Dielectric Functions of Common YBCO Substrate Materials Determined by Spectroscopic Ellipsometry

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***Abstract*—Reference dielectric function data for several common $YBa_2Cu_3O_{7-\delta}$ (YBCO) substrate materials have been determined by spectroscopic ellipsometry over the range 250 nm - 750 nm. These materials include $LaAlO_3$, $BaZrO_3$, $NdGaO_3$, 9.5 mol% Y_2O_3-ZrO_2 (YSZ), $LaSrGaO_4$ (LSGO), and $(LaAlO_3)_{0.3}$-$(Sr_2AlTaO_6)_{0.7}$ (LSAT). The precision of the data was confirmed by comparing SE determined data for $SrTiO_3$ to published values. Agreement to the third decimal point was shown. These data have been used to characterize interfaces between YBCO and some of these materials by SE.**

I. Introduction

Since the development and subsequent advancements in methods used to deposit high quality epitaxial thin films of $YBa_2Cu_3O_{7-\delta}$ (YBCO), there has been a significant interest in the substrates used for these films. It is desired that the chosen substrate results in a film which has the optimum structural, transport and superconducting properties. Guo *et al.* [1] have completed a comprehensive study of structural compatibility between several substrate materials and YBCO. It was concluded that lattice matching between YBCO and the substrate was secondary in importance to the idea of structural similarity. That is, similarities in values such as the ionic distributions and distances should be the primary concern when selecting a substrate. This has significantly expanded the pool of substrates on which epitaxial YBCO can be grown.

With this wide selection of substrates available, characterization of these systems has become crucial. Information regarding crystallinity of the film, surface roughness, substrate/film interfacial roughness and the presence of reaction layers must be determined for each YBCO/substrate combination. Also, how these features affect the superconducting properties must be understood. In past years, characterization of the dielectric function of YBCO via optical methods has received much attention [2]-[6]. Specifically, the effect of oxygen content on the dielectric spectra has been investigated [3], [4]. More recently, work has commenced on optical characterization of the microstructures of YBCO films on various substrates using spectroscopic ellipsometry (SE) [7]. That entails using SE to depth profile these systems and extract information on reaction layers, surface roughness, etc.

In order to use SE to determine these microstructural parameters, it is necessary to have reference dielectric function data ($\varepsilon_1+i\varepsilon_2$) for the individual materials in the sample of interest. Typically, the SE measurement is taken from the near UV to the near IR energy range. Data for this range has been determined for YBCO [4]; however data for the various substrate materials is not as widespread. It is the purpose of this study to determine the dielectric functions of several YBCO substrates using SE.

Manuscript received August 27, 1996.
This work was supported by the Office of Naval Research under Grant No. N00014-94-1-0815.

II. Experimental Procedure

A. Apparatus

A diagram of the rotating analyzer ellipsometer used in this study is shown in Fig. 1. A xenon source is used because it provides a stable output over the wavelength range of interest, 250 nm - 750 nm. A double Czerny-Turner monochromator is used to select the wavelength. The polarizer and analyzer are quartz Rochon prisms, and a photomultiplier tube is used as the detector. The achromatic compensator is based on the design of King and Downs [8] and is made of vitreous silica with a MgF_2 coating. This is a three reflection quarter wave system that can produce an approximate 90° phase shift of the incident light over a wide wavelength range. The entire system is interfaced with a Gateway 2000 486 DX2 computer which controls the data acquisition.

