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A Statistical Analysis of Laser Ablated $Ba_{0.50}Sr_{0.50}TiO_3$ /$LaAlO_3$ Films for Microwave Applications

R.R. Romanofsky*, N.C. Varaljay*, S.A. Alterovitz*, F.A. Miranda*, C.M. Mueller**,F.W. VanKeuls***, J. Kim****, and K.S. Harshavardhan****
*NASA Glenn Research Center, Cleveland, OH 44135
**Hadron, Inc. Cleveland, OH 44135
***Ohio Aerospace Institute, Brookpark, OH 44142
****Neocera, Inc., Beltsville, MD 20705

ABSTRACT

The NASA Glenn Research Center is constructing a 616 element scanning phased array antenna using thin film $Ba_xSr_{1-x}TiO_3$ based phase shifters. A critical milestone is the production of 616 identical phase shifters at 19 GHz with ≈4 dB insertion loss and at least 337.5° phase shift with 3 percent bandwidth. It is well known that there is a direct relationship between dielectric tuning and loss due to the Kramers-Kronig relationship and that film crystallinity and strain, affected by the substrate template, play an important role. $Ba_{0.50}Sr_{0.50}TiO_3$ films, nominally 400 nm thick, were deposited on 48 0.25 mm thick, 5 cm diameter $LaAlO_3$ wafers. Although previous results suggested that Mn-doped films on MgO were intrinsically superior in terms of phase shift per unit loss, for this application phase shift per unit length was more important. The composition was selected as a compromise between tuning and loss for room temperature operation (e.g. crystallinity progressively degrades for Ba concentrations in excess of 30 percent). As a prelude to fabricating the array, it was necessary to process, screen, and inventory a large number of samples. Variable angle ellipsometry was used to characterize refractive index and film thickness across each wafer. Microstructural properties of the thin films were characterized using high resolution X-ray diffractometry. Finally, prototype phase shifters and resonators were patterned on each wafer and RF probed to measure tuning as a function of dc bias voltage as well as peak (0 field) permittivity and unloaded Q. The relationship among film quality and uniformity and performance is analyzed. This work presents the first statistically relevant study of film quality and microwave performance and represents a milestone towards commercialization of thin ferroelectric films for microwave applications.

INTRODUCTION

Scanning phased array antennas could offer a highly desirable solution for futuristic near Earth and deep space science mission scenarios. For example the Laser Interferometer Space Antenna will consist of three spacecraft flying 5 million km apart in the shape of an equilateral triangle. The formation flying spacecraft will form a giant Michelson interferometer, measuring the distortion of space caused by passing gravitational waves. The antenna needs to measure the distance between proof masses separated by 5 million km with an accuracy of 20 picometers. Hence mechanically induced vibrations from gimbaled parabolic communications system antennas are unacceptable. While other solutions exist an affordable and efficient phased array would be preferred. Numerous applications for phased arrays exist for low Earth orbiting (LEO) scientific and commercial spacecraft. And a suitable scanning phased array technology is also being sought for high data rate LEO-to-ground communications, automotive radar, and other remotes sensing and industrial applications.

The current state-of-practice in scanning phased arrays is represented by GaAs monolithic microwave integrated circuit (MMIC) technology or ferrite phase shifters[1,2]. Cost and weight are still significant problems. Moreover, conventional manifold fed arrays suffer from beam-forming loss that places considerable burden on MMIC amplifiers. The inefficiency can result in severe thermal management problems. A ferroelectric reflectarray antenna that overcomes these limitations is being assembled at the NASA Glenn Research Center [3]. The enabling component of the array is a low loss thin film $Ba_{0.50}Sr_{0.50}TiO_3$ coupled microstrip phase shifter. While we have demonstrated good performance from these devices (≈60°/dB) it was not known if a sufficient quantity of high quality and uniform films could be produced economically. And, the correlation among film thickness, crystallinity and microwave performance was still under investigation.

