. The outer windows remain at room temperature so a rubber O ring can be used to make the vacuum seal. The 1 atm pressure drop across the window provides a uniform and reproducible force which pushes the window against the O ring. Since the window only makes contact with the holder through the rubber O ring, strains created in bolting the Conflat knife edge into the copper gasket do not strain the window.

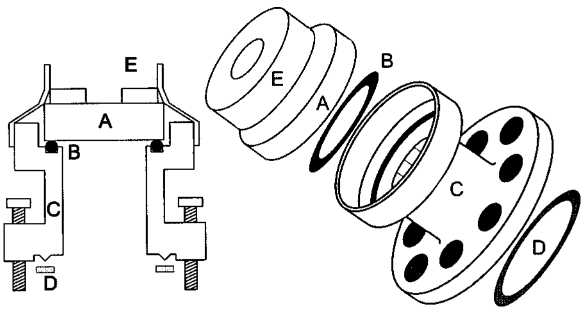

FIG. 4. Outer vacuum window. The precision annealed 2 in. SF-57 window (a) fits into the stainless steel holder (c) and maintains a vacuum tight seal via the rubber O ring (b). This is then bolted to the outer stainless steel vacuum can, with a copper gasket (d). Stresses induced by bolting the bottom plate are not transmitted to the glass. The temperature controlled aluminum donut (e) greatly reduces long term drift and noise. Flexible PVC shrink tubing helps hold the donut in place.

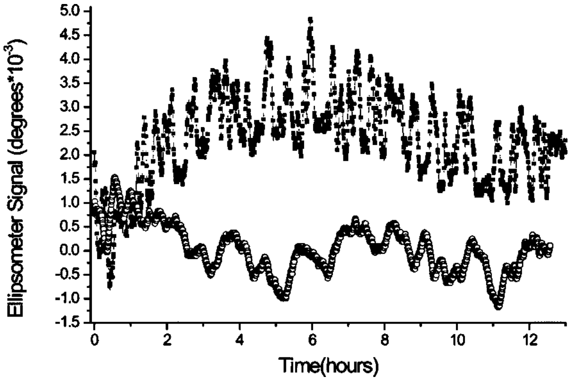

FIG. 5. The measured ellipsometer signal in millidegrees versus time. The ellipsometer is measuring a bare substrate in vacuum, and ideally would trace a horizontal straight line. The first curve (a) was taken before thermally regulating the outer windows and the bottom curve (b) with temperature control. They show how well the drift was constrained, noting that 1 Å of liquid helium on gold corresponds to a ~0.0022° shift.

These improvements in the design of the windows reduced the fluctuations in the ellipsometric signal to approximately ±0.004° over 10 h with a higher frequency component with variations of ±0.002° every 20 min, as shown in Fig. 5(a). These fluctuations are correlated with small changes in the ambient temperature. In particular, the oscillations with a 20 min period were caused by temperature changes of less than 0.2 K as the climate control unit in the lab cycled on and off. These variations in the ellipsometric signal were not simply proportional to the temperature deviation ΔT. Experiments in which we changed the temperature of the room by several degrees showed that the ellipsometer response was more nearly proportional to $d\Delta T/dt$ than to ΔT itself. The stability of the ellipsometer signal was greatly improved by actively controlling the temperature of the outer windows, as shown in Fig. 5(b). This was accomplished using an aluminum donut with a heater and thermometer which was placed onto the outer face of the window using vacuum grease to increase thermal contact. The inner window face was exposed to vacuum and only made contact with the rest of the apparatus through the low conductivity rubber O ring. The aluminum ring was maintained a few degrees above room temperature to within ±0.01 K using a feedback loop. PVC shrink tubing and foam was added for insulation, and a 3/8 in. hole was cut into this insulation for the beam to pass.

Our modulated null ellipsometer consists of the standard polarizer, compensator, sample and analyzer set up similar to that used by Volkmann and Knorr.[1] The same small deviations in room temperature that plagued the outside windows also affected these optical components. Experiments with a heat gun showed that by far the most thermally sensitive optical component was the compensator. The compensator is a first order quartz $\lambda/4$ plate designed for 632.8 nm light. This type of compensator consists of two nearly identical pieces of quartz aligned with their ordinary axes orthogonal to one another so that any change in retardation from one is compensated by an opposing shift in the other. Although first-order wave plates are known to be much less sensitive to temperature variations than multiple-order wave plates, we found that even the residual temperature dependence in our quartz wave plate was large enough to cause significant polarization shifts from small room temperature fluctuations.

Active temperature control of the wave plate outside the cryostat considerably improved the stability of the ellipsometer, but an even simpler and more effective improvement was to mount the wave plate to the outside of the low temperature cell just in front of the window. For our null ellipsometer, the $\lambda/4$ plate orientation is fixed with its axis at 45° to the plane of incidence, so placing it inside the cryostat, where it could not be adjusted, was not an issue. At temperatures below 10 K the thermal expansion coefficient of quartz becomes negligible and the cell temperature is regulated to 0.001 K. Initially we were concerned whether the fragile wave plate would survive the thermal cycling from room temperature to 1.3 K and back but no ill-effects have been observed; even the broadband antireflecting coatings remain intact. The two pieces of quartz which make up the compensator are press fit together without adhesive. Thermal cycling caused them to separate from each other but the optical performance was not degraded.