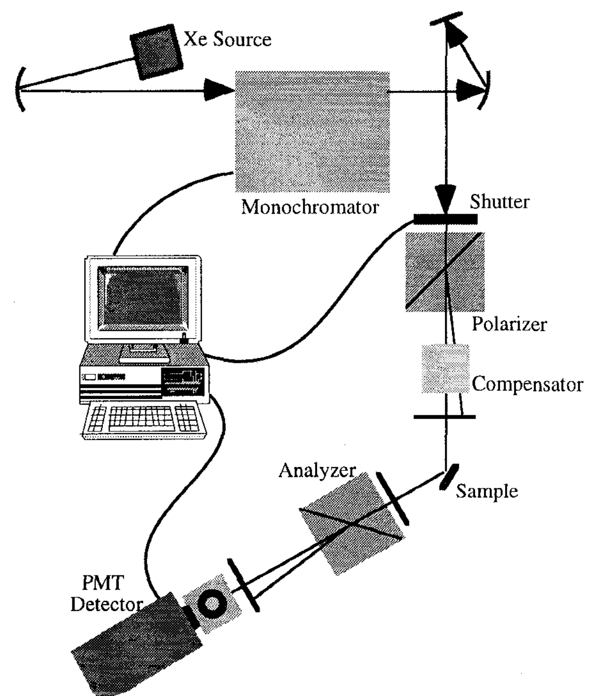

Fig. 1. Diagram of the SE used in this study.

B. Background of SE

In general, SE is a technique where linearly polarized light at several known wavelengths is reflected from a sample and the changes in the relative phase and amplitude of the parallel (p) and perpendicular (s) components of the light are measured. These changes in the polarization state are characteristic of the depth profile of the dielectric function of the sample. The measured ellipsometric parameters, Δ and Ψ, are related to the dielectric function by:

$$\rho \equiv {}^{r_p}\!/_{r_s} = \tan\Psi \exp(i\Delta), \quad (1)$$

where ρ is the complex reflectivity coefficient and r_p and r_s are the complex Fresnel reflection coefficients for the parallel and perpendicular components of the light, respectively.

The parameter Δ is defined as the difference between the phase shifts of the parallel and perpendicular components of the light. For light reflected from a transparent material Δ will be near 0° or 180°. De Nijs and van Silfhout [9] showed that in rotating analyzer ellipsometry (RAE), the experimental errors are proportional to 1/sinΔ. Thus, in measuring transparent materials using RAE the errors can be very significant. Chindaudom [10] overcame this problem by inserting an achromatic compensator into the RAE system. In doing so, an artificial phase retardation of ≈ 90° occurs and the incident light to the sample is circularly polarized rather than linearly polarized. There is still little change in Δ after reflection, however in this case Δ is near 90° and the error is significantly reduced.

The procedure used to measure transparent materials involved obtaining two sets of SE spectra. First, SE data were obtained without the sample and with the ellipsometer in the straight through position (90° angle of incidence). This allowed the exact phase shift of the compensator to be measured as a function of wavelength [10]. Subsequently, data were obtained with the sample in place at an angle of incidence equal to 80°. Finally, Δ and Ψ for the sample were calculated from:

$$\tan\Psi e^{i\Delta} = \tan\Psi_{cs} e^{i\Delta_{cs}} \Big/ \tan\Psi_c e^{i\Delta_c}, \quad (2)$$

where the subscript c refers to the straight through measurement and cs refers to the measurement with the sample in place. SE data from 50 mechanical cycles of the analyzer were averaged for each wavelength. The entire calibration procedure for the compensator requires 5-8 minutes.

As has been described previously [11] the ellipsometer was also calibrated to account for the dark current present during the measurement, and for variations in the ac to dc gain ratio with decreasing light intensity. The resulting system has an estimated accuracy of ±0.001 for the refractive index and the extinction coefficient for even such poor reflectors as vitreous silica.

C. Data Reduction

Once the ellipsometric spectra were determined, a computer program was used to develop a depth profile of the sample and to determine the optical properties. The sample was modeled as a layer of surface roughness on top of an infinite layer of bulk substrate material. A schematic of this geometry is shown in fig. 2. The reference data for the $BaZrO_3$ was obtained from a thin film sample of $BaZrO_3$ on YSZ. In this case, the model consisted of a layer of BZO surface roughness on a layer of dense BZO on the YSZ substrate. The surface roughness was modeled as a mixture of air and substrate material using the Bruggeman effective medium approximation. The refractive index as a function of wavelength ($n(\lambda)$) was modeled as a Sellmeier oscillator function, given by:

$$n^2 = 1.0 + \left(A\lambda \Big/ \left(\lambda^2 - \lambda_o^2\right) \right), \quad (3)$$

where n is the index of refraction, λ is the wavelength in nm and A and λ_o are the variables in the model which are related to the optical properties of the material. For samples which exhibited absorption at higher energies, the data set was truncated so that the undamped Sellmeier oscillator would still be a valid description of the data. The only additional variable was the thickness of the surface layer. The volume fractions in the surface layer were fixed to 50% in the model because the layer thickness on these polished substrates is very low. Letting the volume fraction vary resulted in excessive correlation between variables [10].