In this work we examine a batch of 48 $Ba_{0.50}Sr_{0.50}TiO_3$ on $LaAlO_3$ wafers in terms of film uniformity, microstructure and microwave performance. Variable angle ellipsometry was used to characterize refractive index and film thickness across each wafer. Microstructural properties of the thin films were characterized using high resolution X-ray diffractometry. Finally, prototype phase shifters and resonators were patterned on each wafer and RF probed to measure tuning as a function of dc bias voltage as well as peak (0 field) permittivity and unloaded Q

FILM PREPARATION

A set of film specifications was targeted based on our prior experience with various devices and material from different vendors. All $(Ba_{0.60}Sr_{0.40})TiO_3$ films were to be deposited on 5 cm diameter, 0.25 mm thick single-crystal $LaAlO_3$ substrates. The nominal film uniformity sought was a film thickness of 400 nm ± 40 nm at any point on the wafer except the 2 mm diameter ring at the edge of the wafer. The desired crystalline quality was a full-width, half-maximum value of the (002) peak < 0.12° (432 arc seconds) and the full-width, half-maximum values of the (103) peak <0.20° (720 arc seconds) with a concomitant peak intensity (002) > 4500 counts per second. The expected lattice parameters were a cubic unit cell, where the difference between $a_{<100>}$ and $a_{<001>}$ was less than 0.01 Å. Ideally, both $a_{<100>}$ and $a_{<001>}$ would not deviate by more than 0.03 Å from the bulk lattice parameters for $Ba_{0.5}Sr_{0.5}TiO_3$. Admittedly, these were ambitious goals

The films were deposited at Neocera, Inc. by Pulsed Laser Deposition (PLD). A Lambda-Physik Kr-F excimer laser operating at 248 nm was used. The laser energy density at the target during film deposition was $3 J/cm^2$. The deposition temperature was 800 C. The oxygen partial pressure during film deposition was 300 mTorr. After film deposition the chamber was back filled with 500 Torr of oxygen and the substrates were cooled to room temperature. To obtain uniform film thickness the substrates were mounted on a rotating substrate stage. This special substrate holder facilitates the substrate rotation about their axes as well as about the axis of the stage, executing a planetary rotation. To maximize the thickness uniformity the laser beam is also raster scanned over the target surface using a laser beam scanner. The beam scanner is computer controlled to facilitate a uniform deposition rate over the substrate area. The films were delivered in two batches of 24 each.

ELLIPSOMETRIC ANALYSIS

Experimental Procedure

Ellipsometric measurements were taken on a "J. A. Woollam Co., Inc." spectroscopic ellipsometer model M-2000L at an angle of incidence of 65º at 800 wavelengths in the range 2450-9000Å. Each wafer was measured in the center, 0.5" and 0.75" off center. In some cases, several measurements were done at almost the same spot, to obtain a lower "depolarization" factor. In this study an unusual problem was encountered and it was due to the twins in the $LaAlO_3$ substrates. These twins were scattering part of the incoming light in a non-specular way, thus breaking a main ellipsometry assumption. The fraction of the light which was lost is a measurable quantity and is called the "depolarization" factor. In the present study, non-zero "depolarization" factors were measured for all samples which affected Ψ and Δ experimental results. This could cause increased values for mean square error (MSE) and errors in the value of the sample parameters. A detailed explanation follows.

An example of a large "depolarization" factor experiment is shown in Fig. 1. Adjacent spots on wafer #37, 0.5" off center were measured at an angle of incidence of 65º and gave the "depolarization" factors shown in the figure. The large "depolarization" measurement probably was done with the light being reflected from a spot that included a large twin boundary. Fig. 2 depicts the experimental ψ vs. λ graph showing a large effect of the "depolarization" on the ψ (and Δ) results. As a result of this, the final values obtained in the least squares fit for the sample parameters were also affected, especially keeping in mind the parameters correlations problem (see below). For example, a layer thickness difference of 200Å (out of 3600Å) between the two spots was obtained in the fits, even though such a difference is essentially impossible.

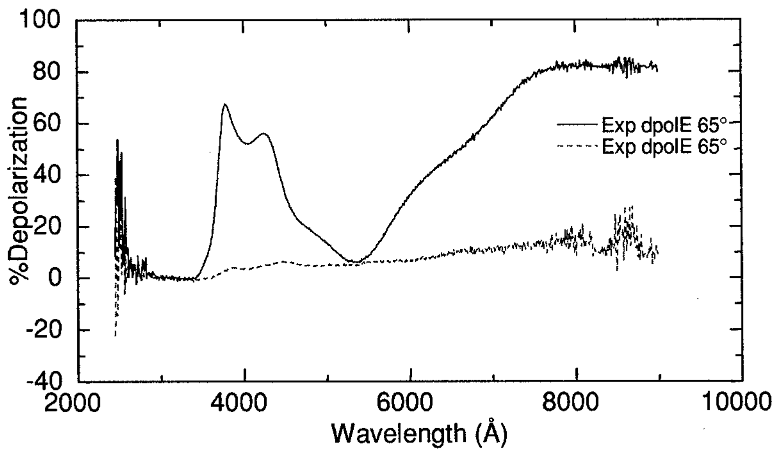

Fig. 1. Experimental "depolarization" factor vs. the wavelength λ for two adjacent spots on wafer #37, 0.5" off center. An ideal sample has a vanishing "depolarization" factor.

