# SYMPOSIUM KK

# Materials Issues for Tunable RF and Microwave Devices

November 30 – December 2, 1999

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<sup>\*</sup> Invited paper

#### SESSION KK1: FREQUENCY AGILE MATERIALS FOR ELECTRONICS

Chairs: Steven C. Tidrow and Xiaoxing Xi Tuesday Morning, November 30, 1999 Room 201 (H)

#### 8:30 AM \*KK1.1

FREQUENCY AGILE MATERIALS FOR ELECTRONICS (FAME) A DARPA PERSPECTIVE. Stuart Wolf, DARPA/DSO, Arlington

The DOD is very interested in frequency agile electronics for radar, coomunications and intelligence applications. The DARPA FAME program is addressing these problems by developing tunable components and subsystems based on the field dependent properties of para-electrics and ferrites as well as MEMs and other novel ideas. This talk will review some of these concepts as well as provide an update of the progress to date.

#### 9:00 AM \*KK1.2

ANALYSIS AND OPTIMIZATION OF THIN FILM FERRO-ELECTRIC PHASE SHIFTERS. Robert R. Romanofsky, Fred W. Van Keuls, Felix A. Miranda, Joseph D. Warner, Carl H. Mueller, NASA Glenn Research Center, Cleveland, OH; and Haq Qureshi, Cleveland State University, Cleveland, OH.

Microwave time delay phase shifters based on various thin ferroelectric film transmission lines have been developed. The best SrTiO3 and  $Ba_xSr_{1-x}TiO_3$  films thus far have been grown using pulsed laser deposition (PLD). The performance of these devices is better than their semiconductor microwave integrated circuit counterparts despite relatively high film loss tangent. Insertion loss less than 5 dB has been achieved at K-band using a dc field of nearly 40 V/ $\mu m$  to affect 360° tuning. Single strip and coupled strip electrode patterns have been analyzed theoretically and experimentally. Line capacitance of the multi-layer dielectric structure is evaluated using a quasi-TEM variational expression. The potential distribution satisfying Poisson's equation is derived using a transverse transmission line method. With this technique, complemented by full-wave electromagnetic simulation, it is possible to dissect the total loss into its various constituents including: substrate, ferroelectric layer, mismatch, and conductor and radiation loss. It is shown that at the microwave frequencies of interest the conductor loss dominates and the ferroelectric layer loss tangent can deteriorate substantially before becoming a problem. Electrode material and thickness and ferroelectric film thickness impact performance as well. The best results to date and optimal designs will be presented. A new design concept for a scanning reflectarray antenna based on these devices, and the impact of device performance on the antenna, will also be discussed.

#### 9:30 AM \*KK1.3

MATERIALS ISSUES IN Ka-BAND FERROELECTRIC PHASE SHIFTERS. J. Talvacchio, W.D. Partlow, S. Gaglione, S.N. Stitzer, T.M. Fertig and S.H. Talisa, Northrop Grumman Corporation, Baltimore, MD.

We will review the materials requirements for Ka-band ferroelectric phase shifters and compare them with materials available today. Trade-offs between tunability and dielectric loss and between operating voltage and conductor losses were evaluated. The specifications that were developed determined specific configurations for prototype phase shifters and a related configuration for evaluating ferroelectric materials at these frequencies. One of the fundamental results is that bulk ferroelectric materials are favored over thin films for this application. Another is that filled or partially filled waveguide designs are favored over microstrip or coplanar transmission lines. \* Supported by DARPA agreement MDA972-99-3-0005

### 10:30 AM \*KK1.4

30 GHz STEERABLE ANTENNAS USING BaxSr<sub>1</sub>\_xTiO<sub>3</sub>-BASED 30 GHZ STEERABLE ANTENNAS USING Bazsr<sub>1</sub>\_xtro<sub>3</sub>-BASED ROOM-TEMPERATURE PHASE SHIFTERS. A. Kozyrev<sup>1</sup>, V. Osadchy<sup>1</sup>, A. Pavlov<sup>1</sup>, A. Golovkov<sup>1</sup>, M. Sugak<sup>1</sup>, D. Kalinikos<sup>1</sup>, C.M. Carlson<sup>2</sup>, T.V. Rivkin<sup>3</sup>, P.A. Parilla<sup>3</sup>, J.D. Perkins<sup>3</sup>, D.S. Ginley<sup>3</sup>, J.C. Price<sup>2</sup>, L.C. Sengupta<sup>4</sup>, L. Chiu<sup>4</sup>, X. Zhang<sup>4</sup>, Y. Zhu<sup>4</sup>, S. Stowell<sup>4</sup>, S. Sengupta<sup>4</sup>; <sup>1</sup>Electrotechnical University of St. Petersburg, St. Petersburg, RUSSIA, <sup>2</sup>University of Colorado, Boulder, CO, <sup>3</sup>National Renewable Energy Laboratory, Golden, CO, <sup>4</sup>Paratek, Inc. Aberdeen MD. <sup>4</sup>Paratek, Inc., Aberdeen, MD.

We report the performance of 4- and 16-element phased array antennas operating at 30 GHz and ambient temperatures. These antennas use BaxSr<sub>1-</sub>xTiO<sub>3</sub>(BST)-based phase shifters to produce the beam steering. Ferroelectric phase shifters offer advantages over more traditional semiconducting devices including lower losses at this and higher frequencies, faster switching speeds, and higher power handling. Since the higher power handling eliminates the need for amplifiers and duplexers at each radiating element, costs are also

reduced. We made phase shifters from laser-ablated epitaxial BST thin films as well as polycrystalline BST-oxide composite thick films. Typical BST thin film phase shifters give almost  $200^\circ$  of phase shift with only 200 V of dc bias. The extrapolated figure of merit is 70°/dB, which is close to the record performance previously reported for superconducting circuit elements. The radiation pattern of the antennas is highly directional with half-power widths as low as  $\sim 13^{\circ}$ and the intensity of the side lobes down by more than 10 dB. With the BST phase shifters incorporated, the central maximum in the radiation pattern can be shifted (or steered) by up to  $\sim 24^{\circ}$  in either direction. These results demonstrate a first step toward prototype steerable antennas for 20-30 GHz satellite communications as well as 70 GHz vehicle radar systems.

#### 11:00 AM \*KK1.5

ELECTRODYNAMIC PROPERTIES OF SINGLE-CRYSTAL AND THIN-FILM STRONTIUM TITANATE . Alp T. Findikoglu, Los Alamos National Laboratory, Los Alamos, NM.

We have pursued a comparative study of broadband (0 - 2 GHz) electrodynamic properties of coplanar waveguides made from single-crystal and thin-film strontium titanate (STO). As a function of temperature, frequency, and dc bias, the single-crystal and thin-film waveguides exhibit marked differences in their electrodynamic properties in terms of nonlinearity, dissipation and dispersion. Such differences also manifest themselves in the performance of prototype nonlinear devices both in the small- and large-signal regimes. At present, we are investigating the relationship between the materials properties and electrodynamic characteristics.

11:30 AM  $\underline{KK1.6}$  FERROELECTRIC MATERIAL FIGURE OF MERIT DEFINED BY DEVICE REQUIREMENTS. E.D. Adler, S.C. Tidrow, A. Tauber, D.D. Brickerd, A. Lee, B. Rod, M.S. Patterson, U.S. Army Research Laboratory, Adelphi, MD; \*Under contract with Geo-Centers, Inc.

Ferroelectrics are tunable electronic materials that as a technology may have a tremendous impact on future radar and communications systems. These materials when incorporated into devices need to operate over very wide bandwidths, handle power requirements without breakdown, and exhibit minimal losses. RF devices include but are not limited to tunable radiating structures, phase shifters, and band select filters. The challenge of developing ferroelectric-based RF devices requires that the materials exhibit wide tuning and low loss characteristics. Unfortunately, as presently defined, the figure of merit of the material does not correspond to the figure of merit for device performance. This can result in development of new materials that have an improved material figure of merit but that result in poorer device performance. Therefore, we need to establish a material's figure of merit in the context with device requirements. For a good device, we tend to want a highly tunable material that is low in loss per device length. We also want the material's  $\epsilon$  to be comparable to the substrate external to the device for minimal impedance mismatch. Finally, we want the material to be fairly consistent over a broadband of frequencies allowing us to approach true-time delay performance. In this paper, we provide a baseline definition for the material figure of merit for a simple microstrip design. We discuss projected device performance based on electromagnetic simulations using the tunability, dielectric constant and dielectric loss tangent for a host of ferroelectric materials.

#### 11:45 AM KK1.7

FIRST DEMONSTRATION OF A PERIODICALLY LOADED LINE PHASE SHIFTER USING BST CAPACITORS. Amit S. Nagra, Robert A. York, ECE Dept., University of California, Santa Barbara, CA; Troy Taylor, Padmini Periswamy, James Speck, Materials Dept., University of California, Santa Barbara, CA.

The cost of phase shifters is a major component of the cost of modern phased array antennas and it is of paramount importance to reduce this cost to ensure widespread acceptance of phased arrays in military/civilian applications. Ferroelectric thin films promise cost reduction because of two factors- 1) the ferroelectric material can be deposited relatively inexpensively using RF sputtering/MOCVD 2) the films can be processed using low cost, high volume monolithic fabrication techniques. Apart from cost, the use of ferroelectric thin films in phase shifter circuits also has potential performance advantages such as low insertion loss, high power handling capability and low DC power requirements.

Our approach is to periodically load a coplanar wave guide transmission line with tunable ferroelectric capacitors. The periodically loaded line has variable phase velocity that can be controlled by changing the value of the ferroelectric capacitors embedded in the transmission line. The design of periodically loaded phase shifters has been optimized by us for low loss performance and we have already demonstrated record low loss K-band phase shifters using this design methodology (with semiconductor varactor loaded

lines). We present here an X-band phase shifter fabricated on high resistivity silicon. The tunable capacitors used here were of the parallel plate type with RF sputter deposited Barium Strontium Titanate as the field tunable dielectric and platinum as the top and bottom electrode metal. The circuit produced 100 degrees of differential phase shift at 10 GHz and the insertion loss was less than 7 dB. Only 8 Volts of DC bias was required for the maximum phase shift, which is much lower than voltages required to control bulk ferroelectric phase shifters. This is the first demonstration of a periodically loaded line phase shifters using ferroelectric thin film capacitors.

> SESSION KK2: ELECTRIC-FIELD TUNING Chairs: James S. Horwitz and Felix A. Miranda Tuesday Afternoon, November 30, 1999 Room 201 (H)

### 2:00 PM \*KK2.1

TUNABLE RF AND MICROWAVE DEVICES — MATERIALS AND PROCESSING ISSUES. K.S. Harshavardhan, H.M. Christen, S.D. Silliman, Neocera, Inc., Beltsville, MD.

The ability to tune room temperature devices as well as cryogenic high temperature superconducting (HTS) filters has been identified as an important step in enhancing the performance of RF and microwave subsystems. With the evolution of a variety of thin film process technologies incorporating ferroelectric, ferrite and HTS films, tunable RF and microwave devices are moving toward system insertions. Technological advances in this area are, however, impeded by the large intrinsic losses in the tunable material, pointing out the need for innovative materials and materials processing approaches. This talk will focus on current issues related to materials processing. Promising approaches that have shown potential in reducing the losses and retaining tunability will be discussed, and experimental data on electrically and magnetically tunable filters will be presented.

BARIUM STRONTIUM TITANATE FILMS FOR TUNABLE MICROWAVE DEVICES. J.D. McCambridge, D.W. Face, L. Bao, DuPont Superconductivity, Wilmington, DE.

We deposited thin epitaxial films of  $Ba_xSr_{1-x}Ti_yO_3$  (x = 0.06-0.60 and y = 0.96-1.00) on lattice-matched 51 mm diameter substrates and buffer layers, including LaAlO<sub>3</sub>, CeO<sub>2</sub>/LaAlO<sub>3</sub>, MgO, and YBCO/CeO<sub>2</sub>/sapphire. We aimed to produce ferroelectric layers suitable for tunable microwave devices both at cryogenic and room temperature. The films were 0.1-0.8  $\mu m$  thick and generally (001) oriented along the growth direction. The substrates were radiantly heated between 775-925 K. We deposited the films by magnetron sputtering at RF power densities between 1.3-2.4 W/cm<sup>2</sup> in 8-18 Pa of 1:1::Ar:O<sub>2</sub>, followed by a cooldown in O<sub>2</sub>. Deposition rates ranged between 5-15 pm/s. We characterized the orientation and strain of these films by x-ray diffraction. Deposition rates, compositions, and impurities were measured by Rutherford back scattering (RBS). We measured the dielectric permittivity, tunability, and loss at low frequencies (< 1 MHz) and microwave frequencies (~ 5 GHz) from 77-325 K. We also examined the effect of post-annealing in  $O_2$  at temperatures up to 1275 K on the structural and dielectric properties of some of the films. We found that the permittivity and loss improved with the crystalline quality but the tunability depended primarily on strain.

# 3:30 PM \*KK2.3

TUNABLE DIELECTRIC THIN FILMS FOR HTS MICROWAVE APPLICATIONS. Brian Moeckly, Conductus, Inc., Sunnyvale, CA.

Thin-film tunable ferroelectrics such as SrTiO<sub>3</sub> have properties which are attractive for active tuning of high performance HTS microwave filters. However, in order to preserve the high Q values of the filters, the dielectric properties of the SrTiO<sub>3</sub> films need to be dramatically improved. We would like films possessing both a significant electric-field tunability of their permittivity and a low loss tangent at an appropriate temperature (~ 60 K). We will discuss our dielectric measurements of films whose growth protocols have been optimized to achieve these goals. We have evaluated films grown on various substrates, with different buffer layers and interlayers, with various dopants, and with low and high temperature oxygen post-anneals. We will present compositional analysis and discuss the nature of the defects in these films and their relation to the dielectric properties. In addition, we will discuss the tunability and loss obtainable for YBCO rf resonator structures incorporating the best available tunable dielectric films.

#### 4:00 PM KK2.4

LOW LOSS FERROELECTRIC  ${\rm Ba_xSr_{1-x}Tio_3}$  FILMS - A COMPARISON OF EPITAXIAL POLYCRYSTALLINE AND AMORPHOUS FILMS. Hua Jiang, Wei Hu, Vladimir Fouflyguine, Jing Zhao and Peter Norris, NZ Applied Technologies, Woburn, MA.