Another source of depolarization of the ellipsometer beam was traced to the Faraday modulator, which provides the modulation and fine tuning to the input linear polarization. It consists of a cylindrical piece of super-precision annealed SF-57 glass 2.5 in. long placed inside a solenoid. Even the lengthy annealing process is insufficient to completely remove all residual strain. Its effects are magnified by the 2.5 in. length of the modulator glass. This length of glass was necessary in order to get the needed polarization rotation of approximately 1° from the solenoid using less than 2 amps of current. The residual birefringence transforms an initially linearly polarized beam into a slightly elliptically polarized beam as it passes through the modulator. This effect can be minimized by rotating the polarizer and modulator with respect to each other until the polarizer's axis is aligned with the modulator's birefringent axis and then locking them together. It should be noted that we initially used terbium gallium garnet (TGG), in the Faraday modulator. TGG has a higher Verdet constant than SF-57, and is commonly used in commercial faraday isolators. We found, however, that TGG was very birefringent and that this birefringence was spatially inhomogeneous and also temperature dependent. TGG was therefore a poor modulation medium for our purposes.

III. RESULTS AND DISCUSSION

Cooling the experiment from room temperature to 1.3 K typically takes about 12 h. The ellipsometer is not used during this period because the thermal contractions that occur within the cryostat change the alignment. Once the temperature has settled, the ellipsometer is aligned and nulled. For a reflecting surface we use the gold electrode on a quartz crystal microbalance(QCM), that sits in vacuum in the experimental cell. The ellipsometric signal from this bare surface in vacuum is monitored for at least 12 h to make sure that the system has stabilized before beginning an isotherm. During the isotherm we adiabatically dose the cell with helium gas until saturation is reached, maintaining the cell temperature constant to within 0.001 K. A computer monitoring the experiment records the temperature, pressure, ellipsometer signal and other relevant parameters. A typical isotherm which shows both the ellipsometer signal and the QCM signal as a function of the reduced pressure P/P0 is shown in Fig. 6.

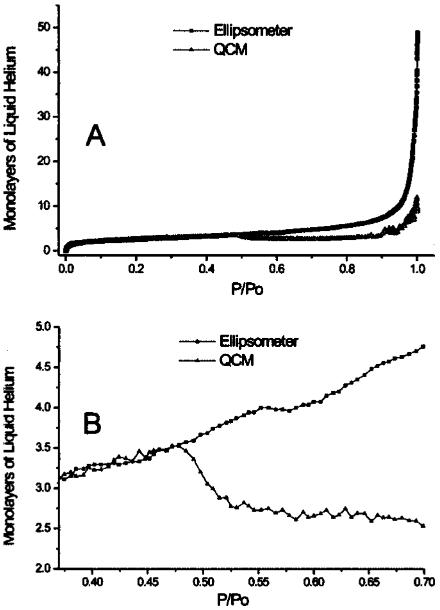

FIG. 6. (A) Helium adsorption isotherm on Bare Gold at 1.5 K from vacuum to saturation. (B) A closer look at the region around 0.5 P/Po. Below this point there is good agreement between the QCM and ellipsometer, both with sub-monolayer resolution. Above 0.5 P/Po a fraction of the film undergoes a superfluid transition and decouples from the QCM. The ellipsometer measures the total coverage, regardless of superfluidity up to saturation.

For an ideal geometry consisting of a flat semi-infinite substrate with known values of the optical index n_s+ik_s, and a flat, transparent film with optical index n_f, the thickness of the film can be computed from the ellipsometer signal using Maxwell's equations. The results of such a calculation show that the ellipsometer signal, is linearly proportional to the film thickness for films <500 Å thick. The measured polarization should shift $\sim 0.0032°$ per angstrom of adsorbed liquid helium. Uncertainties in our value of n_s+ik_s due to temperature dependence, surface roughness, and thin film effects introduce a corresponding uncertainty in the proportionality factor between polarization shift and the film thickness. One of the advantages of performing our ellipsometric measurements directly on the surface of a microbalance is that we have an independent measure of the adsorbed film thickness from the QCM signal which is simple to interpret as long as the film is in the normal state. We can empirically determine the proportionality factor between polarization shift and film thickness by demanding that the ellipsometric thickness and the thickness determined by the QCM agree when the film is non-superfluid. This results in the relation polarization shift $=0.0022°/\text{Å}$ which is 30% less than the value determined from the bulk optical constants of gold; this relation was used in the comparison of the ellipsometer and QCM signals shown in Fig. 6.

Figure 6 shows that for reduced pressures less than 0.455, the ellipsometer and QCM signals agree to within 0.1 layers, and that the point to point fluctuations for the two signals are of similar size. For values of the reduced pressure between 0.455 and 0.53, the QCM signal decreases due to decoupling of the superfluid component of the helium film as it undergoes the superfluid transition. The ellipsometer signal is smoothly increasing through this region because the optical constants do not change at the transition and the total film thickness is expected to monotonically increase as a function of pressure. The absence of any discernable feature in the ellipsometric signal at the superfluid transition indicates that the thermal perturbation caused by the incident beam power of approximately 1 μW is negligible. The deviation between the ellipsometric and QCM signals grows larger as the system approaches saturation at $P/P0=1$. Although the ellipsometric signal measures the total thickness of the film, the QCM is sensitive only to the normal part, and for thick films the response is nonlinear.

In summary, we have constructed a null ellipsometer with sufficient stability and resolution to measure submonolayer changes in the thickness of a helium film. Reducing window birefringence is critical to achieving the low-drift, low-noise signal needed for this level of resolution. The key steps required for this are annealing all of the window glass, utilizing low stress-optic coefficient glass when possible and using low-stress window mounts. We also temperature controlled all of the windows in the cryostat as well as the compensator wave plate.

ACKNOWLEDGMENTS

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