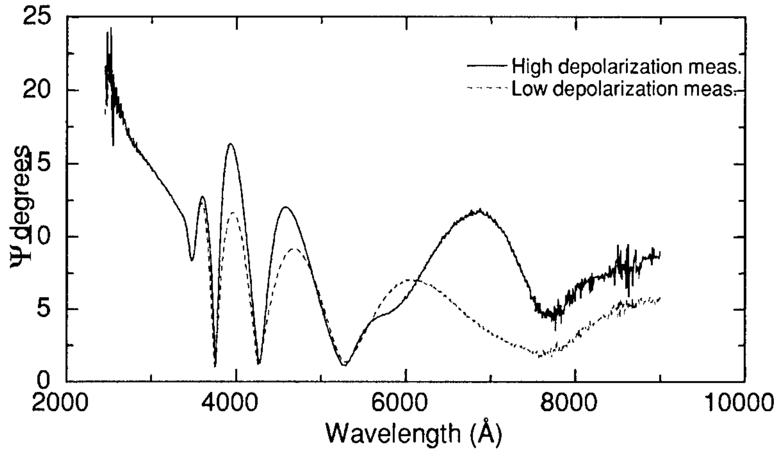

Fig. 2. Experimental ψ vs. the wavelength λ for two adjacent spots showing the "depolarization" effects on other ellipsometric parameters. (Wafer #37, 0.5" off center)

An empirical rule was found which states that least squares fits with 4 variables having MSE below 25 were considered to have good confidence, while fits with MSE above 40 were considered unreliable. However, no exact estimate of the errors was possible. In the case of wafer #37, 0.5" off center, the MSE values were 11 and 76 for the low and high "depolarization" measurements respectively. The substrate dielectric function was measured on a blank $LaAlO_3$ wafer. A "standard" $Ba_xSr_{1-x}TiO_3$ dielectric function was also measured and presented at the Spring 2000 MRS [5]. The mixtures are all calculated using the EMA (Bruggeman Effective Medium Approximation) method. The variables in the least squares fit are: film and surface overlayer thicknesses and void fractions in the film itself and in the overlayer. The λ range of the fits was over the entire 245-900 nm experimental wavelength range. The model includes the $LaAlO_3$ substrate, the ferroeletcirc film and a top roughness layer. The film is assumed to be made of good quality $Ba_xSr_{1-x}TiO_3$, which is simulated as a mixture of $Ba_xSr_{1-x}TiO_3$ similar to that made by NREL using PLD on MgO and voids [4]. The larger the void fraction, the lower is the refractive index of the present film as compared to the NREL film. Lower refractive index is associated with lower quality material when comparing films in the tables. The roughness layer is again a mixture, this time of the film material and variable void fraction. In some cases, a constant value of 18% voids for the overlayer was chosen, as it does fit most of the films, and makes the comparison among wafers more standard.

Results

The full result for wafer #37, 0.5" off center (as an example) was:MSE=11.29, Layer #1 thickness 3616.8 Å, Layer #1 composition 5.05 % void by volume, Layer #2 (overlayer) thickness 106.4 Å, Layer #2 (overlayer) composition 16.8 % void by volume. The quality of the fits obtained after the least squares minimization can be seen in Fig. 3.

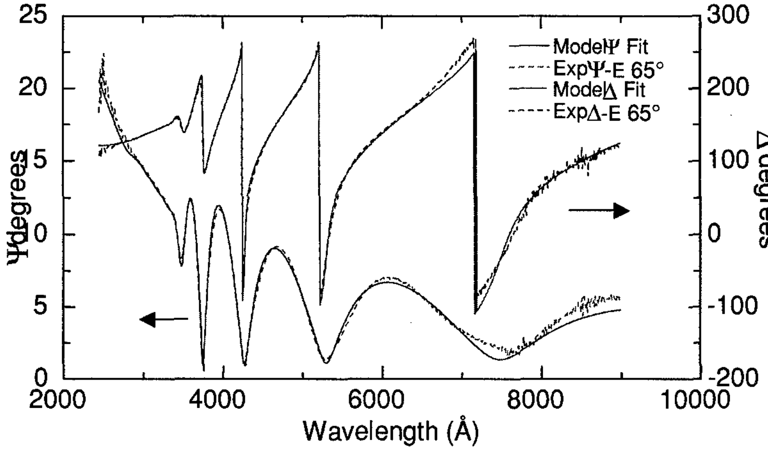

Fig. 3. Comparison of the experimental and the model calculated ψ and Δ vs. the wavelength λ, for the low polarization spot. (wafer # 37, 0.5" off center)