Low dielectric loss and high tunability of ferroelectric films are highly demanded for electrically tunable microwave devices. Many extrinsic factors could contribute to the dielectric loss in addition to the intrinsic single crystal dielectric loss, such as defects, film/substrate interfaces, oxygen deficiencies, and internal stresses. In order to identify the major loss factor, we have grown epitaxial, polycrystalline, and amorphous  $Ba_xSr_{1-x}TiO_3$  (BST) films on LaAlO<sub>3</sub> and yttrium iron garnet (YIG) and GaAs substrates using Metal-Organic Chemical Liquid Deposition (MOCLD), and measured the dielectric losses of the films. Our results indicated that the dielectric tunability of the films was proportional to the film crystallinity, however, the dielectric loss did not show a clear trend to the film crystallinity. As a matter of fact, some polycrystalline and amorphous BST films showed very low dielectric losses (tan  $\delta < 0.003$  at 400KHz), lower than the loss of epitaxial films at the same frequency. This result suggested that the internal stress of the films could be a significant source of loss, since epitaxial films had stronger internal stress due to the film/substrate lattice mismatch than polycrystalline or amorphous ones. We also grew BST multilayer films by alternatively depositing  $\mathrm{BaTiO_3}$  and SrTiO<sub>3</sub> layers. The dielectric losses of such multilayer films were smaller than that of  $\rm Ba_{0.5}Sr_{0.5}TiO_3$  solid solution films using the same MOCLD method. This result can be explained by the reduced internal stress in the multilayer films.

 $4:15\ PM\ \underline{KK2.5}$  ELECTRICALLY TUNABLE MICROWAVE DEVICES PREPARED BY RF-MAGNETRON SPUTTERING: STRUCTURE, PRO-CESSING AND PROPERTY RELATIONSHIPS. B.J. Gibbons, Y. Fan, A.T. Findikoglu, D.W. Reagor and Q.X. Jia, Los Alamos National Laboratory, Los Alamos, NM.

Recently, electrically tunable microwave devices based on  $YBa_{2}Cu_{3}O_{7-x}/SrTiO_{3}$  (YBCO/STO) multilayers have been extensively investigated. It has been demonstrated that one of the most important factors influencing the performance of these devices is dissipation loss. This loss may originate from the bulk of the STO film, the STO/substrate and STO/YBCO interfaces, and the YBCO itself. The majority of the active microwave devices reported in the literature have been fabricated using pulsed laser deposition (PLD). This method results in the presence of macroparticles on the film surface which may affect the properties of devices fabricated from these films. Thus, a deposition method which provides smoother films (interfaces) should result in improved microwave device performance. Using rf-magnetron sputtering, we have deposited a 200-nm-thick epitaxial STO layer on a  $\sim$  35-nm-thick layer of homoepitaxial LaAlO<sub>3</sub> (LAO) on LAO substrates. Subsequently, 400 nm of STO and 400 nm of YBCO were deposited using PLD. Coplanar waveguide structures with a centerline width of 20  $\mu$ m and 40  $\mu$ m wide gaps were patterned into the films. The tunability  $(\Delta f/f)$  of the STO film was found to be 4.9% at 100 V and 7.0% at 200 V (centered at pprox 4 GHz), showing an improvement over similar devices fabricated completely by PLD. Based on these data, sputter deposited-based devices will be tested for their microwave properties and compared to PLD-based devices. In addition, the effects of insitu and post deposition ozone annealing treatments on the sputtered STO films will be investigated. Preliminary data indicate that ozone treatments in the early stages of deposition result in improved structural properties of the sputtered STO.

#### 4:30 PM KK2.6

Ba<sub>1-x</sub>Sr<sub>x</sub>TiO<sub>3</sub> THIN FILM GROWTH PROCESSES AND ELEC-TRICAL PROPERTY RELATIONSHIPS FOR APPLICATION TO HIGH FREQUENCY DEVICES. J. Im , O. Auciello , S.K. Streiffer, J.A. Eastman, Materials Science Division, Argonne National Laboratory, Argonne, IL; A.R. Krauss, Materials Science and Chemistry Divisions, Argonne National Laboratory, Argonne, IL; Jianxing Li, Johnson-Matthey Electronics, Spokane, WA.

Precise control of  $\mathrm{Ba}_{1-x}\mathrm{Sr}_x\mathrm{TiO}_3$  (BST) film composition is critical for the production of high-quality BST thin films. Specifically, it is known that nonstoichiometry greatly affects the electrical properties of BST film capacitors. We are investigating the compositionmicrostructure-electrical property relationships of polycrystalline BST films produced by magnetron sputter-deposition using a single target with a Ba/Sr ratio of 50/50 and a (Ba+Sr)/Ti ratio of 1.0. It is known that such films exhibit high tunability and sufficiently low loss for some device applications, without the restrictions imposed for epitaxial film growth. It was found that the ratios of (Ba+Sr)/Ti and Ba/Sr in the deposited films depend strongly on the total pressure (Ar+O<sub>2</sub>) in the magnetron chamber and on the substrate temperature during film deposition. By changing the total process pressure

(Ar+O<sub>2</sub>), the ratio of (Ba+Sr)/Ti changed from 0.85 to 1.35 within the pressure range studied. We found similar compositional changes as a function of substrate temperature, while changes in the O<sub>2</sub>/Ar ratio did not affect the composition of the BST films. The crystalline quality as well as the measured dielectric constant, dielectric tunability, and electrical breakdown voltage of BST films have been found to be strongly dependent on the composition of the BST films, especially the (Ba+Sr)/Ti ratio. We will discuss the impact of BST film composition control, through film deposition and process parameters, on the electrical properties of BST capacitors for high frequency devices. \*Work supported by the U.S. Department of Energy, BES-Material Sciences, under Contract W-31-109-ENG-38, and by DARPA, contract # 978040.

MICROSTRUCTURE-DIELECTRIC PROPERTY RELATION IN (Ba,Sr)TiO3 FILMS GROWN BY THE COMBINATORIAL SYNTHESIS TECHNIQUES. <u>I. Takeuchi</u>, H. Chang, X.-D. Xiang, Lawrence Berkeley National Laboratory, Materials Sciences Div, Berkeley, CA; E.A. Stach, Lawrence Berkeley National Laboratory, National Center for Electron Microscopy, Berkeley, CA.

In order to better understand the phase formation processes in combinatorial syntheses as well as (Ba,Sr)TiO<sub>3</sub> film growth, we are investigating the relation between microstructural and dielectric properties of the films. To survey a large compositional variation on individual combinatorial libraries, we grow (Ba,Sr)TiO<sub>3</sub> films from; 1) precursor layers of BaF2, SrF2 and TiO2 and 2) stoichiometric layers. In both cases, compounds are formed by post-annealing amorphous layers deposited at room temperature. These methods allow fabrication of films with continuously varying composition and systematically introduced dopants. Quantitative scanning evanescent microwave microscopy is used for dielectric characterization. Novel low loss compositions identified from the combinatorial libraries have been reproduced in films grown by other techniques. The cross-sectional TEM of films made from precursors shows columnar growth of oriented grains with no evidence of isolated amorphous regions which may arise form non-uniformly diffused precursors. We have found that epitaxial films can be obtained on LaAlO3 for all the compositions studied here, except for BaTiO3 grown from stoichiometric layers: these films do not show any peaks in the XRD, but the TEM shows the presences of nanocrystalline grains. Films grown from stoichiometric layers often display higher dielectric constant compared to films deposited in-situ on heated substrates. This is attributed to the smaller amount of stress present in the post-annealed films as seen from the difference in the lattice constant from the XRD.

> SESSION KK3: MAGNETIC-FIELD TUNING Chairs: Daniel E. Oates and John Talvacchio Wednesday Morning, December 1, 1999 Room 201 (H)

### 8:30 AM \*KK3.1

FERRITES FOR TUNABLE RF AND MICROWAVE DEVICES. Gerald F. Dionne, Lincoln Laboratory, Massachusetts Institute of Technology, Lexington, MA.

Microwave systems for communications and radar require control of propagation of the rf signal. Devices that accomplish this function include phase shifters, isolators and circulators, and tunable filters. In many instances, these devices are magnetic and are based on the variable permeability of electrically insulating ferrimagnetic oxides (ferrites). A review of relations between rf propagation and material properties arising from ferrimagnetic resonance will be given, followed by a description of relevant crystallography and chemistry of the spinel and garnet ferrites. Important properties of these oxides that influence device performance include magnetization, magnetocrystalline anisotropy, magnetostriction, and ferrimagnetic resonance linewidth, each of which is sensitive to temperature. Recent advances in microwave ferrite devices have featured superconductor circuitry that promises to virtually eliminate insertion losses due to rf surface resistance in microstrip geometries. Low-loss phase shifters and circulators have been demonstrated at X-band frequencies [1] and high-Q magnetically tunable filters with superconductor resonators are being investigated [2]. For operation at cryogenic temperatures ferrimagnetic spinels and garnets will require chemical redesign in order to realize the full potential of these devices [3]. The use of single crystal ferrite in bulk or film form offers to reduce power losses and switching times. Basic magnetochemistry of ferrites will be described with emphasis on composition design tradeoffs expected in the adaptation of ferrite technology to cryogenic environments. [1] G.F. Dionne et al., IEEE Trans. Microwave Th. & Tech. 44, 1361 (1996) [2] D.E. Oates and G.F. Dionne, 1997 IEEE MTT-S Digest, p. 303. [3] G.F. Dionne, J. Appl. Phys. 81, 5064 (1997). \* This work was sponsored by Defense Advanced Research Projects Agency.

 $9{:}00~\mathrm{AM}~\underline{*KK3.2}$  APPLICATION OF FERROMAGNETIC FILMS IN TUNABLE VHF/UHF DEVICES. Rodger M. Walser, University of Texas, Electrical & Computer Engineering, Austin, TX.

Thin ferromagnetic films have narrow ferromagnetic resonance bandwidths, and large RF permeabilities that are attractive for tunable device applications. They can also be processed at low temperatures and integrated with most RF and microwave materials. At VHF frequencies, discrete tunable devices can be utilized, and magnetic loss tangents  $(Q^{-1})$  less than  $10^{-3}$  can be obtained, if the effect of resonance and eddy current losses is reduced. The material figure of merit (FOM=tuning bandwidth x Q) requires tradeoffs of the permeability, Q, and fill factor; and maximum FOMs are realized by maximizing the magnetization, anisotropy, resistivity, and minimizing magnetic damping. This paper discusses the interplay of these properties, and the fill factor and magnetic bias required for tunable inductors with Q>1000 in the 30-100 MHz range. Progress has been made with reactively sputtered CoFeTa and FeCoHf films. Their large resistivities ( $>200~\mu\text{W.cm}$ ), magnetizations ( $\sim1.5\text{T}$ ), and thicknesses ( $>1~\mu\text{m}$ ) permit large device fill factors, and microlithographed objects with large shape demagnetizing fields (>500 Oe) eliminating the need for bias magnets to achieve Q's  $\sim$ 1000. Additional considerations involve the trade of the device size with its inductance and Q. Optimum trades are achieved with solenoids, and devices with area of  $10^{-2}$  cm<sup>2</sup> and  $Q \sim 800$  have been been integrated with an electroplated tuning yoke and coil. The integration of film inductors into tunable high Q, VHF filters will also be discussed, and simulations of the filter responses of multiple pole, tunable devices will be presented.

#### 9:30 AM KK3.3

STRUCTURAL AND MAGNETIC CHARACTERIZATION OF EPITAXIAL Co-DOPED GARNET FILMS. Darren Dale, G. Hu, Vincent Balbarin, Y. Suzuki, Dept. of Materials Science and Engineering, Cornell University, Ithaca, NY.

Epitaxial crystalline thin films of  $m Y_3Fe_5O_{12}$  (YIG) doped with  $m Co^{2+}$ have been grown on (110) oriented Gd<sub>3</sub>Ga<sub>5</sub>O<sub>12</sub> substrates. The addition of Co<sup>2+</sup> in YIG films enhances the in-plane uniaxial anisotropy by up to an order of magnitude, depending on composition. Such harder garnet films have been developed to serve as an exchange bias layer for magnetically soft undoped YIG films. Exchange biased YIG minimizes unwanted power dissipation that severely limits the high frequency performance of inductor devices with pure YIG. In order to ensure  $\text{Co}^{2+}$  doping into octahedral sites to enhance the magnetic anisotropy,  $\text{Ge}^{4+}$  and  $\text{Ce}^{4+}$  have been substituted for Y<sup>3+</sup> to compensate for the charge differential between  $\text{Co}^{2+}$  and  $\text{Fe}^{3+}$ . These garnet films, prepared using pulsed laser deposition, exhibit excellent crystallinity, as determined from X-ray diffraction and Rutherford backscattering spectroscopy, as well as significant enhancement of the magnetic anisotropy.

### 9:45 AM KK3.4

SUBSTITUTED BARIUM HEXAFERRITE FILMS FOR PLANAR INTEGRATED PHASE SHIFTERS. S.A. Oliver, Northeastern University, Center for Electromagnetic Research, Boston, MA; S.-D. Yoon and C. Vittoria, Northeastern University, Department of Electrical and Computer Engineering, Boston MA.

Thick oriented hexaferrite films are being developed for use in planar integrated ferrite/semiconductor devices operational at microwave or millimeter wavelengths. One application of these materials will be for planar latching phase shifters, which may be either reciprocal or non-reciprocal devices dependent upon the orientation of the in-plane magnetization relative to the direction of wave propagation. Here the use of hexaferrite films having good in-plane square loop behavior and low coercive fields allows for tunability of the phase shift by adjusting the net film magnetization from the demagnetized state up to the full saturation magnetization value. To meet these goals, thick  $\mathrm{BaSc}_x\mathrm{Fe}_{12-x}\mathrm{O}_{19}$  films have been deposited onto A-plane sapphire substrates by pulsed laser ablation deposition, where the substitution of scandium cations for iron allows for adjustment of the film saturation magnetization and anisotropy field values and thus the optimum operating frequency of the device. X-ray diffraction patterns indicate that these films are highly phase-pure, and do not show c-axis diffraction peaks. Electron micrographs on eight micron-thick films show significant surface texturing and the presence of large facets. Magnetization measurements show these films have distinct in-plane uniaxial axis coincident with the sapphire substrate c-axis with these results showing square loop behavior with remanance field values being 90% of the full saturation magnetization and coercive field values of less than 300 Oe. The saturation magnetization and anisotropy field values for x=1.3 films were approximately 3.8 kG and

10 kOe, respectively, while the corresponding values for x=0 films were 4.2 kG and 17 kOe. Here, the saturation magnetization values for the x=1.3 films were greater than those measured for bulk samples. Work is in progress to grow even thicker films for use in fabicating low-loss planar integrated phase shifters.

