5. Alternatively, the relative total energy/shot passing through A_1 and A_2 can be determined with photon drag detectors placed immediately before and after the absorption cell. This permits the study of molecules that do not fluoresce and it also produces a stronger signal.
6. J. I. Davis and W. Clements, Eds., Lawrence Livermore Laboratory Laser Program Annual Report, 1974, UCRL-50021-74, pp.135 *ff*.

Simple ellipsometer design

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The optical community has recently shown considerable interest in simple, inexpensive means of characterizing the index and thickness of thin films.[1,2] In this Letter we present a simplified ellipsometer design which could make the flexibility of ellipsometry for the examination of thin films accessible to most laboratories.

Ellipsometry, in general, is the measurement of the polarization of light reflected from a surface. For light incident at a given angle ϕ on a homogeneous isotropic surface, the reflection can be characterized by the complex reflectances, r_p and r_s, of the components polarized parallel and perpendicular to the plane of incidence. An ellipsometer measures the ratio of these reflectances, $r_p/r_s \equiv \tan\psi \exp(i\Delta)$.

Conventional ellipsometry makes use of rotatable polarizers and a $\lambda/4$ plate to measure ψ and Δ for arbitrary, but usually fixed, incidence angles ϕ. In 1936 O'Bryan[3] described an alternative scheme in which ϕ and ψ are measured at the principal incidence angle, where $\Delta = \pm 90°$. This principal-angle ellipsometry employs only one polarizer and no $\lambda/4$ plate. Recently, Yamamoto and others[4,5] have discussed the use of principal-angle ellipsometry and described the tolerances of O'Bryan's ellipsometer design.

Figure 1 illustrates our modified design for a principal-angle ellipsometer which is easily assembled from standard optical components. L is the light source, which in our instrument is a He–Ne laser. Any collimated monochromatic source could be used, though a narrow beam is helpful, as discussed below. P represents a polarizer in a rotatable mount; we use a film-type polarizer. S is the sample mounted on R, a rotation stage aligned with its rotation axis intersecting the incident light beam. An adjustment micrometer translates the sample across the stage to allow its surface to intersect the same point. M is a spherical mirror with its center of curvature also aligned at the point of intersection. This fixed mirror slightly decollimates the reflected beam and returns it to the sample, through the polarizer, and off a beam splitter, B. We use a pellicle beam splitter, though a mirror with a central hole could also work well.

The principal angles are determined by rotating the polarizer and the sample stage until a null is reached at the detector, D. Detection can be done by focusing the beam onto a photodiode, by focusing it onto a screen, or by direct viewing. We have preferred the visual approaches for speed and convenience. Direct viewing is appropriate for precisely locating nulls from high-quality low-reflectance surfaces.

The major innovation in this ellipsometer is the spherical mirror, which replaces a gear-driven mirror mechanism in the previous design. The spherical mirror also eliminates the need for adjustments to correct for tilt in the sample. Tilt in the plane of incidence simply shifts the base line for measurement of ϕ, and tilt perpendicular to the plane of incidence only affects ϕ to second order.

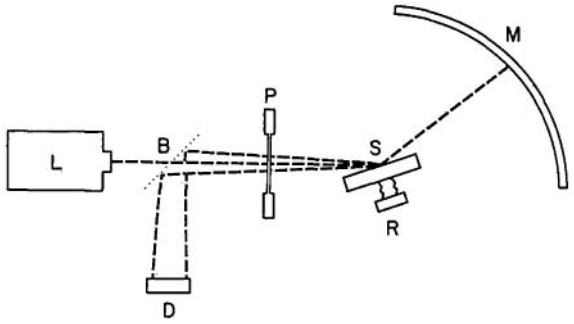

Fig. 1. Layout of simplified principal-angle ellipsometer: B, beam splitter; D, detector; L, light source; M, spherical mirror; P, polarizer; R, rotatable holder; S, sample.