For comparison, the high "depolarization" spot in the same general area gave the following results: MSE=76.46, Layer #1 thickness 3422 Å, Layer #1 composition -2.42 % void by volume, Layer #2 (overlayer) thickness 138.1 Å, Layer #2 (overlayer) composition 18.3 % void by volume. The result above shows that the sample parameters are quite different from the low "depolarization" spot. The quality of the fits obtained after the least squares minimization in this case can be seen in Fig. 4, and it shows a much poorer fit than in Fig. 3.

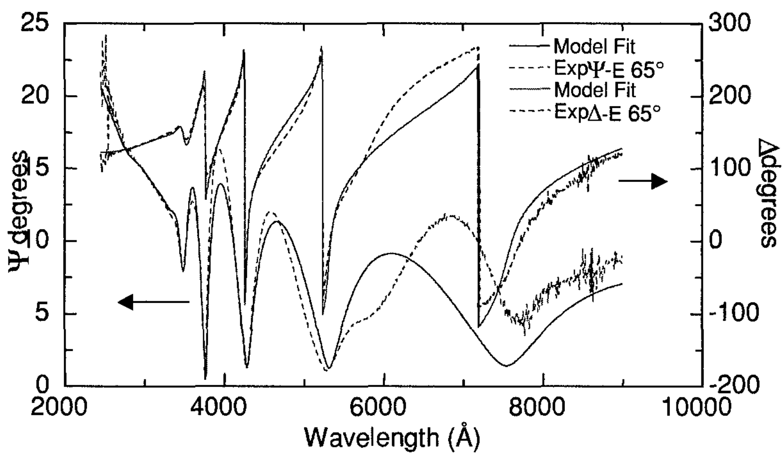

Fig. 4. Comparison of the experimental and the model calculated ψ and Δ vs. the wavelength λ, for the high polarization spot. (wafer # 37, 0.5" off center)

Another important consideration for any least squares fit is the resulting parameter correlations. The correlations, which were rather high among all 4 parameters in these measurements, mean that any value obtained for a single parameter out of the 4 parameters is not as reliable as needed for accurate comparisons. This correlation problem was in addition to the fact that different "depolarization" factors were measured on different wafers. These comparisons among wafers required very accurate values for each parameter, with accuracy much higher than the differences among the wafers. It was found that a one-parameter description of the films, although not as good mathematically as a full 4 parameters model, was a reasonably good description of the film for wafer comparison purposes. This one parameter model assumes that the film is a uniform NREL BSTO quality material, and the thickness is the only variable. This thickness is called the "equivalent" thickness and is conceptually connected with the total amount of $Ba_xSr_{1-x}TiO_3$ material in the film having the same high density as the NREL material.

The "equivalent" thickness values obtained in these simplified fits are similar (to within ~2%) to the values calculated using the film thickness multiplied by density and added to the same product for the overlayer. Here density means (1-void fraction) and is really the density normalized to the NREL $Ba_xSr_{1-x}TiO_3$ density. The quality of the fits as measured by the MSE is much poorer than for the 4 parameter fits, as the actual films do have an overlayer and have a lower density than the NREL films.

An example of a result for wafer #37, 0.5" off center, low depolarization spot: MSE=63.69, "Equivalent" film thickness 3573 Å. Result for wafer #37, 0.5" off center, high depolarization spot: MSE=104.1,"Equivalent" film thickness 3586.5 Å. This result shows the small sensitivity of the one-parameter model to the "depolarization" problem (figure 5). Numerical results for all samples are shown in figure 6.

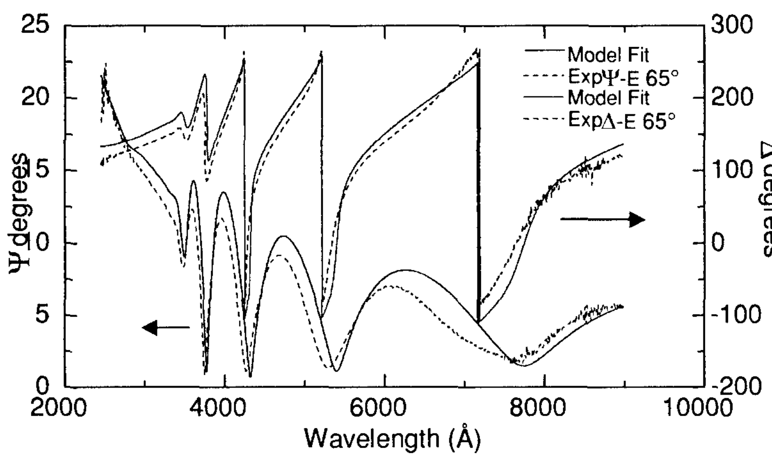

Fig. 5. Comparison of the experimental and the model calculated ψ and Δ vs. the wavelength λ, low polarization spot, one parameter model. (wafer # 37, 0.5 " off center)