#### 10:30 AM \*KK3.5

CHARACTERIZATION OF FERROMAGNETIC PEROVSKITES FOR MAGNETICALLY TUNABLE MICROWAVE SUPER-CONDUCTING RESONATORS. <u>Jaroslaw Wosik</u>, Michael Strikovski, Lei-Ming Xie, Maged Kamel, Stuart A. Long, University of Houston, Dept. of Electrical and Computer Engineering and Texas Center for Superconductivity, Houston, TX; Piotr Przyslupski, Institute of Physics, Polish Academy of Sciences, Warszawa, POLAND.

Ferrite substrates are not crystallographically compatible with high-T<sub>c</sub> superconducting (HTS) materials and can only be used in hybrid HTS microwave structures. Therefore, there is a need for materials that would be compatible with monolithic HTS thin film technology. Perovskites exhibiting ferromagnetic properties are interesting candidates for replacing ferrites. We report here on investigations of magnetic properties and microwave losses of  $Nd_xSr_{1-x}MnO_{3-y}$  (NSMO) and  $La_xCa_{1-x}MnO_{3-y}$  (LCMO) systems and on studies of magnetic tunability of superconducting microwave resonators controlled by magnetic field-dependent effective permeability  $\mu(H)$  of these materials. Several films as well as bulk samples with different oxygenation level and doping were tested for surface impedance dependence on the dc magnetic field dependence The films exhibited Curie temperatures  $T_f$  ranging from 220 K to 60 K, saturation magnetization  $4\pi M$  from 0.1 T to 0.3 T. Both a 13 GHz sapphire dielectric resonator and 3 GHz S-shaped microstripline resonator, coupled in a flip chip configuration to ferromagnetic perovskite were used. It was found that for tuning purposes the film properties have to be optimized to combine the significant magnetization and high resistivity. The correlation between the microwave losses and magnetic properties of NSMO and LCMO films as well as optimization of film properties, regarding minimum microwave losses, will be discussed.

#### 11:00 AM KK3.6

FERROMAGNETIC RESONANCE IN SINGLE-CRYSTAL BIS-MUTH IRON GARNET FILMS. V.P. Denysenkov and <u>Alex Grishin</u>, Condensed Matter Physics, Department of Physics, Royal Institute of Technology, Stockholm, SWEDEN; Takashi Okuda and Nobuyasu Adachi, Nagoya Institute of Technology, Nagoya, JAPAN.

Bi-substituted magnetic garnets exhibit large magnetooptical effects while Bi<sup>3+</sup> substitution does cause no essential optical absorption in the near infrared thus they meet all the requirements for materials for micro- and millimeter-wave nonreciprocal components and integrated optics. Thin films of pure  $\rm Bi_3\,Fe_5\,O_{12}$  (BIG)  $^1$  are of special interest since they exhibit high magnetooptical figure of merit: giant Faraday rotation of 7.35 deg/ $\mu$ m and absorption coefficient of 1.8\*10<sup>3</sup> cm<sup>-</sup> 633 nm. One of the important task in practical wave-guiding type device design and manufacture is a control of ferrite film birefringence caused by growth induced and cubic crystalline magnetic anisotropy. Here we report results on extensive BIG films characterization by Ferromagnetic Resonance (FMR) technique. BIG films have been grown onto (111)  $\rm Sm^3(Sc,Ga)^5O^{12}$  single-crystal substrate by a reactive ion-beam-sputtering technique and demonstrated by x-ray diffraction to be epitaxial. Room temperature VSM measurements revealed the in-plane magnetization of  $4\pi M = 1450 G$  with the coercive field of 50 Oe. Perfect film-substrate lattice match results in narrow FMR linewidth of 68 Oe at 16.9 GHz frequency and resonance field of 6 kOe. Full angular dependence of the resonance field gave a complete information on magnetic anisotropy: both growth induced uniaxial and cubic. The fields of anisotropy have been calculated to be:  $2K_u/M = -1310$  Oe and  $K_1/M = 60$  Oe for uniaxial and cubic magnetic anisotropy correspondingly. The experimental results on broad band spectroscopy up to 40 GHz also will be presented. <sup>1</sup> T Okuda et al, IEEE Trans. J. Magn. Japan 3, 483 (1988).

#### 11:15 AM KK3.7

 $\frac{\text{MAGNETIC}\overline{\text{ALLY}}}{\text{TUNED}} \ \text{SUPERCONDUCTING FILTERS}. \\ \frac{\text{D.E. Oates}}{\text{MA}} \ \text{and A.C. Anderson, MIT Lincoln Laboratory, Lexington,} \\ \frac{\text{MA}}{\text{MA}}.$ 

We have demonstrated magnetically tunable superconducting filters consisting of microwave circuits coupled to ferrite substrates in monolithic structures using both niobium at 4 K and YBCO at 77 K. A three-pole 1% bandwidth filter with 10-GHz center frequency, 1-dB insertion loss, and greater than 10% tunability has been demonstrated and will be described. A closed magnetic circuit and operation in the partially magnetized state prevent degradation of the surface resistance of the superconductor. This technology has the potential for compact, light weight, multipole filters with very low insertion loss, <0.5 dB and very rapid tunability, <1 $\mu$ s, due to the low

inductance of the magnetic circuits. We will also present the use of lightly coupled ferroelectric films to introduce trimming of the individual resonators in the magnetically tuned filters. The materials issues that limit performance and means to improve present performance will be discussed.

This work was supported by DARPA.

#### 11:30 AM KK3.8

NARROW-LINEWIDTH YTTRIUM IRON GARNET FILMS FOR HETEROGENEOUS INTEGRATION. M. Levy, R.M. Osgood, Jr., Columbia University, Dept of Applied Physics, New York, NY; F.J. Rachford, Naval Research Laboratory, Washington, DC; A. Kumar, H. Bakhru, SUNY at Albany, Physics Dept, Albany, NY.

Narrow-linewidth magnetic garnets are of interest in microwave systems because of their use as tunable low-loss filters. Of particular importance in this regard is the development of low-cost integrated technologies for wireless systems and phased array antennas. Here we report on the fabrication, characterization and bonding of singlecrystal films of yttrium iron garnet (YIG) for heterogeneous integration onto growth-incompatible substrates. The films are originally grown on < 111 >-oriented gadolinium gallium garnet (GGG) substrates by liquid-phase epitaxy. Separation from this substrate is done by crystal ion slicing (CIS). The CIS separation process entails the implantation of energetic helium ions into the front surface of the material. This results in the formation of a fast etching sacrificial layer. Flash annealing is used to remove residual damage in the film before detachment either by wet chemical etching or thermal shock. The virgin material has a  $4\pi M_S$  of 1070 G, and 9.5 GHz ferromagnetic resonance (FMR) linewidth of 0.55  $\pm$  0.1 Oe. The implantation (5 X 10<sup>16</sup> cm<sup>-2</sup> dosage, 3.8 MeV energy) significantly reduces the growth induced uniaxial anisotropy in these films. Upon implantation the FMR linewidth increases from 0.55 to 3 Oe. The detached (8µm-thick) samples, treated by rapid thermal annealing before etching, have linewidths ranging from 1.7 Oe to 2.5 Oe. Further reduction by post-detachment annealing is expected and is presently under investigation. Integration onto non-compatible substrates is done by direct wafer bonding. Low-temperature bonding of implanted YIG onto Si, InP and GaAs exhibits bond strengths 50% to 60% that of Si onto Si. The bond survives thermal treatment up to 400°C to  $500\,^{\circ}\mathrm{C},$  despite significantly different thermal expansion coefficients. M.L. and R.M.O. acknowledge support by DARPA/FAME under Contract No. N00173-98-1-G014. 1.M. Levy, R.M. Osgood, Jr., A. Kumar, H. Bakhru, Appl. Phys. Lett., vol.71, 2617 (1997).

#### 11:45 AM KK3.9

FREQUENCY AGILE MICROWAVE APPLICATIONS USING (BaSr)TiO<sub>3</sub>/Y<sub>3</sub>Fe<sub>8</sub>O<sub>12</sub> MULTILAYER GROWN BY PULSED LASER DEPOSITION. W.J. Kim\*, Wontae Chang, S.B. Qadri, H.D. Wu\*, J.M. Pond, S.W. Kirchoefer, J.S. Horwitz and D.B. Chrisey, Naval Research Laboratory, Washington, DC.

Ba,Sr)TiO3 thin films deposited on dielectric substrates using pulsed laser deposition (PLD) are currently being used to develop a new class of frequency agile microwave electronics based on the electric field dependent dielectric constant. A concern in the development of these devices is the large change in the characteristic impedance of the device when the dielectric constant is reduced by a factor of four or more. A novel approach to this problem is to fabricate the device from a ferrroelectric/ferrite multilayer. In a microwave transmission line, it will be possible to simultaneously tune both the inductance per unit length (magnetic field tuning of the ferrite) and capacitance per unit length (electric field effect in the ferroelectric) such that the characteristic impedance remains constant while its phase velocity through the line is continuously varied. Towards this goal, multilayers  ${\rm BST/Y_3Fe_5\,O_{12}}$  (YIG) have been fabricated. The structure of the multilayer has been characterized using X-ray diffraction and cross-section TEM. Epitaxial BST film growth was observed on (111) and (001) YIG substrates. The orientation of the BST films ((001) and (111)) on YIG strongly depend on the deposition temperature. The microwave dielectric properties of the multilayer have been measured as a function of bias field (E  $\leq$  200 kV/cm and/or H  $\leq$  5 tesla) from 1 - 20 GHz using interdigitated capacitors or coplanar transmission lines deposited on top of the dielectric film. More than a 30% change in the dielectric constant with 100 kV/cm dc field are achieved with (001) BST/ (111) YIG and polycrystalline BST/ (001) YIG. A summary of the electric field and magnetic filed effects as a function of the BST/YIG structure as well as its impact on future device structures will be presented.

#### SESSION KK4/Y10: JOINT SESSION: HIGH-FREQUENCY APPLICATIONS OF FERROELECTRICS

Chairs: Quanxi Jia and Susan Trolier-McKinstry Wednesday Afternoon, December 1, 1999 Room 304 (H)

#### 1:30 PM \*KK4.1/Y10.1

MATERIALS ISSUES IN ELECTRIC FIELD TUNABLE RF AND MICROWAVE DIELECTRICS. <u>L. Eric Cross</u>, Materials Research Laboratory, The Pennsylvania State University, University Park, PA.

Ferroelectric crystals are in general highly nonlinear dielectrics, but in the ferroelectric phase extrinsic domain wall processes generally give rise to very high dielectric loss levels. To achieve dielectric tunability under electric field, but retain low dielectric loss there has been a focus upon the paraelectric phase in incipient ferroelectrics in the perovskite structure oxides. Strontium titanate (SrTiO<sub>3</sub>) and Potassium Tantalate (KTaO<sub>3</sub>) are favorable candidates and solid solutions based on these end members, such as  $(\mathrm{Ba}_x\mathrm{Sr}_{1-x})\mathrm{TiO}_3$  and  $\mathrm{K}(\mathrm{Ta}_{1-x}\mathrm{Nb}_x)\mathrm{O}_3$  and composites containing these solid solutions are under study for room temperature application.

The focus on the paraelectric phase is understandable as both theory and experiment confirm low loss (tan  $\delta \sim \! 0.00065$  at 10 GHz) and high tunability in single crystal SrTiO3. In thin film form however, which is essential for many important device applications, both dielectric loss levels and E-field tunability are significantly compromised although adequate tunability is maintained because of the enhanced dielectric strength of the thin films. Recent improvements in film processing and performance will be reviewed and the developing understanding of the importance of crystal quality in the films underscored.

Strontium titanate has been an excellent vehicle for the study of the roles of both isovalent and aleovalent substituents in the perovskite lattice and these works will be summarized and discussed. New work to explore the low temperature pyrochlore structure ferroelectrics will be described and measurements showing promising results for both loss levels and tunability presented.

With molecular beam techniques it is possible to develop completely new single phase examples of the strontium titanate Ruddleston Popper phase crystals and to make new layer structure compounds in the  $(SrTiO_3)_x$   $(BaTiO_3)_{1-x}$  compositions. Work on these systems will be described and the dielectric properties of these new artificial compounds briefly reviewed.

#### 2:00 PM \*KK4.2/Y10.2

DIELECTRIC PROPERTIES OF (Ba,Sr)TiO<sub>3</sub> THIN FILMS TAILORED FOR TUNABLE MICROWAVE APPLICATIONS. C.L. Canedy, M.R. Burr, D. Steinhauer, S. Anlage, T. Venkatesan and R. Ramesh, Materials Research Science and Engineering Center (MRSEC), University of Maryland, College Park, MD; F.W. Van Keuls, R.R. Romanofsky, N.D. Varaljay and F.A. Miranda, NASA Glenn Research Center, Cleveland, OH.

Ferroelectric materials enjoy a large nonlinearity in their dielectric response, which gives rise to an electric field dependent permittivity. This feature makes them particularly attractive for use as electronically tunable microwave components. However, early studies have reported prohibitively large dielectric losses in the microwave frequency regime. Recently though, progress in materials processing and deposition techniques has dictated the fabrication of thin ferroelectric films of high crystalline quality and corresponding losses which make them competitive for use as resonators, filters or phase shifters. Nonetheless, it is still unclear as to how basic material properties are correlated with dielectric losses and/or the degree of tunability. In this study, we report on the evolution of the dielectric properties in epitaxial  $\mathrm{Ba}_{1-x}\mathrm{Sr}_x\mathrm{TiO}_3$  (BSTO) thin films with varying extrinsic parameters, such as stress and oxygen defect concentration. The films were grown on four separate substrates using pulsed laser deposition. These included LaAlO<sub>3</sub>,MgO,(LaAlO<sub>3</sub>)<sub>0.3</sub>(Sr<sub>2</sub>TaAlO<sub>6</sub>)<sub>0.7</sub> (LSAT) and (ZrO<sub>2</sub>)<sub>0.9</sub>(Y<sub>2</sub>O<sub>3</sub>)<sub>0.1</sub> (YSZ) coated Si. In addition, the defect chemistry was altered in a controlled fashion by annealing in reducing atmospheres of oxygen. The dielectric properties at low frequency were measured using Au/Ti interdigitated electrodes. Furthermore, assessment of the BSTO films at microwave frequencies was accomplished by incorporating the films in an actual microwave phase shifter device based on a novel coupled microstripline phase shifter (CMPS) geometry. Strain relaxation in the films was studied using glancing x-ray diffraction while oxygen defect concentration was inferred using positron annihilation studies. Finally, a novel scanning microwave microscope was used to detail the local microwave response of our films. This work was partially funded by NSF-MRSEC.

### 2:30 PM KK4.3/Y10.3

COPLANAR CAPACITORS WITH HIGH DIELECTRIC CONSTANT BaSrTiO<sub>3</sub> FOR MICROWAVE APPLICATION.