The spherical mirror must be large compared to the sample and must have a very low f number. To cover the range $45° \leq \phi \leq 90°$, the mirror's diameter must cover $\geq 90°$ of arc. Several smaller mirrors can be arranged around the sample stage instead, although this complicates the initial alignment. The mirror decollimates the input beam, leading to a divergence of $\Delta\theta \simeq d/r$ where d is the beam diameter and r is the mirror's radius of curvature. The practical effect of this divergence is to reduce the intensity on the optical axis where the ellipsometric nulls should be observed.

The polarizer, whether a prism or film type, should have high efficiency, good uniformity, and low wedge. If an ordinary beam splitter is used, the polarizer should be tilted slightly ($\leq 1°$) so that the reflection of the beam from its first surface does not enter the detector.

The major components of our ellipsometer are an 8-cm diam film-type polarizer in a rotatable mount and a 19-cm diam mirror with a 15-cm radius of curvature. Though our instrument was assembled from components readily at hand, we have priced the total cost of these components at less than $2,000. This compares favorably with commercial ellipsometers which range upward from $10,000.

In preparing our samples for measurement, we have coated or deposited the thin films onto nontransparent substrates, usually aluminum or silicon. If a transparent substrate is used, it must be thick enough so that reflections from the back surface do not enter the detector (much thicker than the incident-beam diameter).

The principal angle and corresponding value of ψ are determined as differences of measured quantities. The sample stage may be zeroed by rotating it until the light beam is reflected directly back to the source. The beam is rotated back onto the spherical mirror, and the sample is translated via the micrometer until the reflected and incident beams again coincide. The polarizer and sample stage are now alternately rotated until a null is reached. The principal angle, ϕ_p, can now be read directly from the stage. An independent null can be found in the next quadrant of the polarizer, and the difference between the two measured polarizer angles is 2ψ (or $180° - 2\psi$, depending on which quadrants are used). At the principal angle we must have $\Delta = \pm 90°$. The sign ambiguity can be resolved by inserting a simple circular polarization filter between the sample and the mirror, as discussed by Yamamoto.[4]

We have chosen to use graphical means to reduce measured values of ψ, Δ, and ϕ_p to the desired optical parameters of the sample. We have programmed a desk-top computer to plot

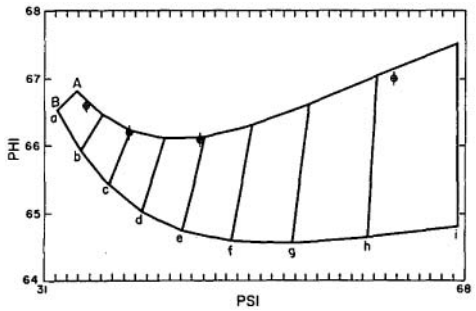

Fig. 2. Ellipsometry for cellulose nitrate coatings on silicon (see text).

ϕ and ψ (Δ determines the sign of ψ) for the largest principal angle of a multilayer film in which one layer is given a range of both thickness and complex refractive index.

As an example, we have measured the thickness and index of low-molecular-weight cellulose nitrate (RS 18-25, Hercules) spin coated on silicon. Observations of five uncoated substrates yielded $\phi_p = 75.48° \pm 0.04°$ and $\psi = 1.07° \pm 0.06°$, which are interpreted as representing 1.3 ± 0.2 nm of oxide ($n = 1.5$) over the bulk silicon with a nominal index of $3.86 + 0.091\,i$ (at 633 nm). Figure 2 shows the measurements for four cellulose nitrate films spin coated at 6000, 5000, 4000, and 3000 rpm from a 5% solution in cyclohexanone. Error bars indicate the typical precision of our instrument. Superimposed on the data are the values calculated for a uniform coating over the substrate structure described above. Lines A and B correspond to refractive indices of 1.50 and 1.55 respectively, and lines a through i correspond to thicknesses of 80–120 nm ($n = 1.50$) and 73–113 nm ($n = 1.55$) in steps of 5 nm. Graphical comparison of these data (including a data point at 2000 rpm) indicates a polymer index of $n = 1.505 \pm 0.003$ at 633 nm, in agreement with the manufacturer's estimate of 1.51.