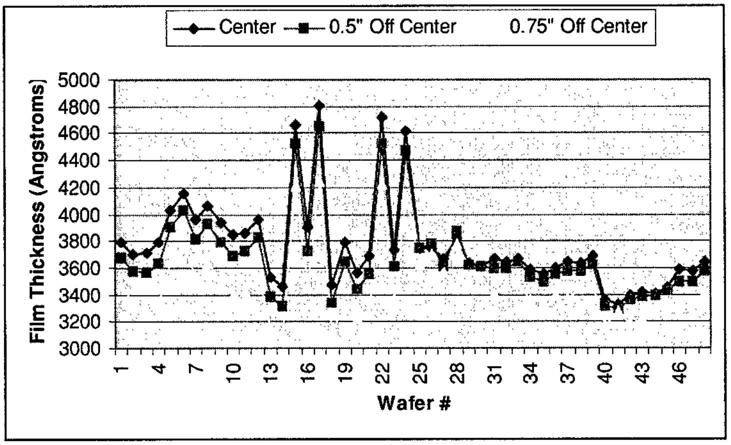

Fig 6. Summary of the film thickness variation within and across the 48 wafers.

The mean and (standard deviation) for the film thickness at the center, 0.5" and 0.75" off center are 3767 Å (340 Å), 3677 Å (313 Å), and 3469 Å (273 Å), respectively. Results have the following general features, as derived mostly from the "equivalent" thickness results:

1. The variation in film thickness among wafers is much larger in the group of wafers #1-24 vs. the group of wafers #25-48.
2. In the wafers group #1-24, the film thickness drops by about 150Å from the center to the 0.5" off-center point, and another ~200Å to the 0.75" off-center location. This is way above the requirement of ±4% around the thickness at the center. In the wafers group #25-48, there is only a negligible drop between the center and 0.5" off center and around 100Å to the 0.75" location.
3. The film thickness for the wafer group #1-24 is in the range 4000±800Å, with 8 wafers having the center location outside the required range of 3600-4400Å (4 are too thin and 4 are too thick). In the wafer group #25-48 the thickness range is 3550±350Å with 11 samples having the center location thickness lower than the required range of 3600-4400Å. These 11 wafers are all in the last group of wafers #34-48.
4. An estimate of the error in the refractive index as measured by the void fraction is in the range 1-2%. Using this error estimate, there is only a negligible difference in the quality of the film when we move from the center toward the periphery.

HIGH RESOLUTION X-RAY DIFFRACTION

The crystalline quality of the film was evaluated using a Phillips PW3720 HRXRD in the double-axis mode. The measurements were made at five different locations, at points lying successively further from the center of the wafer: (0,0 mm); (5,5 mm); (10, 10 mm); (15, 15 mm); and (18, 18 mm). Data for the center (0,0 mm) and practical usable limit insofar as device placement is concerned (15, 15 mm) is shown in figures 7 and 8.