The input to the modeling program consisted of a range of values for each variable. Then, the program calculated Δ and Ψ spectra for each combination of variables and compared them to the experimental values. After the initial search determined acceptable starting parameters, a Levenberg-Marquardt algorithm was used to determine the best fit values, 90% confidence limits and the correlation between the variables. The calculated data were compared to the experimental using the unbiased estimator of the error, σ, defined as:

$$\sigma = \frac{1}{N-P-1} \sum_{i=1}^{N} \left[\left(\Delta^i_{exp} - \Delta^i_{calc}\right)^2 + \left(\Psi^i_{exp} - \Psi^i_{calc}\right)^2 \right]^{1/2}, \quad (4)$$

where N is the number of data points (typically 202), P is the number of variables in the model (3) and the subscripts exp and calc refer to the experimental and calculated parameters. This weighting function has been shown to be the most

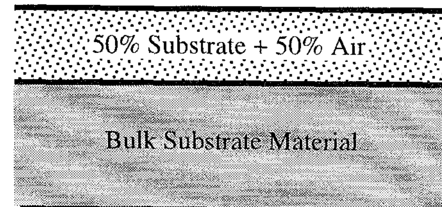

Fig. 2. Schematic model for substrate used in SE data analysis.

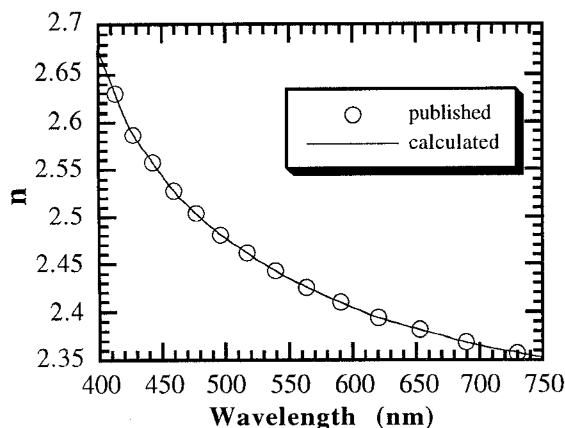

Fig. 3. Comparison of published dielectric function data to SE determined data for $SrTiO_3$.

appropriate for analysis of transparent samples where the values of Δ are low [12].

With the depth profile and the Sellmeier constants of the material determined, it was possible to calculate the dielectric function ($\varepsilon_1+i\varepsilon_2$) for this wavelength range. It is important to emphasize here that these data were obtained while taking into account the surface roughness. In other methods where this inhomogeneity is not accounted for, the calculated data can be misleading. Specifically, anomalous values for the refractive index (n) and the extinction coefficient (k) can be obtained.

The substrates characterized in this study were: $LaAlO_3$, $NdGaO_3$ and $(LaAlO_3)_{0.3}$–$(Sr_2AlTaO_6)_{0.7}$ (from Applied Technology Enterprises, Irmo, SC); $LaSrGaO_4$ (from A. Dabkowski and H.A. Dabkowska, McMaster University); $BaZrO_3$ (from Chang-Beom Eom, Duke University); and 9.5 mol% Y_2O_3-ZrO_2 (from Commercial Crystal Labs, Inc., Naples, FL).