Our primary use for this ellipsometer has been to measure the optical constants of layers which absorb 633-nm light. In such cases it is necessary to either evaluate samples of different thicknesses (to increase the number of measured parameters) or use transmission measurements through transparent substrates to determine the imaginary part of the index. We have also used this instrument to measure refractive indices at other wavelengths, by combining, for a series of samples, thickness determinations (made at 633 nm) with transmission and reflection measurements.

One important fundamental difference between conventional ellipsometry and the principal-angle ellipsometry we have described is the extent of knowledge of the substrate. Evaluation of ψ and Δ for a bare substrate completely characterizes its ellipsometric properties at the given incidence angle, regardless of the correctness of the assumed substrate model (i.e., the index depth profile). Since conventional ellipsometry keeps ϕ fixed, the substrate model is not critical. However, since ϕ_p varies considerably with film thickness, a previous measurement of ϕ_p and ψ for the uncoated substrate is not sufficient to support a principal-angle calculation without a detailed substrate model.

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Aluminum black films

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Over the years there has been interest in producing metallic black films. One of the most common has been made from gold. We report here that it is possible to make a similar black film from aluminum. This sort of film could have advantages because of its low atomic number and cost of materials. Also, at elevated temperatures it appears that the aluminum blacks are less likely to return to their shiny metallic appearance than blacks made from gold.

The aluminum blacks are prepared by vacuum evaporation of aluminum in a helium atmosphere at a pressure of 1 Torr. At this pressure the substrate must be ~15 cm from the source. Greater source–substrate separation tends to require lower pressures.

One way of characterizing metallic black films is to measure the scattered light for a given incident beam. We have done this using the 633-nm line from a He–Ne laser and the 477-nm line from an Ar-ion laser. In both cases the laser beams were polarized in the plane of observation, which is the plane defined by the incident beam and the line of observation. The scattered intensity is measured relative to the incident intensity with a detector composed of a 3.2-mm diam fiber-optic bundle coupled to a silicon solar cell operated as a linear detector.[2] The scattered intensity is measured at a number of angles with respect to the normal incident direction. The results for the scattered 477-nm laser light are shown in Fig. 1. The scatter angles are measured with respect to the incident beam. The asymmetry is probably due to the plane of the substrate being tilted away from normal with the source.

If one assumes azimuthal symmetry in the scattering about the incident beam in the case of normal incidence, an estimate of the total scattered light can be made. Assuming scattering into 2π sr the fraction of the incident intensity to be scattered is 0.02% for the 633-nm radiation and 0.03% for the 477-nm line. Comparison with other blacks in the literature,[1] such

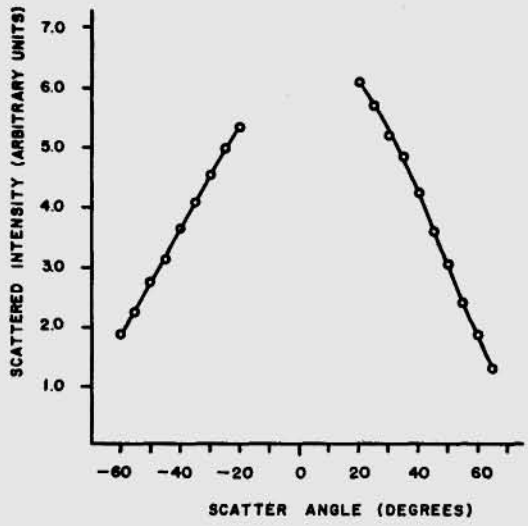

Fig. 1. Scattered intensity of 477-nm radiation measured with respect to the incident direction.