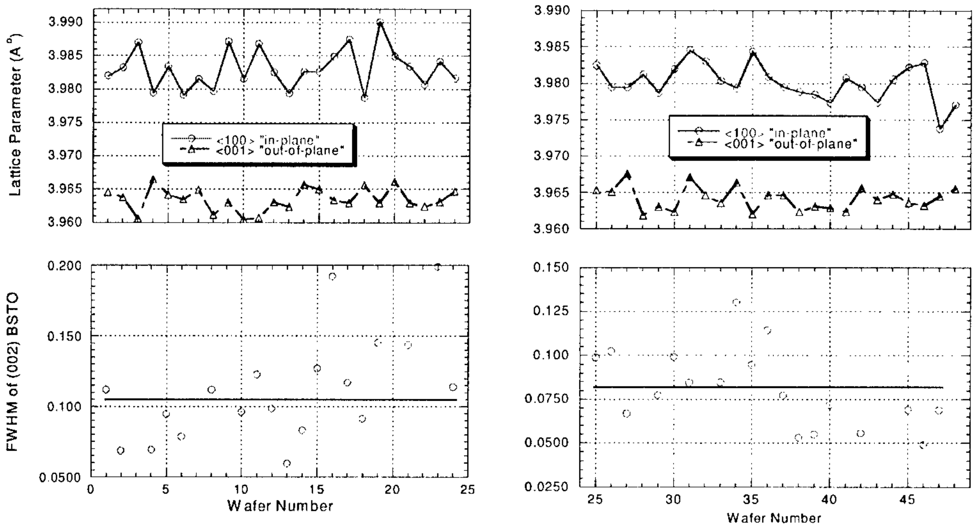

Fig 7. Lattice parameters and full width half maximum values for the 48 $Ba_{0.50}Sr_{0.50}TiO_3/LaAlO_3$ wafers at the center of each wafer (0,0 mm).

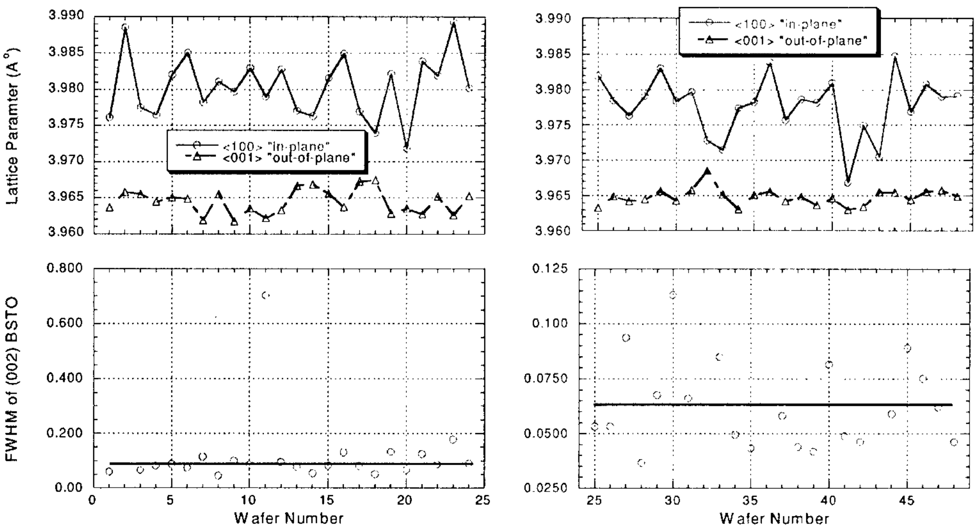

Fig 8. Lattice parameters and full width half maximum values for the 48 $Ba_{0.50}Sr_{0.50}TiO_3/LaAlO_3$ wafers near the perimeter of each wafer (15,15 mm).

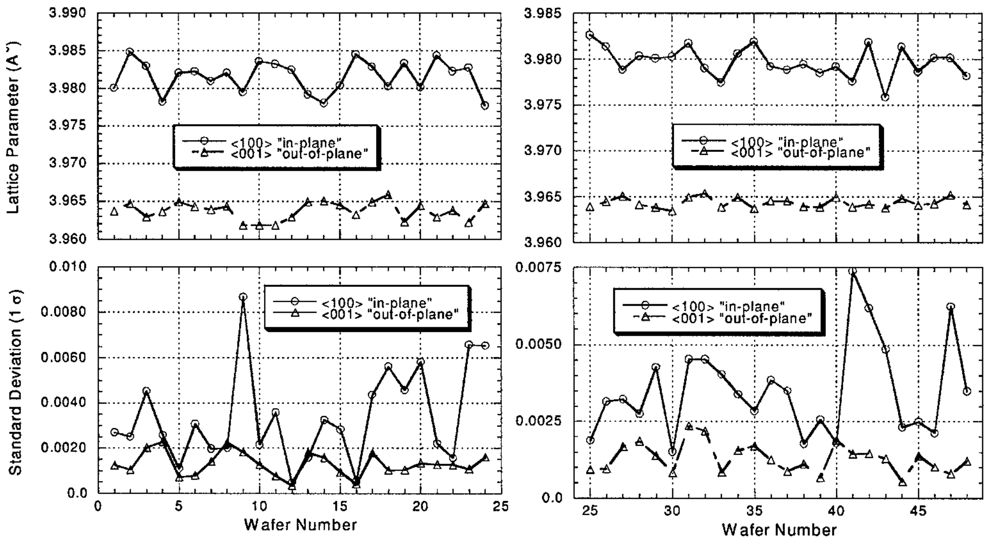

Fig. 9 Average of the lattice parameters and standard deviation across each wafer (all 5 spots). For wafers 1-24, the mean in-plane lattice parameter was 3.9816Å (σ=0.0021) and the mean out-of-plane lattice parameter was 3.9637 Å (σ=0.0012). For wafers 25-48 the mean in-plane lattice parameter was 3.9797Å (σ=0.0016) and the mean out-of-plane lattice parameter was 3.9643 Å (σ=0.0005).