<u>Wei Hu</u>, Valdimir Fouflyguine, Jim Chi, NZ Applied Technologies, Inc. Woburn, MA.

MIM Capacitors with BST dielectrics normally exhibits a nonsymmetrical IV characteristics. This nonsymmetrical behavior can be primarily attributed to the thermal degradation of the bottom BST-electrode interface during film preparation. High leakage current and low breakdown voltage in one direction of bias are resulted. The present work examined the IV nonsymmetry and proposed a solution for this problem. For this study, MIM structures were fabricated on Si and GaAs substrates. The BST film was prepared by the sol-gel method. The deposition temperature was varied from 400 to 600 C The relative dielectric constants are typically around 300 and can be as high as 470. The bottom electrode is Pt e-beam deposited on several combination of metals and dielectrics to minimize the stress in the BST film. After capacitor fabrication, the IV characteristics were measured over a temperature range from room temperature to  $200\mathrm{C}$ with voltage varied from -15 V to 15 V. The IV characteristics are analyzed assuming Schottky emission mechanism. It is found that the barrier height for the bottom interface is significantly lower than the top. This barrier lowering may be explained by the thermal degradation during the film deposition. Since the lowered barrier height will cause a higher large leakage current in one direction, it needs to be avoided for circuit applications. To solve this problem, a novel electrode structure is proposed to eliminate the nonsymmetrical IV characteristics by putting the two electrodes both on the top side of the BST film. This structure is seen to be effective in reducing the leakage current and increasing the breakdown voltage. The capacitance per unit area, however, is reduced by a factor of four. Such a reduction is tolerable, since the BST capacitors is still one order of magnitude higher than the conventional capacitors with the silicon oxynitride as the inter-electrode dielectrics. In addition to reducing the leakage and increasing the breakdown voltage, an additional benefit for the present structure is the extremely low parasitic inductance compared with the standard MIM structure. Such a low parasitic results in excellent high frequency properties. Both these benefits with the present structure may advance the application of BSTs to the microwave devices and circuits.

#### 2:45 PM KK4.4/Y10.4

DOPED BARIUM STRONTIUM TITANATE THIN FILMS FOR INTEGRATED CAPACITORS. Gregory T. Stauf, Phil S. Chen, Witek Paw and Jeffrey F. Roeder, Advanced Technology Materials Inc., Danbury, CT.

There has been significant interest recently in use of BaSrTiO $_3$  (BST) thin films for integrated capacitors; these devices have benefits for high temperature and high frequency operations, particularly when electrically tunable devices are required. We will discuss the electrical properties of BST thin films which make them suitable for these applications, as well as the impact of processing conditions on specific film properties. Voltage withstanding capability and high frequency loss are particularly important parameters for determining applications for these devices. By addition of a dopant to BST films grown by metalorganic chemical vapor deposition (MOCVD), we have increased breakdown voltages in BST films by as much as a factor of two, to approximately 2 MV/cm, potentially raising their energy storage density values significantly.

#### 3:30 PM \*KK4.5/Y10.5

THE COMBINATORIAL APPROACH TO TUNABLE FERRO-ELECTRIC MATERIALS. XiaoDong Xiang, Lawrence Berkeley National Laboratory, Materials Sciences Division, Berkeley, CA.

We have established a comprehensive combinatorial methodology in order to search for novel and improved ferroelectric materials for tunable microwave devices. Using a discrete combinatorial scheme, we perform systematic doping study where up to thousands of different combinations of dopants and host materials are generated on individual chips. We have identified several dopants that help reduce the microwave loss in (Ba,Sr)TiO<sub>3</sub>. Alternatively, we have also developed a continuous-compositional-spread technique where refined compositional search is performed in a given chip. We have identified a novel low loss compositional region from a (Ba,Sr,Ca)TiO3 "ternary-phase-diagram" chip. We use the scanning evanescent microwave microscope (SEMM) to rapidly scan and screen our combinatorial samples. SEMM allows quantitative electrical impedance microscopy of ferroelectric/dielectric materials with high spatial resolution (< 0.1 micron. The microscopy is also used to investigate tunability by applying a voltage to a localized spot in a

#### 4:00 PM KK4.6/Y10.6

BISMUTH PYROCHLORE FILMS FOR DIELECTRIC APPLICATIONS. Wei Ren, Ryan Thayer, Clive A. Randall, Susan Trolier-McKinstry, Materials Research Laboratory and Department of Materials Science and Engineering, The Pennsylvania State University, University Park, PA.

Bismuth pyrochlore ceramics have low temperature coefficients of capacitance, good microwave properties, and can be prepared at relatively modest temperatures (~1000 - 1100°C). This work focuses on the preparation and characterization of thin films in this family for the first time. A sol-gel procedure using bismuth acetate in acetic acid and pyridine, in combination with zinc acetate dihydrate and niobium ethoxide in 2-methoxyethanol was developed. The solution chemistry was adjusted to prepare  $(\mathrm{Bi}_{1.5}\mathrm{Zn}_{0.5})(\mathrm{Zn}_{0.5}\mathrm{Nb}_{1.5})\mathrm{O}_7$  and  $\mathrm{Bi}_2(\mathrm{Zn}_{1/3}$ Nb<sub>2/3</sub>)<sub>2</sub>O<sub>7</sub> films. Solutions were spin-coated onto platinized Si substrates and crystallized by rapid thermal annealing. In both cases, crystallization occurred by 550°C into the cubic pyrochlore structure.  $(\mathrm{Bi}_{1.5}\mathrm{Zn}_{0.5})(\mathrm{Zn}_{0.5}\mathrm{Nb}_{1.5})\mathrm{O}_7$  films remained in the cubic phase up to crystallization temperatures of 750°C, while the structure of the  $\mathrm{Bi}_{2}(\mathrm{Zn}_{1/3}\mathrm{Nb}_{2/3})_{2}\mathrm{O}_{7}$  thin films is dependent of the firing temperature: cubic below 650°C and orthorhombic above 750°C. A mixture of cubic and orthorhombic structures is found at 700°C. The resulting BZN films are dense, uniform, smooth (rms roughness of < 5 nm) and have a very small leakage current. In terms of the dielectric properties, cubic bismuth zinc niobate films show dielectric constants up to 150, a negative temperature coefficient of capacitance, TCC, (= - 400 ppm/°C), tan  $\delta$  < 0.01, and a field tunable dielectric constant. Orthorhombic films showed smaller dielectric constants (~80), low tan  $\delta$  (1%), positive TCC, and field independent dielectric constants. TCC could be adjusted to new 0 ppm/°C using a mixture of orthorhombic and cubic material.

#### 4:15 PM KK4.7/Y10.7

STRUCTURAL AND DIELECTRIC PROPERTIES OF PULSED LASER DEPOSITED PYbN-PT THIN FILMS. Veronique Bornand, Susan Trolier-McKinstry, The Pennsylvania State University, Department of Materials Science and Engineering, Materials Research Laboratory, University Park, PA.

The demonstration of high strain, high piezoelectric coefficients, and high electromechanical coupling constants in relaxor ferroelectric single crystals in the Pb[B/B//]O<sub>3</sub>-PbTiO<sub>3</sub> family make them promising candidates for micro-electromechanical systems (MEMS) Thus, this study focuses on the preparation and characterization of Thus, this study locates on the preparation and characterization of epitaxial (1-x) Pb[Yb<sub>1/2</sub>Nb<sub>1/2</sub>]O<sub>3</sub> - x PbTiO<sub>3</sub> (PYbN-PT) thin films. The high Curie point (~360°C) at the morphotropic phase boundary  $(x \sim 0.5)$  should greatly improve the high temperature capabilities of piezoelectric driven MEMS relative to the lead magnesium niobate-based compounds. Heterostructures consisting of (100) LaAlO<sub>3</sub> substrates, a SrRuO<sub>3</sub> metallic oxide bottom electrode and PYbN-PT ferroelectric film were deposited by pulsed laser deposition. The influence of deposition parameters such as the chamber pressure, the substrate temperature, the laser frequency, and the target composition on the microstructural and dielectric properties was investigated. High quality films could be grown in the 600-660°C temperature range, with a dynamic O<sub>3</sub>/O<sub>2</sub> pressure of 300 mTorr and high laser repetition rates. 4-circle X-ray diffraction analyses, performed on optimized samples, confirmed the (001) orientation of the perovskite sub-cells in each layer, as well as the < 110 >PYbN-PT | [110] SrRuO<sub>3</sub> in-plane heteroepitaxial relationships. Scanning electron microscopy studies revealed well-defined and homogeneous microstructures. Most of the films show room temperature dielectric constant greater than 1500 associated with low dielectric losses ( $\sim 0.03$ -0.05) and exhibit saturated hysteresis loops with remanent polarizations up to  $P_r=40~\mu\mathrm{C.cm}^{-2}$ . Furthermore, the stabilization of the < 001 >-orientation was observed to enhance the fatigue resistance. < c>-axis oriented PYbN-PT thin films do not show polarization fatigue up to  $10^{11}$  cycles, making these heterostructures attractive for non-volatile ferroelectric memories, in addition to ferroelectric actuators or sensors.

### 4:30 PM KK4.8/Y10.8

PULSED LASER DEPOSITED  $Na_{0.5}K_{0.5}NbO_3$  THIN FILMS: FROM SUPERPARAELECTRIC STATE TO FERRO-ELECTRICITY. Choong-Rae Cho and Alex Grishin, Condensed Matter Physics, Dept of Physics, Royal Institute of Technology, Stockholm, SWEDEN.

 $\mathrm{Na}_x\mathrm{K}_{1-x}\mathrm{NbO}_3$  is the continuous solid solution of potassium and sodium niobates, which shows a number of ferroelectric phases experienced coupled structural-polarization phase transitions.  $\mathrm{Na}_{0.5}\mathrm{Ko}_{0.5}\mathrm{NbO}_3$  (NKN) possesses a high spontaneous polarization of  $30~\mu\mathrm{C/cm}^2$ , high piezoelectric constant  $\mathrm{d}_{33}$  of  $160~\mathrm{pC/N}$ , strong dependence of dielectric permittivity on electric field with low loss. Therefore it is considered as a promising candidate for various applications as piezoelectric micro-sensors and actuators, electrically tunable micro- and millimeter waveguides, surface and bulk acoustic wave devices. Recently we have reported self-assembled highly c-axis oriented single-phase NKN thin films pulsed laser deposited (PLD) onto polycrystalline and amorphous substrates (Appl. Phys. Lett., 75, July 12, 1999) and epitaxial NKN/(La,Sr)MnO\_3

heterostructures on SrTiO<sub>3</sub> single-crystal (Mat. Res. Soc. Symp. Proc., MRS Spring 1999 meeting). Here we present improved as well as extended crystalline and electrical properties of PLD-NKN thin films on polycrystalline Pt and oxidized Si substrates. Phase composition and microcrystalline properties of fabricated NKN films have been found to be crucially dependent on ambient gas pressure, laser fluence and repetition rate, substrate temperature and target-to-substrate distance. These effects will be discussed in the framework of the model of  $discriminated\ thermalization$  of laser-ablated ions and neutral species in laser plume. Electrical performance of NKN films has been successfully tailored from low loss - high electrically tunable superparaelectric state to ferroelectric state characterized by high remnant polarization and piezoelectric activity. Films deposited at low oxygen pressure regime contain NKN nanocrystallites embeded in the sodium poor potassium niobate matrix, exhibit low loss  $tan\delta$  of 0.3%, dielectric permittivity of 175 which is frequency independent (from audio frequencies up to 100 kHz) and 5% tunable at the field of 100 kV/cm. Films grown at high oxygen pressure have been found to be single-phase as well as strongly c-axis oriented due to the effect of self-assembling: FWHM of NKN-200 reflection is less than 2° while Pt-200 substrate reflection was about 15°. Ferroelectric measurements yield remnant polarization of 20  $\mu \rm C/cm^2$ , spontaneous polarization of 30  $\mu \rm C/cm^2$  at electric field of 80 kV/cm, electric resistivity of 100 G $\Omega$ -cm at 10 kV/cm, dielectric permittivity of 500 and  $\tan\delta$  less than 3%. Results of piezoelectric measurements for ferroelectric films also will be presented.

#### 4:45 PM KK4.9/Y10.9

A SYSTEMATIC STUDY OF FERROELECTRIC Ba<sub>x</sub>Sr<sub>1-x</sub>TiO<sub>3</sub> FILMS. Hua Jiang, Vladimir Fouflyguine, Wei Hu, Jing Zhao and Peter Norris, NZ Applied Technologies, Woburn, MA; A. Drehman, S. Wang and P.Yip, Air Force Research Laboratory, Hanscom AFB, MA.

Metal-Organic Chemical Liquid Deposition (MOCLD) has many advantages in fabricating ferroelectric films. Its simplicity and economic nature made it most suitable for doping and multicomponent materials development. Using MOCLD technology, we have systematically grown  $\mathrm{Ba_xSr_{1-x}TiO_3}$  (BST) films (x = 0, 0.1, 0.25, 0.4, 0.5, 0.6, 0.75, 0.90, and 1) on a variety of substrates and at different temperatures. Excellent epitaxial BST films have been obtained. The c-axis and a-b plane alignments of the BST films grown on LaAlO3 substrates were about 0.06° and 0.5°, respectively, measured by a four-circle high resolution x-ray diffractometer. The correlation of film quality (in terms of orientation, grain size, dielectric constant, dielectric tunability, dielectric loss and leakage) to the growth temperature and oxygen treatment have been investigated. We also doped BST films with Ca, Mg, and rare earth elements, and studied the film structures and dielectric properties influenced by the doping. This work demonstrated that MOCLD can be used to grow very low loss (  $\tan\delta < 0.003)$  BST ferroelectric films. We also grew multilayer of alternating BaTiO<sub>3</sub>/SrTiO<sub>3</sub> and BST1/BST2 (BST1 has different composition from BST2) using the same MOCLD method. We found that the dielectric constant value of the BaTiO<sub>3</sub>/SrTiO<sub>3</sub> multilayer structure was very similar to that of a  $(\mathrm{Ba}_{0.5}\mathrm{Sr}_{0.5})\mathrm{TiO}_3$  solid solution film except that it remained nearly constant over a wide temperature range (-200°C to 0°C). This structure may find a very important application where temperature variation is a key consideration.

# SESSION KK5/Y15: JOINT POSTER SESSION: FERROELECTRICS

Chairs: Orlando Auciello and Quanxi Jia Wednesday Evening, December 1, 1999 8:00 P.M. Exhibition Hall D (H)

#### KK5.1/Y15.1

EFFECT OF THE OXYGEN PARTIAL PRESSURE ON THE MICROSTRUCTURE AND PROPERTIES OF BARIUM STRONTIUM TITANATE THIN FILMS SYNTHESIZED BY PULSED LASER DEPOSITION. C.G. Fountzoulas, Eric H. Ngo, C.W. Hubbard, P.C. Joshi and M.W. Cole, Army Research Laboratory, Weapons Materials Directorate, APG, MD.