III. Results and Discussion

The accuracy of the dielectric function data obtained by this method was checked by calculating data for a $SrTiO_3$ substrate

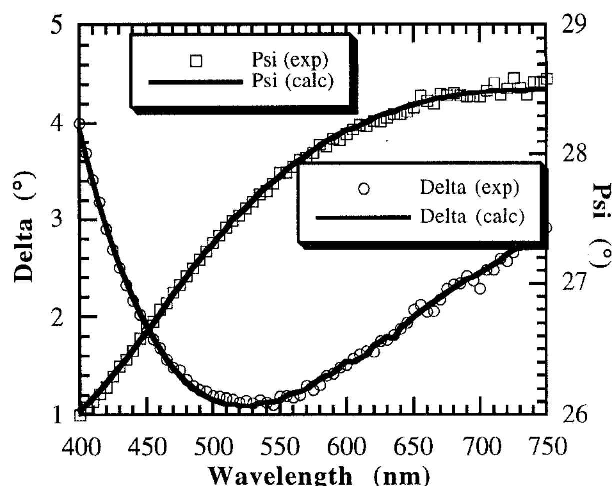

Fig. 5. Experimental and calculated Δ and Ψ data for $BaZrO_3$.

and comparing it to published data obtained by the angle of minimum deviation method [13]. Fig. 3 shows the excellent agreement between the two sets of data. The values agree throughout this range to ±0.001. Values below 400 nm cannot be directly compared because the available published data is obtained from measurements which do not account for the surface roughness.

Fig. 4 shows the experimental and calculated Δ and Ψ spectra for 9.5 mol% Y_2O_3-ZrO_2 (YSZ). The difference between the two spectra is on the order of the accuracy of the instrument (±0.01° for Ψ and ±0.03° for Δ). The thickness of the surface roughness layer was determined to be 36 Å ± 0.2 Å.

Fig. 5 shows a similar plot of the measured and calculated ellipsometric parameters for $BaZrO_3$ (BZO). Again, it is clear that the fit is very good. For this material, the data could only be modeled as a Sellmeier oscillator over the range of 400 nm - 750 nm. At lower wavelengths the BZO begins to absorb the incident light. Thus, the assumption of a classical oscillator begins to break down as described previously. For the lower wavelength range, the experimental data were directly inverted to obtain the complex dielectric function.

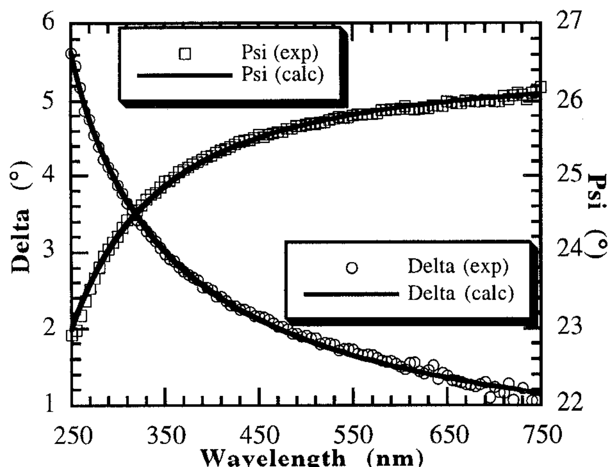

Fig. 4. Experimental and calculated Δ and Ψ data for 9.5 mol% Y_2O_3-ZrO_2.

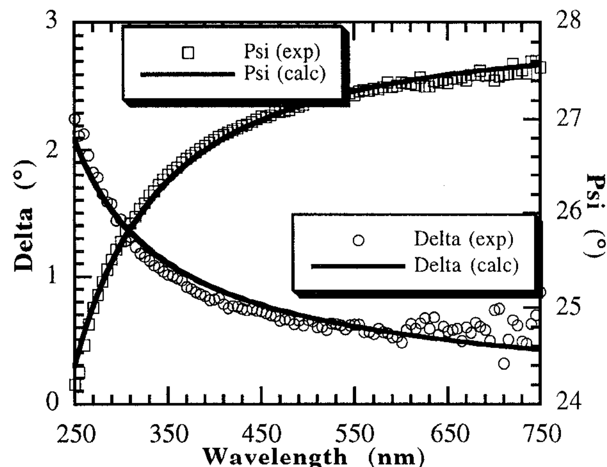

Fig. 6 Experimental and calculated Δ and Ψ data for LSAT.