Very good reproducibility was obtained both across the wafers and from wafer to wafer. Only wafer 11 was considerably poorer that the others based on the increase in FWHM. The film quality and reproducibility of the second set of wafers (25-48) was slightly better that the original set (1-24). The difference between in-plane and out-of-plane lattice parameters was approximately 0.015 Å. It would have been preferred to have the in-plane lattice parameters closer to 3.965 rather than 3.980 (so as to match the out-of-plane parameter).

MICROWAVE MEASUREMENTS

Several microwave test structures were patterned near the perimeter of each of the 48 wafers (approximately 0.75" from center) using standard lift-off techniques. The nominal metalization consisted of 250 Å Ti, 500 Å Au, 1.5 μm Ag, a 2500 Å Au cap. The test structures included λ/2 and λ ≈42 Ohm gap coupled microstrip resonators and a single coupled microstrip phase shifter element. The resonator coupling gap, that serves to isolate the resonator and facilitate extraction of unloaded Q, was 12.5 μm wide. The phase shifter section consisted of coupled microstrip lines that were 425 μm long separated by a 12 μm gap. Each device incorporated a coplanar virtual ground structure to allow on-wafer testing with 250 μm pitch ground-signal-ground probes. This technique has been described elsewhere [5]. The probes were calibrated to the device input/output ports. S11 and S21 were measured for the resonators and phase shifters, respectively, using a HP 8510C automatic network analyzer. The effective permittivity was determinedd from the λ/2 and λ resonator measurements using well-known techniques [6,7] and peak (0 dc field) permittivity of the

$Ba_{0.50}Sr_{0.50}TiO_3$ layer was deduced a quasi-TEM model [8]. The unloaded Q (Q_o) was determined from the λ/2 resonator. The resonant frequencies of the λ/2 and λ resonators were typically ≈17.5 and 18.5 GHz, respectively. The insertion phase shift was measured at 19 GHz by applying 0 and 200 V to the device. This step was conducted in vacuum (≈100 mTorr) to help prevent any breakdown problems across the coupled lines. Figure 10 summarizes the phase shift and Q_o for all available wafers.

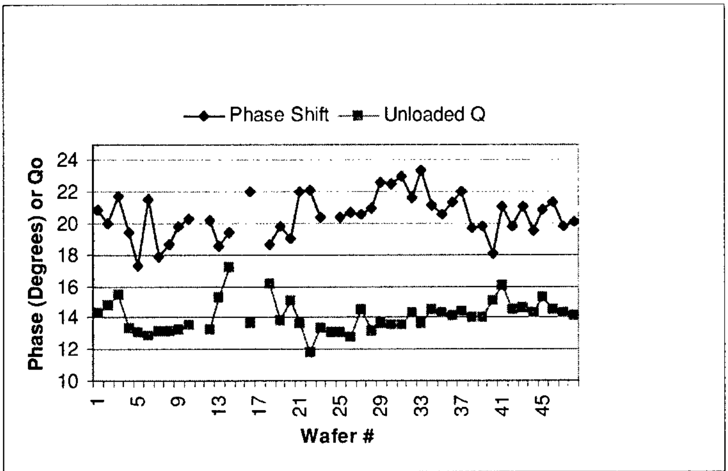

Fig 10. Insertion phase shift at 19.0 GHz for a single 425 micron long coupled line between 0 and 200 V and unbiased, unloaded Q (Q_o) near 17.5 GHz for a 42 Ohm microstrip resonator.

Wafers 15 and 17 were sacrificed earlier for device optimization purposes and were not included in this part of the study. The λ resonator on device 11 had a lithographic error but the λ/2 device yielded a Q_o near 16. The probe structure on phase shifter 24 shorted during testing. There were no other problems. The mean Q_o was 14.1 with a standard deviation of 1.0. The mean insertion phase shift was 20.5° with a standard deviation of 1.4°. The insertion loss near the mid-band of the phase shift structure (about 15 GHz), which included the effects of the probe transitions and 0.6 cm of microstrip, typically changed from 2.2 dB to 1.6 dB with a bias of 0 V and 200 V, respectively.