Thin films of novel barium strontium titanate (BSTO), deposited by the pulsed laser deposition (PLD) technique exhibit excellent electronic properties including tunable dielectric constants and low electronic loss. The microstructure of the film influences the electronic, mechanical properties (internal stresses and adhesion), important factors affecting the mechanical integrity and reliability of a device made of these thin films, which in turn influence the performance of the film. Films of 1  $\mu \rm m$  nominal thickness were deposited simultaneously on silicon and on single crystals of sapphire. The synthesis of the films took place at 700°C and various partial oxygen pressures. The microstructure and crystallinity of the BSTO films were studied with the aid of x-ray diffraction analysis, scanning

electron microscopy (SEM) and FT-Raman spectroscopy. The nanohardness, modulus of elasticity, cohesion and adhesion and wear properties of the films were studied with the aid of a nanohardness indenter and a ball-on-disk tribometer. The electronic, mechanical, physical properties and an initial microstructural zone model of these films will be discussed as a function of the partial oxygen pressure. These results will be combined with the results of our previous work<sup>1</sup> on the effect of substrate temperature on above electronic and mechanical properties. The substrate influence on the microstructure and properties will also be presented and discussed.

<sup>1</sup> 'Mechanical Properties of Ferroelectric Composite Thin Films', C.

<sup>1</sup> 'Mechanical Properties of Ferroelectric Composite Thin Films', C G. Fountzoulas\* and Somnath Sengupta, Thin Films-Stresses and Mechanical Properties VII, MRS Vol. 505 (1998)

#### KK5.2/Y15.2

PARAELECTRIC COMPOUNDS BASED ON SUBSTITUTED BaTiO<sub>3</sub>. S.C. Tidrow, <u>A. Tauber</u>\*, D.D. Brickerd, A. Lee, B. Rod, E.D. Adler, M.S. Patterson, U.S. Army Research Laboratory, Adelphi, MD; \*Under contract with Geo-Centers, Inc.

Cubic compounds exhibiting paraelectric behavior have been identified in systems of solid solutions between BaTiO<sub>3</sub> and perovskites that are dielectrics. Over the composition ranges studied, many compounds are cubic and their lattice parameters obey Vegard's Law. Many compositions exhibit a ferroelectric transition below room temperature. Within the paraelectric range the dielectric constant, and loss tangent are characterized with regard to temperature and microwave radiation. The variation of dielectric constant with applied dc voltage is also reported. Thin films, prepared using pulsed laser deposition, mainly grow epitaxial on LaAlO<sub>3</sub> and LSAT substrates as shown from x-ray diffraction data.

#### KK5.3/Y15.3

MECHANISMS OF TUNABILITY AND LOSS IN FERRO-ELECTRIC THIN FILMS. X.X. Xi, A.A. Sirenko, Anna M. Clark, I.A. Akimov, J. H. Hao and Weidong Si, Penn State University, Department of Physics, University Park, PA.

Strain, oxygen vacancies, and interfacial effects have been recognized to play important roles in determining the tunability and loss properties in ferroelectric thin films. In this talk, I will use incipient ferroelectric  $SrTiO_3$  thin films as an example to illustrate these effects in the thin films. I will present our structural, optical, and dielectric measurements in  $SrTiO_3$  thin films deposited by pulsed laser deposition. We found that strain, both lattice mismatch-induced and due to local defects such as oxygen vacancies, dramatically influences the properties of the thin films. Because of strain, the thin films are in a tetragonal structure, as evidenced by the superlattice peak in x-ray diffraction due to the tilting of the Ti-O octahedron, instead of a cubic structure as observed in single crystals, at room temperature. The cubic-to-tetragonal structural phase transition, which occurs in single crystals at about 105 K, is shifted above 800 K. The Raman scattering measurements show that the symmetry-forbidden optical phonons are active in the thin films, indicating the reduction of symmetry due to the strain. The line shape of the polar phonon shows a Fano asymmetry, indicating the existence of micro polar regions, or other local polar structures around the oxygen vacancies. The dielectric properties depends on the film thickness which can be treated as the result of dead layers at the interface with the electrodes. Further, we have measured the soft mode in the thin films using far infrared ellipsometry and showed that the soft mode is considerably hardened in the thin films compared to the single crystals. The thickness dependence of the dielectric constant can be understood in the framework of lattice dynamics in the thin films.

#### KK5.4/Y15.4

PHASE TRANSITIONS IN SrTiO<sub>3</sub> SINGLE-DOMAIN EPITAXIAL THIN FILMS. Alexander Tagantsev, Nava Setter, EPFL Swiss Federal Institute of Technology, Ceramics Laboratiry, Materials Department, Lausanne, SWITZERLAND; Nicholas Pertsev, A.F. Ioffe Physico-Technical Institute, St. Petersburg, RUSSIA.

A Landau-Ginsburg-Devonshire-type theory is used to describe the mechanical substrate effects upon equilibrium states and phase transitions in (001)-  $SrTiO_3$  single-domain epitaxial thin films. The misfit straintemperature phase diagram of  $SrTiO_3$  films is developed allowing for the existence of two coupled instabilities (structural and ferroelectric) in this crystal. It is shown that  $SrTiO_3$  films remain paraelectric down to 0 K only over a narrow range of small, negative misfit strains. It is found that the mechanical interaction with the substrate can result in changes in the sequence of phase transitions. For example, under certain conditions, the first transition upon cooling can be ferroelectric and only on further cooling the material undergoes the antiferrodistorsive phase transition.

#### KK5.5/Y15.5

MICROSTRUCTURE AND MICROWAVE RESPONSE OF YBCO AND YBCO / SrTiO<sub>3</sub> THIN FILMS ON 3-INCH DIAMETER SAPPHIRE WAFERS GROWN BY PLD. Michael Lorenz, Holger Hochmuth, Dieter Natusch, Thomas Thaerigen, University of Leipzig, Faculty of Physics and Earth Sciences, Leipzig, GERMANY; Vasily L. Svetchnikov, H.W. Zandbergen, National Center for High Resolution Electron Microscopy, Delft, THE NETHERLANDS; Gerhard Kaestner, Dietrich Hesse, Max-Planck-Institute for Microstructure Physics, Halle/Saale, GERMANY.

A highly reproducible pulsed laser deposition (PLD) process is set up for YBCO:Ag and YBCO:Ag / SrTiO3 thin films on both sides of 3-inch diameter sapphire wafers to be used as tunable microwave filters for satellite and mobile communication systems. The routinely deposited YBCO:Ag films on CeO2 buffered sapphire show laterally homogeneous maps of microwave surface resistance  $R_s$  of about 45  $m\Omega$  at 145 GHz and 77 K. The  $R_s$  at 8.5 GHz and 77 K determined in the center position of the YBCO:Ag films is about 380  $\mu\Omega$  and remains constant up to a microwave surface magnetic field of  $7-10~\mathrm{mT}$ . The optimum Ag-content of the PLD-YBCO target was determined to be about 4 weight-%. The DC-electrical and microwave properties of YBCO:Ag thin films on dielectric SrTiO<sub>3</sub> films for electrically tunable microwave devices depend remarkably on the microstructure of the underlying SrTiO3 layer. For deposition of YBCO / SrTiO3 multilayers on CeO2 buffered sapphire the flexible PLD technique appears as a very suitable and effective deposition technique. TEM cross sections of the large-area and double-sided PLD-YBCO:Ag thin films on R-plane sapphire with CeO<sub>2</sub> buffer layers show typical defects like stress modulation, stacking faults, a-axis oriented grains, precipitates and interdiffusion layers. Lower oxygen partial pressure during PLD increases the number of stacking faults in the YBCO structure, but reduces the a-axis oriented fraction, resulting in lower microwave surface resistance. The results show the state of the art and hints to a further improvement of YBCO and YBCO / SrTiO3 thin films on technologically important substrates for applications as microwave devices. This work is supported by the German BMBF and by Robert BOSCH GmbH Stuttgart.

#### KK5.6/Y15.6

OPTIMIZATION OF RF SPUTTERED BARIUM STRONTIUM TITANATE (BST) THIN FILMS FOR HIGH TUNABILITY. T.R. Taylor<sup>1</sup>, P. Padmini<sup>2</sup>, J.S. Speck<sup>1</sup> and R.A. York<sup>2</sup>. <sup>1</sup>Materials Research Laboratory <sup>2</sup>Department of Electrical and Computer Engineering, University of California, Santa Barbara, CA.

Ferroelectric thin films are currently being used to develop tunable microwave circuits based on the electric field dependence of the dielectric constant. (Ba<sub>0.5</sub>Sr<sub>0.5</sub>)TiO<sub>3</sub> films prepared on Pt/TiO<sub>2</sub>/ SiO2/Si substrates are found to exhibit a tunability {defined as  $\epsilon_{r(max)}/\epsilon_{r(min)}$ ;  $\epsilon_{r(max)}$  is the relative dielectric constant at zero bias and  $\epsilon_{r(min)}$  is the relative dielectric constant at a higher or defined field} of nearly 4:1. This is the highest reported tunability to the authors knowledge for sputtered films and until now such tunabilities were only realizable by MOCVD. Our main focus has been to optimize the sputtered BST films for higher tunability for applications such as varactor-diode replacements in transmission lines and the non-linear medium in frequency triplers. BST films were systematically prepared under a range of Ar/O<sub>2</sub> ratios, total pressure and substrate temperature; it was found that the texture of the deposited film depended on each of these parameters. Predominatly (100) texturing results from an Ar/O<sub>2</sub> ratio of 90/10 (sccm), a sputtering pressure of 50 mT and a 550°C substrate temperature. The dielectric permittivity  $(\epsilon_r)$  versus bias (V) characteristics of these films shows the highest tunability for (100) textured films. The large field dependence of the dielectric permittivity of the (100) textured films has been attributed to the biaxial tensile stress imposed by Si on BST making the quasi-polar axis (c-axis) oriented in-plane; thus, leading to the quasi a-axis parallel to the film normal. In a proper tetragonal ferroelectric  $\epsilon_c < \epsilon_a$ , which is in agreement with field induced quasi poling and lowering of the dielectric permittivity. The tensile stress in the film arises due to the difference in thermal expansion coefficients between the film  $\sim 7-8\times 10^{-6}/^{\circ}\mathrm{C}$  and the Si 2.5 ×10<sup>-6</sup>/°C substrate. This has prompted us to study growth on other substrates with low to high thermal expansion coefficient such as glass  $\{0.75\times10^{-6}/^{\circ}\mathrm{C}\}\$  and sapphire  $\{8\times10^{-6}/^{\circ}\mathrm{C}\}\$ .

### KK5.7/Y15.7

EPITAXIAL Ba<sub>x</sub>Sr<sub>1-x</sub>TiO<sub>3</sub> THIN FILMS FOR TUNABLE DEVICES: CORRELATION OF DIELECTRIC PROPERTIES WITH MICROSTRUCTURE. C.M. Carlson<sup>1</sup>, T.V. Rivkin<sup>2</sup>, P.A. Parilla<sup>2</sup>, J.D. Perkins<sup>2</sup>, J.C. Price<sup>1</sup>, P. Ahrenkiel<sup>2</sup> and D.S. Ginley<sup>2</sup>; <sup>1</sup>University of Colorado, Boulder, CO, <sup>2</sup>National Renewable Energy Laboratory, Golden, CO.

 $\mathrm{Ba}\mathit{x}\mathrm{Sr}_{1-}\mathit{x}\mathrm{TiO}_3$  (BST) is a ferroelectric material that has a large

electric-field-dependent dielectric constant and relatively low losses above the ferroelectric transition temperature, which can range from a few Kelvin to over 400 K depending on the Ba:Sr ratio. This makes BST potentially useful as the tunable element in a variety of tunable devices over a large temperature range both at RF and microwave frequencies. Using pulsed laser deposition, we have deposited high-quality epitaxial BST films on MgO and LaAlO3 (LAO) substrates. These BST films exhibit peak dielectric constants  $\epsilon/\epsilon_0$ 6000 with a change in dielectric constant of >65% with an applied dc field of  $\sim 7 \text{ V}/\mu\text{m}$ . We characterize the BST films both at 1 MHz and 1-2 GHz using coplanar capacitors patterned on the BST surface. We examine the correlation between structural and dielectric properties of the BST films before and after an ex-situ anneal in flowing  $O_2$  at  $\sim$ 1100 °C for 5 hours. According to recent theories, the biaxial strain imposed on the film by the substrate affects the temperature dependence of  $\epsilon/\epsilon_0$ .[1,2] We attempt to accurately measure this biaxial strain before and after the anneal using x-ray diffraction and compare with the observed  $\epsilon/\epsilon_0(T)$  measurements. X-ray diffraction also shows that annealing can improve the crystal orientation. Atomic force microscopy (AFM) shows that the anneal decreases the already low surface roughness to  $\sim 3$  Å RMS, and transmission electron microscopy (TEM) shows that the density of dislocations near the substrate interface decreases from  $> 10^{12}$  cm<sup>-2</sup> to  $\sim 10^{11}$  cm<sup>-2</sup>, resulting in higher maximum  $\epsilon/\epsilon_0$  and tuning. The optimization of the tunable dielectric properties through control of microstructure will allow the best possible device performance. [1] S.B. Desu, V.P. Dudkevich, P.V. Dudkevich, I.N. Zakharchenko,

S.B. Desu, V.P. Dudkevich, P.V. Dudkevich, I.N. Zakharchenko, and G.L. Kushlyan, Mat. Res. Soc. Symp. Proc. 401 (1996) 195.
 N.A. Pertsev, A.G. Zembilgotov, and A.K. Tagantsev, Phys. Rev. Lett. 80(9), (1998) 1988.

#### KK5.8/Y15.8

GROWTH AND CHARACTERIZATION OF  $Ba_{1-x}Sr_xTiO_3$  (BST) THIN FILMS FOR HIGH FREQUENCY DEVICES. <u>P.K. Baumann</u>, S.K. Streiffer, O. Auciello, Argonne National Laboratory, Materials Science Div, Argonne, IL; D. Kaufman, R.A. Erck, J. Giumarra, Argonne National Laboratory, Energy Technology Div, Argonne, IL.