Fig.6 is a plot showing the fit to the experimental data for $(LaAlO_3)_{0.3}$—$(Sr_2AlTaO_6)_{0.7}$ (LSAT). The quality of the model is shown again by the excellent agreement between the measured and calculated spectra. The scatter in the data at longer wavelengths is due to the very low intensity of the reflected light. The roughness layer determined here is 16 Å ± 0.3 Å. LSAT is a relatively new substrate material being studied for epitaxial growth of YBCO. The advantage of LSAT over pure $LaAlO_3$ is that there is no twin structure present which can disturb the crystallinity of the YBCO upon cooling after growth.

Table I lists the results of the SE modeling for each substrate material. In addition to the materials listed above, data for $LaAlO_3$ (LAO), $NdGaO_3$ (NGO) and $LaSrGaO_4$ (LSGO) were determined. The constants A and λ_o from (3) are given along with the values of the unbiased error estimator, σ. The value of λ_o is related to the natural frequency of the oscillator. A is related to the magnitude of the dielectric function over this particular wavelength range. The models shown in Fig. 4 - Fig. 6 are indicative of the quality of all the fits.

Finally, Fig. 7 is a plot of the real part of the dielectric function (ε_1) for all of the materials in this study. Each curve was derived using (3) and the Sellmeier constants determined from the SE data. The relationships between A and λ_o and the shape of the curve can be clearly visualized with this plot. As A increases, the height of the curve follows. And as λ_o increases, the increase in the slope of the curve is sharper at the lower wavelengths.

TABLE I
VALUES FOR THE CONSTANTS OF THE SELLMEIER OSCILLATOR DETERMINED FROM MODELING

Material	*A*	λ_o *(nm)*	σ
$LaAlO_3$	3.09 ± 0.01	125.5 ± 2.2	0.12°
$BaZrO_3$	3.04 ± 0.01	159 ± 6	0.06°
$NdGaO_3$	3.26 ± 0.01	140.2 ± 1.3	0.11°
YSZ	3.50 ± 0.01	143.9 ± 0.8	0.07°
LSAT	2.92 ± 0.01	144.9 ± 1.2	0.08°
LSGO	2.88 ± 0.01	140.6 ± 1.4	0.10°

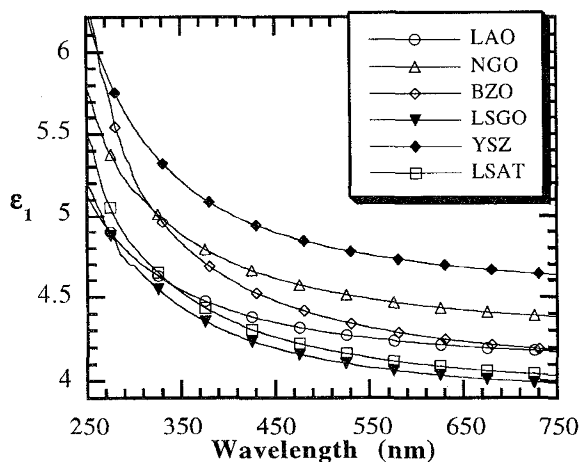

Fig. 7. Plot of the real part of the dielectric function for several YBCO substrate materials as determined by SE.

IV. CONCLUSION

In order to use optical methods (i.e. spectroscopic ellipsometry) to characterize the microstructures of YBCO films on various substrates, it is necessary to have reference dielectric function data for these materials. There exists several sources of data for YBCO, however data for many of the substrate materials are not available. In this study, dielectric spectra for $LaAlO_3$, $NdGaO_3$, $BaZrO_3$, $LaSrGaO_4$, 9.5 mol% Y_2O_3—ZrO_2 and $(LaAlO_3)_{0.3}$—$(Sr_2AlTaO_6)_{0.7}$ have been determined by SE. The data was shown to be accurate to the third decimal point. In determining the data, the surface roughness layer was accounted for.

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