The estimated peak dielectric constant of the $Ba_{0.50}Sr_{0.50}TiO_3$ layer for each wafer is shown in figure 11. The mean peak dielectric constant was 2129 with a standard deviation of 149.

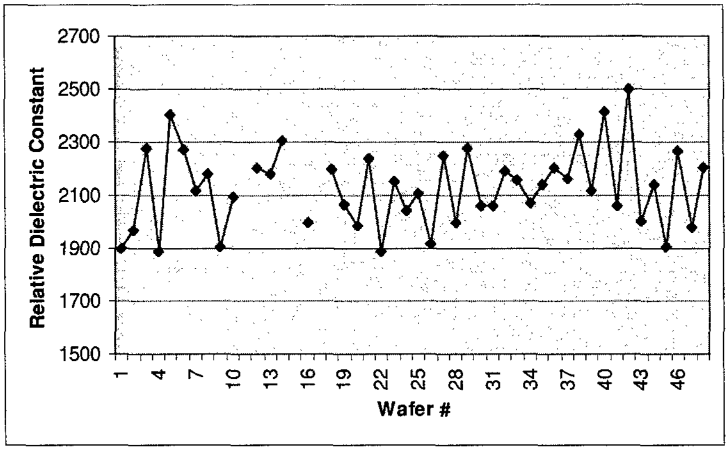

Fig 11. Peak (0 field) dielectric constant of the $Ba_{0.50}Sr_{0.50}TiO_3$ layer extracted from the $\lambda/2$ and λ microstrip resonators.

CONCLUSIONS

In order for any phased array antenna to be realized with ferroelectric materials, or any device technology for that matter, the phase shifters must be reproducible in terms of insertion loss and insertion phase shift. For example, a random 2 dB insertion loss variation may be tolerable (i.e. adequate beam profile maintained) but can degrade the power in the main beam by about 33 %. Similarly, a random phase error of $\pi/16$ may be negligible but a $\pi/8$ random phase error may degrade the beam by about 22%.

Very good reproducibility was obtained both across the 48 wafers and from wafer to wafer in terms of crystal quality. For wafers 1-24, the mean in-plane lattice parameter was 3.9816Å (σ=0.0021) and the mean out-of-plane lattice parameter was 3.9637 Å (σ=0.0012). For wafers 25-48 the mean in-plane lattice parameter was 3.9797Å (σ=0.0016) and the mean out-of-plane lattice parameter was 3.9643 Å (σ=0.0005). The FWHM of the (002) peak was generally better than 0.1° and significantly better than this for the second batch of 24 wafers.

Based on the ellipsometric analysis, The mean and (standard deviation) for the film thickness at the center, 0.5" and 0.75" off center are 3767 Å (340 Å), 3677 Å (313 Å), and 3469 Å (273 Å), respectively. There is a direct effect of the thickness measurements on peak permittivity calculations. The sensitivity of the calculation to measurement uncertainty is nearly linear. For example, a film thickness measurement uncertainty of 10% translates into about a 9% change in peak dielectric constant for the $Ba_{0.50}Sr_{0.50}TiO_3$ layer. We believe that the uncertainty is much smaller than this. Notice that several wafers had significant excursions in thickness from the mean value but their corresponding peak dielectric constant was consistent with the average value.

Despite some variations in thickness and crystallinity, the microwave performance was very consistent. The mean Q_o was 14.1 with a standard deviation of 1.0. The mean insertion phase shift was 20.5° with a standard deviation of 1.4°. The insertion loss near the mid-band of the phase shift structure (about 15 GHz), which included the effects of the probe transitions and 0.6 cm of microstrip, typically changed from 2.2 dB to 1.6 dB with a bias of 0 V and 200 V, respectively. In the actual ferroelectric reflectarray application, the phase shifter will consist of ≈6 coupled line sections like the one used here and be operated in a reflection mode. Thus the insertion phase envelope can be expected to be better than plus or minus 18° or π/10 based on our statistical analysis. The loss variation quoted above represents an extreme case (0 to 200 V) and we expect the average loss variation to be within a plus or minus 1 dB envelope. The Q measurements suggest that the film quality, in terms of loss tangent, is very reproducible.

In summary, we believe that the two batches totaling 48 PLD $Ba_{0.50}Sr_{0.50}TiO_3/LaAlO_3$ wafers are suitable, from a microwave point of view, for fabrication into phase shifters and incorporation into a state-of-the-art phased array antenna. In the near future, the remaining 46 wafers will be patterned with approximately 30 phase shifters (or more) each. Then the phase shifters will be on-wafer sampled (about 10% tested) and inventoried for use in the ferroelectric reflectarray antenna.

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