We are investigating the synthesis of  $Ba_{1-x}Sr_xTiO_3$  (BST) thin films for applications such as capacitors for resonant circuits and high-frequency phase-shifters. BST thin films have been deposited at 650°C on platinized silicon with good thickness and composition uniformity using a large area vertical metal organic chemical vapor deposition (MOCVD) system. A computer-controlled liquid delivery system was used to inject precursors into the deposition chamber to control the films composition and ensure good reproducibility. The Ti content of the BST films was varied from 50 - 53%, and the Ba/Sr ratio was 70/30. The composition of the BST films has been analyzed using x-ray fluorescence (XRF) and Rutherford backscattering spectrometry (RBS). Patterned Pt top electrodes were deposited onto the BST films at 350°C through a shadow mask using electron beam evaporation. Annealing the BST films at 500°C in 1 mTorr of oxygen prior to Pt deposition was found to reduce the dielectric loss. A dielectric constant of approximately 900 was measured at room temperature, zero field and 1 kHz for 120nm thick films. Dielectric tunability as high as 3.5:1 was measured for an electric field of 400 kV/cm. We will report on a systematic study of the electrical properties as a function of BST growth conditions and annealing conditions of the entire capacitor structure. Work supported by the U.S. Department of Energy, BES-Material Sciences, under Contract W-31-109-ENG-38, and by DARPA, contract number 978040.

### KK5.9/Y15.9

IDENTIFICATION OF TE MORPHOTROPIC PHASE BOUNDARIES IN LEAD PEROVSKITE TERNARY SYSTEMS WITH THE COMPOSITION SPREAD METHOD. Hauyee Chang, Ichiro Takeuchi, Tsuyoshi Ohnishi, XiaoDong Xiang, Lawrence Berkeley National Laboratory, Materials Sciences Division, Berkeley, CA.

Lead containing pervoskites such as Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub> - PbTiO<sub>3</sub> have been shown to have morphotropic phase boundaries (MPB), where the dielectric constants and tunabilities are extremely large. These compositions are promising candidates for a large variety of applications in areas including piezoelectric, electro-optic and frequency agile microwave devices. The existence of MPB at different compositions for mixtures of different lead compounds is also of great theoretical interest. Thus, there is a large impetus for identification and improvement of such compositions. The combinatorial and composition spread approaches that allow the simultaneous fabrication and analysis of a large number of different compositions are well suited for the task. We have fabricated the entire ternary composition spread of PMN-PT-PZ (PbZrO<sub>3</sub>) and found peaks in the dielectric constants at the known MPB compositions. Thin film ternary composition spreads of  $Pb(A,Nb)O_3$  -  $PbTiO_3$  -  $PbZrO_3$ , where A=Ni, and Fe, are also investigated. Most compositions within the composition spreads form epitaxial thin films on LaAlO<sub>3</sub> with the layer by layer precursor method of synthesis. Entire phase

boundaries within each ternary system have been identified using the scanning evanescent microwave microscope, which measures the dielectric constants and loss of thin films. Issues concerning the synthesis of these lead-containing films will be discussed.

#### KK5.10/Y15.10

DIELECTRIC PROPERTIES OF  $Ba_{1-x}Sr_xTiO_3$ . Y. Gim, Y. Fan and Q.X. Jia, Materials Science and Technology, Los Alamos National Laboratory, Los Alamos, NM.

We report on the dielectric properties of  $\mathrm{Ba_{1-x}Sr_xTiO_3}$  (BSTO) films at a frequency of 1 MHz. We have used targets with different Ba concentrations, x = 0.1 to 0.9 at an increment of 0.1, and deposited the films by using pulsed laser deposition at 800 degrees and in an oxygen pressure of 200 mTorr. X-ray 6-20 scans show that the all the films are c-axis oriented and the c-axis lattice constant decreases with x, as expected from the bulk values. To measure the dielectric properties of the BSTO film, we have deposited 2000 Å thick Au and defined 2 mm long electrodes with a separation of 10  $\mu\mathrm{m}$ . The dielectric constant increases slowly with x, becomes a maximum of 3200 at x=0.4, and then decreases rapidly when x is larger than 0.5. To test the tunability of our films, we have also performed the capacitance vs applied voltage (C-V) measurement. The tunability ratio,  $\Delta\mathrm{C}/\mathrm{C}_{film}$ , reaches a maximum of 80% for x = 0.3 - 0.4. From the C-V curves, we have found that the peak voltage which produces a maximum capacitance monotonically decreases with x.

#### KK5.11/Y15.11

MICROSTRUCTURAL ARCHITECTURE OF (Ba,Sr)TiO<sub>3</sub> THIN FILMS FOR TUNABLE MICROWAVE APPLICATIONS. Wontae Chang<sup>1</sup>, James S. Horwitz<sup>2</sup>, Won-Jeong Kim<sup>3</sup>, Jeffrey M. Pond<sup>2</sup>, Steven W. Kirchoefer<sup>2</sup> and Douglas B. Chrisey<sup>2</sup>; <sup>1</sup>Institute for Materials Science, School of Engineering and Applied Science, George Washington University, NW, Washington, DC, <sup>2</sup>Naval Research Laboratory, Washington, DC, <sup>3</sup>SFA Inc., Largo, MD.

 $Ba_xSr_{1-x}TiO_3$  (BST, x=0.5 and 0.6) thin films have been deposited onto (100) MgO single crystal substrates by pulsed laser deposition (PLD). The room temperature capacitance and dielectric quality factor  $(Q=1/\tan\delta)$  have been measured as a function of electric field (≤ 200 kV/cm) at microwave frequencies (1 to 20 GHz) using silver interdigitated electrodes deposited on top of the BST film. It has been observed that the dielectric constant of the film and its change with electric field are closely related to film crystallinity and strain which affects the ionic polarization of the film. Amorphous BST films show high dielectric Q (> 100) with low dielectric constant (~30-200) and low dielectric tuning (< 1%), presumably due to small ionic polarization. Crystalline films have a higher dielectric constant  $(\sim 1000)$  and a higher dielectric tuning  $(\sim 65\%)$  but a lower dielectric Q (~20). As an optimal microstructure of the film for tunable microwave applications, strain-relieved large-grained (~5000 Å) polycrystalline films were deposited using a thin amorphous buffer layer of BST  $(\sim 50 \text{ Å})$ . To increase grain size (up to a few microns), BST films were prepared by the deposition of a series of a thin amorphous layers (≤ 1000 Å) deposited at room temperature followed by an encapsulated anneal at temperatures < 900°C. We will present results of the film strain and grain size on the dielectric properties of BST films and show how careful control of microstructure can lead to films with optimal properties for the tunable microwave devices.

# KK5.12/Y15.12

HELIUM-IMPLANTATION INDUCED LAYER DETACHMENT AND ELECTRICAL PROPERTIES OF SINGLE-CRYSTAL POTASSIUM TANTALATE FILMS. M. Levy, R.M. Osgood, Jr., Columbia University, Applied Physics Dept, New York, NY; R. Guo, A. Bhalla, L.E. Cross, Materials Research Laboratory, Pennsylvania State University, University Park, PA; A. Kumar, H. Bakhru, SUNY at Albany, Dept of Physics, Albany, NY.

Current interest in the development of tunable microwave systems for phased array radar and wireless communication applications has led to significant progress in ferrite and ferroelectric fabrication technologies. Various film deposition approaches are being investigated for the preparation of low-loss frequency-agile materials for integrated microwave applications. Single-crystal ferroelectric films are particularly attractive because of their low-loss and high-bandwidth. Here we report on the fabrication of mesoscopic  $(5-10\mu\text{m-thick})$ single-crystal films of potasium tantalate (KTaO3) by energetic helium ion implantation and subsequent thermal treatment and wet etching. A highly stressed sacrificial layer forms below the film upon implantation at dosages near 1 X  $10^{16}~{
m cm}^{-2}$  and energies of 3.8 MeV. Exfoliation occurs spontaneously by implantation above this threshold dosage or can be induced by wet etching or thermal treatment at lower dosages.  $8\mu$ m-thick free-standing films up to 1 mm<sup>2</sup> in size have been fabricated and their capacitances measured. Low-temperature dielectric measurements show enhancement in the susceptibility, as

expected for Curie-Weiss behavior, with a Curie temperature near 20K. The temperature evolution of the capacitance indicates incipient ferroelectric characteristics. Rutherford backscattering (RBS) and x-ray crystallographic measurements are used to study the evolution of strain leading to exfoliation with implantation dosage. Direct wafer bonding of implanted single-crystal KTa<sub>3</sub> onto semiconductor platforms will also be presented. M.L. and R.M.O. acknowledge support by DARPA/FAME under Contract No. N00173-98-1-G014.

#### KK5.13/Y15.13

ROLE OF SrRuO<sub>3</sub> BUFFER LAYERS IN ENHANCING DIELECTRIC PROPERTIES OF Ba<sub>0.5</sub>Sr<sub>0.5</sub>TiO<sub>3</sub> TUNABLE CAPACITORS. <u>S.J. Park</u>, J.H. Sok and E.H. Lee, Electronic Materials Laboratory, Samsung advanced Institute of Technology, Suwon, KOREA; J.S. Lee Analytic Engineering Laboratory, Samsung advanced Institute of Technology, Suwon, KOREA.

Ba<sub>0.5</sub>Sr<sub>0.5</sub>TiO<sub>3</sub> (BST) is a first candidate material for the development of voltage-tunable microwave devices, for example, filter, phase-shifter and VCO. In this work, the crystal structures and dielectric properties of BST film are investigated with and without SrRuO<sub>3</sub> (SRO) buffer layers. BST and SRO thin films are sequentially prepared by PLD and Au/Ti metal electrodes are ex-situ fabricated by a DC magnetron sputtering system. The capacitance and dielectric loss of the capacitors have been measured as a function of bias voltages at room temperature using a low frequency LCR meter. For the high frequency characteristics (~2GHz), a microstrip resonator with ~2GHz resonance frequency and the center coupling design is fabricated. Using flip-chip attached BST capacitor at the position of the center coupling on the microstrip resonator, we obtained its dielectric loss and tunability. The microwave loss probably due to the interfacial reaction, oxygen deficiency in the surface of the BST film was obviously enhanced by the SRO buffer layer.

SESSION KK6: POSTER SESSION:
MAGNETIC AND OTHERS
Chairs: Daniel E. Oates and Jaroslaw Wosik
Wednesday Evening, December 1, 1999
8:00 P.M.
Exhibition Hall D (H)

#### KK6.1

TUNABLE PHOTONIC CRYSTALS. Alexander Figotin, Yuri Godin, Ilia Vitebsky, University of California at Irvine, Department of Mathematics, Irvine, CA.

A photonic crystal is called tunable if its electromagnetic spectrum can be controllably altered by an external steady or quasi steady field, strong electromagnetic radiation, etc. The property of tunability has a tremendous impact on a variety of practical applications. For instance, one of the obstacles for applications of two- and three-dimensional photonic crystals has been a certain degree of unpredictability of their bandgap structure. Since it is unfeasible to redo or even to slightly modify the already built physical composite structure, the property of tunability seems to be the only way to solve the problem. Furthermore, if we can substantially alter the physical characteristics of one of the constitutive components, then the same photonic device can be used for several different purposes or in a broader frequency range. Another opportunity emerges when an external field alters qualitatively the dielectric properties of the medium. For instance, an external magnetic field can produce appreciable electromagnetic nonreciprocity. This effect can open up new opportunities in applications of photonic crystals. In this communication we explore different realizations of tunable photonic crystals as well as put forward some new ideas related to the electromagnetic properties of periodic composite structures exposed to an external magnetic field.

#### KK6.2

OPTIMAL DOPING PROFILE FOR A GaAs MM-WAVE LINEAR PHASE MODULATOR. <u>Vladimir Sokolov</u>, Philip Cheung, Stephanie M. Carlson, Xiaobo Zhang, Justice J. Calane, and Timothy T. Childs, TLC Precision Wafer Technology, Inc., Minneapolis, MN.

Because of the ability of molecular beam epitaxy (MBE) to precisely control the semiconductor doping profile, it is possible to optimize the profile for a specific device or circuit function. This paper describes a mathematically-derived and physically realizable doping profile for the epitaxial n-layer of a GaAs varactor diode that can be used to fabricate a linear phase modulator (LPM) monolithic microwave integrated circuit (MMIC) providing exactly linear and continuous phase shift versus applied voltage. Specifically, the design of a 32 GHz LPM that allows over 270 degrees of voltage-controlled linear phase shift is described. LPMs are important components for modern mm-wave communications systems, phased array radar systems, and

for general electronic phase tuning and trimming. The LPM design consists of a 4-varactor reflection circuit and a 3-dB quadrature coupler for separating the incident and reflected signals. Two identical branches, each consisting of a pair of varactor diodes connected in shunt to a 50-ohm transmission line and separated by a quarter-wave length of line, are connected to the coupled and direct ports of the 3-dB coupler. The phase-shifted output is taken from the normally isolated port of the coupler. A common voltage applied to all four varactors controls the phase. Starting with the expression for the reflection coefficient of the ideal lossless circuit, the analysis derives an expression for the varactor doping profile, which when used in conjunction with the reflection circuit, results in a linear phase-versus-applied voltage characteristic. Phase characteristics other then linear are also possible which can lead to novel circuit functions that are briefly described. Epitaxial wafers with this special doping profile have been grown by MBE at TLC. A monolithic, 32 GHz, LPM incorporating coplanar waveguide (CPW) circuitry is currently in development. Experimental results are expected by the time of the conference.

#### KK6.3

SPIN-DEPENDENT MICROWAVE LOSSES IN TRANSITION METAL ION DOPED INSULATORS. Rakhim R. Rakhimov, David E. Jones, George B. Loutts, Center for Materials Research, Norfolk State University, Norfolk, VA.

Non-resonant microwave absorption near zero magnetic field was observed in crystalline yttrium aluminates and oxide glasses doped with chromium, manganese and iron. The response is due to dielectric losses induced by low magnetic field and we describe it as magnetoinduced microwave conductivity in insulating oxides that derives from the spin-dependent charge migration in the first coordination sphere of a paramagnetic dopant ion. In contrast to the spin-polarized tunneling that was described in ferromagnets between different valence forms of transition metal ions, for example Mn<sup>3+</sup> and Mn<sup>4+</sup> in manganites, the observed effect is due to spin-dependent tunneling that occurs in the vicinity of the paramagnetic ion dilutely distributed in the material. Magneto-induced changes in conductivity can be measured by means of microwave response with the applied magnetic field and is known as microwave magneto-resistance that is commonly observed in manganites. It was found that transition to superconductive state in high-temperature superconductor ceramics also leads to non-resonant microwave absorption centered at zero magnetic field. Thus, magneto-resistance effects are usually considered in materials containing highly concentrated transition metals, as a result of coupling between metal ions in different valence states. In this presentation we will describe microwave response near zero magnetic field in the case of diluted paramagnetic systems. We will discuss the mechanism of the spin-dependent electron transport in the vicinity of a paramagnetic ion without the change in its valence state and with no need for interaction between dopant ions. The financial support for this research from the National Science Foundation (HRD-9805059) and the U.S. Department of Energy (DE-FG01-94EW11493) is gratefully acknowledged.

#### KK6.4

MICROSTRUCTURE AND MAGNETISM OF SPINEL COBALT FERRITE THIN FILMS. G. Hu, Y. Suzuki, Dept. of Materials Science and Engineering, Cornell Univ, Ithaca, NY; J.H. Choi, Chang-Beom Eom, Dept. of Mechanical Engineering and Materials Science, Duke Univ, Durham, NC.

As a highly insulating magnetostrictive material, cobalt ferrite is of interest for high frequency applications. To understand some of the issues facing high frequency ferrite materials, we have investigated the microstructure and magnetic properties of spinel CoFe<sub>2</sub>O<sub>4</sub> thin films. Epitaxial cobalt ferrite thin films have been fabricated to study the effects of strain on the magnetic properties. For films grown under compression, strain relaxation, observed by grazing incidence x-ray diffraction, is consistent wirh the inverse relationship between anisotropy field and film thickness. Study of the microstructure by Transmission Electron Microscopy, shows that the strain is relaxed by forming dislocations. In (110) oriented cobalt ferrite films, after been annealed at 1000°C in air, the in-plane easy and hard directions switch. Since bulk work has shown that the Co²+ ions in octahedral sites and tetrahedral sites exhibit crystal anisotropy of opposite sign, a study of the Co<sup>2+</sup> ion migration is under investigation. For films grown under tension, the easy direction changes from the normal to the in-plane directiion, as the film thickness is increased. All these observations confirm the dominant role of strain in the magnetic anisotropy. We have also varied the cobalt concentration to study the role of cobalt in enhancing the magnetic anisotropy and magnetostriction.

### KK6.5

ELECTRONIC STRUCTURE OF TUNABLE MAGNETIC MATERIALS MnAl AND MnGa. A. Chantis, D.O. Demchenko,

A.G. Petukhov, South Dakota School of Mines and Technology, Rapid City, SD; W.R.L. Lambrecht, Case Western Reserve University, Cleveland, OH.

Manganese based GMR compounds MnGa and MnAl can be epitaxially grown on GaAs. Being ferromagnetic they show great promise for merging semiconductors with magnetic technologies in novel structures and devices: magnetically tunable Schottky barriers, spin-dependent transistors, non-volatile magnetic memory elements, and integrated magneto-optic devices. These applications are stipulated by intriguing magnetic and magneto-transport properties of MnAl and MnGa, such as unusual magnetic anisotropy and extraordinary Hall effect. Our ultimate goal is to perform predictive calculations of the magnetic and magneto-transport properties of MnAl/GaAs and MnGa/GaAs heterostructures. The first-principle calculations of the electronic structures of bulk MnAl and MnGa materials is a necessary step in this direction. Here we report the results of our calculations of basic electronic, magnetic and structural properties of these materials. Since the unusual magnetic anisotropy of these compounds is related to their structural anisotropy in  $\tau$ -phase, our primary goal was to find equilibrium atomic structures of τ-MnAl(Ga) which are characterized by certain tetragonal distortions. It has been achieve by means of the local-density-functional calculations in the framework of the full-potential linear muffin-tin orbital (FP-LMTO) method. The lattice constant a and the c/a ratio were calculated by minimizing the total energy of crystal lattices and were found to be in good agreement with high-resolution transmission electron microscopy data. The band structure and density of states of MnAl(Ga) were calculated by means of the LMTO method with spinorbit interaction taken into account. We also investigated the magneto-crystalline anisotropy energy as a function of two lattice parameters of tetragonal MnAl(Ga). We found that due to the magnetic anisotropy, the spontaneous magnetization in GaAs/MnAl heterostructures grown in (100) direction is perpendicular to a heteroepitaxial plane. Implications of this effect on magneto-transport properties are discussed.

#### KK6.6

TUNABLE FERROMAGNETIC FILMS WITH HIGH Q's AT VHF/UHF FREQUENCIES. Alaka P. Valanju, Winston N. Win and Rodger M. Walser, University of Texas, Electrical and Computer Engineering, Austin, TX.

Due to their large saturation magnetization  $(M_s)$  and process compatibility, metallic ferromagnetic films are attractive for fabricating high Q tunable devices for VHF/UHF frequencies. To be compatible with a device Q~1000, these films must exhibit narrow FMR lines in the 1 - 10 GHz range, requiring planar anisotropy fields  $H_k$  of 100 - 1000 Oe. Since the maximum  $H_k$  that can be induced during deposition is ~100 Oe, film devices will either require an external magnet, or another source of anisotropy. Here we report the use of shape anisotropy to increase the ferromagnetic resonance frequency into the GHz frequency range. A range of shape anisotropies was induced by microlithographically patterning 1.9 micron thick films of CoFeMoSiB and 1.0 micron thick films of CoZrNb alloy thin films with as-deposited  $H_k$  values of  $\sim 20$  and  $\sim 30$ Oe respectively. The etch pattern was designed to vary  $H_k$  between 100 and 1000 Oe. The  $M_s$  and  $H_k$  of these films were measured using a B-H loop tracer and a VSM magnetometer. The measured values were found to be in excellent agreement with values computed from the shape geometry. The permeability spectra measured with a permeameter and a single loop inductor confirmed that the ferromagnetic resonance frequency varied in accordance with the Snoek's law. Even though the etched films exhibited little magnetic remanence, their measured DC and microwave permeabilities were in good agreement that calculated from Stoner-Wohlfarth theory, indicating that their multidomain configurations exhibited a single domain response. The importance of these results in the fabrication of high Q tunable VHF/UHF devices will be discussed.

#### KK6.7

NANOCRYSTALLINE THIN FILMS FOR TUNABLE HIGH Q VHF/UHF DEVICES. Winston N. Win, Jin-Young Park, and Rodger M. Walser, University of Texas, Electrical and Computer Engineering, Austin, TX.

The Stoner-Wohlfarth theory of single domain ferromagnetic thin films with small damping, shows that their magnetic loss tangents are  $<10^{-3}$  at frequencies  $\sim0.1$  their FMR frequency. Thus, high Q VHF/UHF tunable devices operating at these frequencies can be fabricated from these films, if eddy current losses can be eliminated, and the required device fill factors are realized. High film resistivities  $\rho$  are needed to reduce the eddy current losses. To eliminate external magnets for biasing the FMR well above VHF/UHF operating frequencies, large magnetizations  $M_s$  and in-plane anisotropies  $H_k$  of 100 -1000 Oe are required. Recent reports indicate that reactively sputtered nanocrystalline magnetic films have a number of attractive

features for these applications including;  $4\pi\,\mathrm{M}_s\,>\!1.5$  T,  $\rho\,>\,100\text{-}500$  $\mu\text{-}\Omega\text{-}\mathrm{cm}.$  Well oriented films with thicknesses of  $>\!1\mathrm{micron}$  can be deposited with an in plane  $\mathrm{H}_k \sim \! 80$  Oe. These films are sputtered near the metal-nonmetal percolation threshold and their nanostructural morphology is determined by compositional and process parameters that control the relative mobility, selective reactivity, and morphological exclusion of the reactive, non-magnetic Ta ions. We report on the magnetic and microwave properties of reactively sputtered (RF diode) FeCrTa:O/N and on the correlation of these properties with their composition and nanoheterogeneous structure. X-ray confirmed that the films were nanocrystalline, and that the dominant phase varied with variations in process parameters. The films were granular, with an intergranular oxynitride phase that increased with increased reactive gas pressure. Hysteresis and permeability measurements suggest that near the percolation threshold, their morphology consisted of 3 D granules with anisotropies that, in the limit, approached that of isolated magnetic spheres. Lithographed objects of these films exhibited anisotropies >800 Oe corresponding to the sum of the internal and external shape demagnetizations. The implications of these results for tunable VHF/UHF devices will be discussed.

SESSION KK7: FUNDAMENTALS

Chairs: Brady J. Gibbons and Xiaoxing Xi Thursday Morning, December 2, 1999 Room 201 (H)

8:30 AM \*KK7.1 MECHANISMS OF DIELECTRIC LOSS IN MICROWAVE MATERIALS. Alexandre Tagantsev, EPFL Swiss Federal Institute of Technology, Ceramics Laboratory, Materials Department, Lausanne, SWITZERLAND.

The present knowledge of mechanisms for dielectric loss at microwave and higher frequencies is reviewed. Relation between the loss and other properties of the material (e.g. dielectric constant) and external conditions (e.g. DC bias and mechanical stress) is discussed. The theoretical knowledge is compared to the existing experimental results.

#### 9:00 AM \*KK7.2

ANOMALOUS STRUCTURAL PHASE TRANSITIONS IN SrTiO<sub>3</sub> THIN FILMS. B.O. Wells, University of Connecticut, Dept of Physics, Storrs, CT; M. v.Zimmermann, S.M. Shapiro, Brookhaven National Laboratory, Dept of Physics, Upton, NY; Y. Zhu, Brookhaven National Laboratory, Dept of Applied Science, Upton, NY; A. Clark, X.X. Xi, Pennsylvania State University, Dept of Physics, State College, PA

The cubic to tetragonal phase transition in SrTiO3 at  $T_C \approx 105 \text{ K}$  is well known for being driven by an instability in the zone boundary phonon mode. It is the prototypical system confirming the soft-mode theory of structural phase transitions. In recent years, there has been great interest in using  ${\rm SrTiO_3}$  in frequency agile electronics, particularly at cryogenic temperatures. Such practical applications require the use of thin film samples, which in some cases have shown significantly different physical properties than bulk crystals. We have used x-ray and electron diffraction to investigate the structural phases of SrTiO<sub>3</sub> thin films. For films with the sandwich structure LaAlO<sub>3</sub>/  $\rm SrRuO_3/SrTiO_3,$  the low temperature tetragonal phase persists to at least T = 800 K. This is true for films of varying thickness, up to at least 2 microns. For films with the sandwich structure SrTiO3/ SrRuO<sub>3</sub>/SrTiO<sub>3</sub> the cubic to tetragonal transition temperature is essentially the same as for bulk crystals. This dramatic change in transition temperature is surprising and helps explain several measurements on similar samples. Work at BNL supported by the U.S. Department of Energy under contract No. DE-AC02-98CH10886. A.D. Bruce and R.A. Cowley, Structural Phase Transitions (Taylor and Francis, London, 1981) and references therein

# 9:30 AM \*KK7.3

FERROELECTRIC PHASE BEHAVIOR IN THIN FILMS: LINKS TO TUNABILITY AND LOSS. S.K. Streiffer, O. Auciello, P.K. Baumann and J. Im, Materials Science Division, Argonne National Laboratory, Argonne, IL; D.Y. Kaufman, J. Giumarra and R. Erck, Energy Technology Division, Argonne National Laboratory, Argonne,

The electric field tunability of permittivity in ferroelectric materials results directly from the nonlinear interactions governing the ferroelectric phase transition. Dielectric loss in many instances is strongly influenced by these mechanisms as well. Therefore, for applications utilizing ferroelectric thin films as tunable elements, it is essential to understand how the thin film microstructure affects the ferroelectric phase behavior. Here we address the impact of film

orientation, stress/strain, grain structure, and the substrate constraint on ferroelectric phase behavior, and relate these to quantities of interest for RF and microwave devices. Examples of the influence of these parameters will be drawn from  $(Ba_xSr_{1-x})Ti_{1+y}O_{3+z}$  films produced at Argonne National Laboratory by both metalorganic chemical vapor deposition and magnetron sputtering. Tunability and loss as a function of process conditions for the two different deposition techniques will be briefly discussed.

 $10{:}30$  AM  ${*}\mathrm{KK}7.4$  THE EFFECT OF STRESS ON THE MICROWAVE DIELECTRIC PRPOERTIES OF  ${\rm Ba_{0.5}Sr_{0.5}TiO_3}$  THIN FILMS. <u>James S. Horwitz</u>, Naval Research Laboratory, Washington, DC; Wontae Chang, George Washington University, Washington, DC; Wonjeong Kim, SFA Inc., Largo, MD; Syed B Qadri, Jeffrey M. Pond, Steven W. Kirchoefer, and Douglas B. Chrisey Naval Research Laboratory, Washington, DC.

Epitaxial  $Ba_{0.5}Sr_{0.5}TiO_3$  (BST) films have been deposited onto (100) MgO and (100) LaAlO<sub>3</sub> (LAO) substrates by pulsed laser deposition and it has been determined that film stress has a significant effect on the dielectric properties at microwave frequencies. The dielectric constant and loss tangent has been measured at room temperature and at microwave frequencies as a function of electric field (\le 200 kV/cm). The stoichiometry, doping, oxygen vacancies, growth conditions, substrate type and post-deposition annealing temperature affect the properties of the film. For BST films deposited onto (100) MgO, it is observed that, after a post-deposition anneal in O<sub>2</sub> (T  $\leq$  1000 °C), both the dielectric constant and the dielectric loss decrease. The opposite behavior is observed on LAO. In general, the dielectric constant and dielectric loss for films on LAO is higher ( $\epsilon \sim 1500$ ) compared to BST films on MgO ( $\epsilon \sim 1000$ ). The magnitude of the change in the dielectric constant with an applied DC field is directly proportional to the dielectric constant. Therefore, films on LAO exhibit more tuning than films on MgO. X-ray diffraction analysis indicates that the BST films are tetragonally distorted. The magnitude of the distortion depends on the substrate, oxygen deposition pressure and post-deposition annealing temperature and is directly proportional to the magnitude of the film dielectric constant. The properties of epitaxial BST films deposited on different substrates can be attributed to the differences in film stress caused by the mismatch between the lattice constants and thermal expansion coefficients of the film and substrate. A thin amorphous buffer layer of BST ( $\sim 50$  Å) has been used to relieve film stress due to the lattice mismatch. Stress relieved films show improved dielectric behavior for tunable microwave applications, i.e., improved tuning and lower loss.

#### 11:00 AM \*KK7.5

RESPONCE OF PERMITTIVITY OF (Ba.Sr) TiO<sub>3</sub> FILM ON ELECTRIC FIELD AND MICROSTRUCTURE. Yuri A. Boikov, Tord Claeson, Chalmers University, Physics Department, Goteborg, SWEDEN

Response of dielectric permittivity of (Ba,Sr)TiO<sub>3</sub> film on electric field depends on the chemical composition and microstructure. Substrate material and processing conditions have dramatic impact on the film microstructure. Thin  $(300-1000~\rm nm$ ) epitaxial layer of (Ba,Sr)TiO<sub>3</sub> was inserted by laser ablation between metallic oxide and/or superconducting electrodes at different condensation temperatures. Temperature, frequency and electric field dependencies of permittivity, conductance and loss tangent of the (Ba,Sr)TiO<sub>3</sub> films were investigated in the temperature range 4.2-350 K. Position of maximum in the temperature dependence of permittivity for the (Ba,Sr)TiO<sub>3</sub> film was well correlated with that for the bulk samples. Improvement of the (Ba,Sr)TiO<sub>3</sub> film microstructure was accompanied by increase in tunability of the permittivity and decrease in loss tangent at T<100 K. Loss tangent of the (Ba,Sr)TiO<sub>3</sub> film combined with superconducting electrodes is dramatically contributed by dc conductance (T=300 K). The conductance of the ferroelectric film is suppressed up to two orders of magnitude if superconducting electrodes are substituted by metallic oxide ones. Diffusion of components between bottom superconducting electrode and (Ba,Sr)TiO<sub>3</sub> during nucleation and ferroelectric film growth influence its microstructure and, as result, decrease nonlinearity of permittivity and increase conductance. Oxygen vacancies formed in a (Ba,Sr)TiO<sub>3</sub> film due to defects in the cation sublattice are responsible for high charge carrier concentration in the (Ba,Sr)TiO<sub>3</sub> film grown on superconducting electrode at high (T>750°C) condensation temperature.

# 11:30 AM KK7.6

HARDENING OF THE SOFT-MODE PHONONS IN SrTiO3 THIN FILMS DETERMINED BY FAR-INFRARED ELLIPSOMETRY AND RAMAN SCATTERING. Andrei Sirenko, Anna Clark, Ilia Akimov, Jianhua Hao and Xiaoxing Xi, Department of Physics, The Pennsylvania State University, University Park, PA; Christian Bernhard and Andrzej Golnik, Max-Planck-Institut fuer Festkoerperforschung, Stuttgart, GERMANY.

We report the experimental studies of the dielectric properties and vibrational modes of SrTiO<sub>3</sub> (STO) films with thicknesses ranging from 100 nm to 2  $\mu m.$  The films were grown using pulsed laser deposition on LaAlO  $_3$  and STO substrates with  $\rm SrRuO_3$  buffer layers. Two complementary techniques, namely Fourier-transform infrared (FT-IR) ellipsometry and conventional Raman scattering, were used for the investigation of the phonon spectra. The temperature dependence of the phonon frequencies in the films reveals a hardening of the soft-modes at low temperatures which is different from that fund in bulk crystals. This hardening is equivalent to the application of an effective electric field, and may be explained by the existence of ferroelectric micro-polar regions induced by oxygen vacancies. Results of FT-IR ellipsometry between 30 and 700 cm $^{-1}$  have been compared with the corresponding Raman spectra and radio-frequency measurements of the static dielectric function. These observations are in a complete agreement with the Lyddane-Sachs-Teller formalism, which allows us to describe the difference between the dielectric properties of bulk STO crystals and thin films by means of a hardening of the soft-modes.

11:45 AM KK7.7
THE EFFECT OF THE GROWTH MODE ON THE DIELECTRIC PROPERTIES OF SrTiO<sub>3</sub> THIN FILMS. Mikk Lippmaa, Naoyuki Nakagawa, Masashi Kawasaki, Tokyo Institute of Technology, Dept. of Innovative and Engineered Materials, Yokohama, JAPAN; Satoru Ohashi, Hideomi Koinuma, Tokyo Institute of Technology, Materials and Structures Lab, Yokohama, JAPAN.

 $SrTiO_3$  is an important material in oxide-based structures and devices. It is particularly attractive in cryogenic microwave applications where it can be used together with high-T $_c$ superconductors as a substrate, insulator, or a voltage-tunable capacitor. We have studied the influence of the growth mode on the dielectric properties of  $\rm SrTiO_3$  films by extending the pulsed laser deposition technique to temperatures well above 1000°C in an oxygen atmosphere.  $\rm SrTiO_3$  films grown at above 1000°C in the step-flow mode showed consistently higher dielectric constants, exceeding 10,000 at cryogenic temperatures, than films grown in layer-by-layer mode below 900°C. We attribute this difference to the higher crystal quality obtained by step-flow growth, as shown by AFM observation of the film surface and x-ray diffraction studies. We also present data on the growth mode monitoring by real-time reflection high-energy electron diffraction.

#### SESSION KK8: MATERIALS CHARACTERIZATIONS

Chairs: Felix A. Miranda and Ichiro Takeuchi Thursday Afternoon, December 2, 1999 Room 201 (H)

# 2:00 PM \*KK8.1

BROADBAND CHARACTERIZATION OF NOVEL DIELECTRIC THIN FILM MATERIALS AT MICROWAVE FREQUENCIES USING WAFER PROBING TECHNIQUES. James C. Booth, Leila R. Vale, Ronald H. Ono, NIST, Boulder, CO.

We present measurement results demonstrating the use of wafer probing techniques to evaluate the broadband dielectric constant and loss tangent, and also the tunability, of ferroelectric thin films of interest for frequency agile applications. The measurements are performed in a device configuration using coplanar transmission lines, which are formed by the deposition of metallic conductors onto the dielectric thin film samples. On-chip microwave calibrations are then performed using a variable temperature microwave probe station, and yield the capacitance and conductance (for a given temperature and bias voltage) of the coplanar transmission lines over the broad frequency range from 100 MHz to 26 GHz. The dielectric constant and loss tangent vs. frequency are then extracted from these calibrated measurements using simple models. These measurements allow for the rapid characterization of new dielectric materials in a device configuration over a wide range of frequencies and temperatures, in order to evaluate thier suitability for device applications.

#### 2:30 PM \*KK8.2

MICROWAVE DIELECTRIC TUNING AND LOSSES IN EPI-TAXIAL LIFT-OFF THIN FILMS OF STRONTIUM TITANATE. Charles T. Rogers, Mark J. Dalberth and John C. Price, Department of Physics, University of Colorado, Boulder, CO.

Nonlinear dielectric materials have a dielectric response that can be changed by an applied electric field. Typical nonlinear dielectrics, such strontium titanate (STO), can show nearly factor of ten changes in the dielectric function for applied electric fields on the order of 1 V/micron. Capacitors constructed from such materials are potentially

useful as tunable elements in a variety of radio frequency and microwave circuits. However, the dielectric loss observed in thin films is a major impediment to the realization thin film devices. We will report results from a study of the dielectric response and loss of STO films. Our measurements cover a frequency range from 10 kHz to 1 GHz, and temperatures from room temperature to 4 K. We find that removing the STO films from the growth substrate, through epitaxial lift-off, causes a significant improvement in dielectric tuning and a reduction in dielectric loss. For lift-off films, we observe dispersion in the real and imaginary parts of the dielectric response that can be fitted by a simple Debye dielectric model, which incorporates a low density of two-state dipolar defects in otherwise bulk-like STO. We will discuss our efforts to understand the nature of these defects and the improvements in device performance that can be realized through the use of epitaxial lift-off.

3:30 PM \*KK8.3 LOCAL OPTICAL PROBES OF DIELECTRIC LOSS IN FERRO-ELECTRIC THIN FILMS. Charles Hubert, Oleg Tikhomirov, Jeremy Levy, Dept of Physics and Astronomy, University of Pittsburgh, Pittsburgh, PA.

Ferroelectric thin films such as  $Ba_xSr_{1-x}TiO_3$  (BST) are attractive materials for frequency-agile microwave electronics due to their nonlinear dielectric properties. However, the dielectric loss in thin films is typically much higher than for corresponding bulk materials. To help understand the physical mechanisms for dielectric loss, we have developed a number of high spatial and temporal resolution optical probes. Confocal scanning optical microscopy (CSOM) is a powerful technique that can be used to image the polarization in ferroelectric thin films with sub-micrometer spatial resolution. Using a Ti:Sapphire laser to generate a synchronous microwave excitation, it is possible to image stroboscopically ferroelectric polarization dynamics. Our measurements on BST films with interdigitated top electrodes show local regions of high dielectric loss at microwave frequencies (2-4 GHz). The loss is directly attributable to the ferroelectric mode. A second technique, apertureless near-field scanning optical microscopy (ANSOM), has been used to image BST films with 30 Angstrom spatial resolution. With ANSOM we are able to investigate the relation between the film nanostructure and dielectric loss. By combining the time and spatial resolution we hope to gain insight into the fundamental processes that govern ferroelectric domain dynamics and dielectric dispersion in these materials. This work is supported by ONR N00173-98-1-G011 and by NSF DMR-9701725.

### 4:00 PM <u>KK8.4</u>

PERFORMANCE OF A HYBRID FERROELECTRIC TUNABLE PRE-SELECT FILTER/LOW NOISE AMPLIFIER. Guru Subramanyam, Dept. of ECE, University of Dayton, Dayton, OH; Felix Miranda, Fred Van Keuls, Robert Roomanofsky, Joe Warner, Carl Mueller, Vernon Heinen, NASA Glenn Research Center, Cleveland, OH.

Due to frequency spectrum utilization demands the use of ferroelectric tunable components may prove to be critical for the next generation of satellite communication systems. A ferroelectric tunable bandpass filter (BPF) has been developed for integration with a low-noise amplifier for a possible gain-compensated down-converter targeted for the next generation of K-band satellite communication system. Electrical tunability in this filter is obtained through the nonlinear electric field dependence of the relative dielectric constant of a ferroelectric thin-film such as strontium titanate (SrTiO<sub>3</sub>) or barium strontium titanate ( $Ba_xSr_{1-x}TiO_3$ ). The BPF has been designed using a conductor (gold or HTS)/ferroelectric/dielectric modified microstrip structure for a center frequency of 19 GHz, a bandwidth of 150 MHz, and for a minimum frequency tunability of 10%. Experimental results show that the BPFs are tunable by more than 10%, with a bipolar biasing scheme employed. Within a specific range of applied dc bias, one can tune the filter's parameters such that they meet or exceed the minimum specifications. Such a BPF is integrated with a low-noise amplifier (LNA) to study the effect of tuning on the hybrid circuit's performance especially on the amplifier's noise-figure and the gain. The performance of such a hybrid circuit will be

# 4:15 PM KK8.5

IMAGING OF DIELECTRIC CONSTANT OF TUNABLE DIELECTRIC (Ba,Sa)TiO<sub>3</sub> THIN FILM USING SCANNING MICROWAVE MICRÓSCOPE. S. Hyun, J.H. Lee, J. Kim, D.S. Kim and K. Char, Department of Physics, Seoul National University, Seoul, KORÉA; S.J. Park, J. Sok, and E.H. Lee, Microelectronics Lab, Materials and Device Sector Samsung Advanced Institute of Technology, Suwon, KOREA.

We developed a scanning microwave microscope (SMM)[1] which operates at rf/microwave frequency range to investigate the dielectric properties of the electrically tunable thin films. The real parts and the imaginary parts of permittivity could be obtained by investigating the shifts of resonant frequency and the change of quality factor, respectively, of a quarter wavelength coaxial resonator in near-field regime[2]. After deposition of the contact electrodes, the pulsed laser deposited  $(Ba,Sr)TiO_3$  thin films were imaged at various applied DC biases. By imaging the (Ba,Sr)TiO3 thin films as a function of the applied bias, we can observe the change of dielectric constant and loss. We will discuss the mechanism of the dielectric loss in electrically tunable thin films as well as the relation between this changes and applied bias. We also observed the increase of dielectric loss at the boundary of patterned  $(Ba,Sr)TiO_3$ , which indicated existence of damage of (Ba,Sr)TiO<sub>3</sub> films by the etching process. In addition, we will also report on our use of the scanning microwave microscope in investigating the various materials such as high temperature superconductor, low dielectric materials, ferroelectric materials and so on. [1] T. Wei, X.-D. Xiang, W.G. Wallace-Freedman, and P.G Schultz, Appl. Phys. Lett. 68, 3506 (1996). [2] C. Gao and X.-D. Xiang, Rev. Sci. Instrum. 68, 3846 (1998).

IMAGING OF MICROWAVE PERMITTIVITY AND TUNABILITY IN NONLINEAR DIELECTRIC THIN FILMS. D.E. Steinhauer, F.C. Wellstood, Steven M. Anlage, Univ of Maryland, Center for Superconductivity Research, Dept of Physics, College Park, MD; C Canedy, R. Ramesh, Univ of Maryland, Dept of Materials and Nuclear Engineering, College Park, MD.

We will describe the use of a near-field scanning microwave microscope  $^{1-4}$  to quantitatively image the dielectric constant and tunability of nonlinear dielectric thin-film samples with a spatial resolution of about 1  $\mu$ m. The microscope consists of a coaxial resonator terminated at one end by an open-ended coaxial probe with a sharp, protruding tip which is held in gentle contact with the sample. By monitoring a selected resonant frequency of the microscope resonator, we can quantitatively extract the local dielectric constant as well as nonlinear dielectric terms. This technique is useful for evaluating the dielectric properties and homogeneity of thin film samples at microwave frequencies (0.1 - 50 GHz), and pinpointing regions of low dielectric constant or tunability. We will present quantitative images and local hysteresis loops of barium strontium titanate thin films, as well as a comparison between our microwave data and the material microstructure using other scanning techniques. Steven M. Anlage et al., IEEE Trans. Appl. Supercond., June, 1999.

D.E. Steinhauer et al., Appl. Phys. Lett., 72, 861 (1998). See http://www.csr.umd.edu/research/hifreq/micr\_microscopy.html

<sup>4</sup> This work has been supported by the Maryland/NSF MRSEC and the Center for Superconductivity Research.

#### 4:45 PM KK8.7

NEAR-FIELD IMAGING OF THE MICROWAVE DIELECTRIC PROPERTIES OF PbTiO<sub>3</sub> CRYSTALS AND  $Sr_{1-x}Ba_xTiO_3$  FILMS. Yu Guo Wang, Mark E. Reeves, Department of Physics, The George Washington University, Washington, DC; Wontae Chang, James S. Horwitz, Wonjeong Kim, Naval Research Laboratory, Washington,

A near-field scanning microwave microscope is used to image the variations of dielectric properties at the submicron range in insulating thin films and single crystals. We will report periodical profiles of dielectric susceptibility and loss due to alternating a- and c-axis domains in PbTiO3 crystals. Defects originated from the flux-growth process are detected with lower dielectric susceptibilty and higher loss. In  $Sr_{1-x}Ba_xTiO_3$  films, we observe local variations of dielectric properties. These are correlated with structural and morphological studies. This work is supported by DARPA.