SYMPOSIUM O

Complex Oxide Heteroepitaxy

November 26 – 28, 2001

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* Invited paper

SESSION O1: CRYSTALLINE OXIDES ON SEMICONDUCTORS
Chairs: Charles H. Ahn and Philippe Ghosez
Monday Morning, November 26, 2001
Room 203 (Hynes)

8:30 AM O1.1
EPITAXIAL GROWTH AND MICROSTRUCTURE CHARACTERIZATION OF $SrRuO_3$ FILMS DEPOSITED ON SILICON WITH SrO BUFFER LAYER. Y.X. Chen, J. Koike, Tohoku Univ, Dept of Materials Science, Sendai, JAPAN; T. Higuchi, S. Iwashita, M. Ishida, T. Shimoda, SEIKO EPSON Co, Base Technology Research Center, Nagano-ken, JAPAN.

Among various conductive oxide thin films, $SrRuO_3$ exhibits excellent conductivity and has a pseudo-cubic perovskite structure, which makes $SrRuO_3$ one of the most favorable candidates as a bottom electrode layer as well as an epitaxial seed layer for ferro- or piezo-electric devices. For a better device performance, it is desirable to grow a single crystal $SrRuO_3$ layer on a (001) Si substrate, using a buffer layer. Our aim is to grow an epitaxial $SrRuO_3$ layer on a (001) Si substrate using SrO as a buffer layer by pulsed laser deposition, and to characterize the microstructure and crystallographic orientation relationship among the constituting layers using X-ray diffraction, transmission electron microscopy, high-resolution electron microscopy and energy dispersive X-ray spectroscopy. SrO buffer layer was epitaxially grown on the silicon (001) surface in spite of the presence of an amorphous silicon oxide layer. Subsequently, $SrRuO_3$ film was epitaxially grown on the SrO surface. Three types of domains having surface normal directions of [110], [1$\bar{1}$0] and [001] were observed in $SrRuO_3$ crystalline. As a result, three kinds of crystallographic orientation relationships among Si, SrO and $SrRuO_3$ were observed. Stacking faults and twins were formed along the orthorhombic {202} lattice planes of $SrRuO_3$. The formation mechanisms of these domains and planner defects, and their effects on the subsequent growth of ferroelectric films will be discussed.

8:45 AM O1.2
TUNGSTEN OXIDE CHEMIRESISITIVE FILMS INTEGRATED ONTO SILICON BY USE OF AN EPITAXIAL BaF_2 BUFFER LAYER. L. Doucette, L.J. LeGore, R.J. Lad, Laboratory for Surface Science & Technology, University of Maine, Orono, ME; F. Santiago, S.L. Moran, Naval Surface Warfare Center, Dahlgren Division, Dahlgren, VA.

Chemical sensors based on semiconducting metal oxides, such as WO_3, require an insulating substrate so that conductivity changes in the oxide caused by gas interactions with the oxide surface can be measured. Our previous work has shown that heteroepitaxial WO_3 films can be grown on sapphire substrates. In this work, we have grown and characterized WO_3 films on BaF_2/Si substrates. The epitaxial insulating BaF_2 layer allows integration of miniaturized WO_3 gas sensors and associated control electronics directly onto the same chip using Si-based microfabrication. The 30 nm thick epitaxial BaF_2 buffer layers were grown on Si (100) by mbe and exhibit four equivalent (111) orientations relative to the Si substrate as characterized by RHEED, LEED, and XRD pole figures. WO_3 films were grown on the BaF_2/Si substrates using low rate rf magnetron sputtering of a W target in an Ar/O_2 plasma over the temperature range 30 ? 650°C. The as-deposited films exhibit microstructures ranging from random polycrystalline to highly oriented depending on the exact deposition conditions. Post-deposition annealing in ambient air above 400°C leads to oxidation at the WO_3/BaF_2 interface. This result is significant since the chemiresistive sensors are routinely operated for long times between 100 and 450°C. XPS analysis in conjunction with ion beam depth profiling suggests the formation of BaO at the interface when the WO_3 film is polycrystalline. However, highly oriented WO_3 films appear to result in a more stable interface. STM and AFM imaging reveals that the surface roughness of the WO_3 films also correlates with deposition conditions and film microstructure.

9:00 AM O1.3
EPITAXIAL GROWTH AND PROPERTIES OF CeO_2 ON (001) InP. Mat Ivill, David Norton, Mitesh Patel, Kyunghoon Kim, Hyung-Jin Bae, and Stephen Pearton, University of Florida, Department of Materials Science and Engineering, Gainesville, FL; John Budai, Hans Christen and Matthew Chisholm, Oak Ridge National Laboratory, Oak Ridge, TN.

In recent years, there has emerged significant interest in the integration of electronic oxides with semiconductor materials. This is true for III-V compound semiconductors, such as GaAs and InP, where the functionality of oxides could prove enabling in the development of integrated photonic or microwave systems. In addition, the formation of epitaxial oxides on III-Vs would be useful in exploring MOSFET devices. The epitaxial growth of oxides on InP is particularly challenging due to the volatility of phosphorus from the substrate surface. In this talk, we will report on the successful growth of epitaxial CeO_2 on InP using hydrogen-assisted physical vapor deposition. Both pulsed-laser deposition and reactive sputter deposition have been used to achieve (001) CeO_2 on (001) InP. Epitaxy is achieved in the presence of molecular hydrogen that is introduced during nucleation to reduce native In_2O_3 from the InP surface. X-ray diffraction along the surface normal shows that the films are (001) oriented. In-plane four-circle X-ray diffraction scans confirm a cube-on-cube epitaxial relationship between the oxide film and the InP substrate. Rapid heating of the substrate to the deposition temperature proved important in order to avoid significant decomposition of the InP surface prior to film growth. We will describe the structure, morphology, and interface properties. The growth of CeO_2 on InP should enable the integration of electronic oxide functionality with InP-based semiconductor technologies, and may also provide a means to explore InP MOSFET structures.

9:15 AM O1.4
CRITICAL ISSUES IN THE HETEROEPITAXIAL GROWTH OF COMPLEX OXIDES ON SILICON. J. Lettieri, J.H. Haeni, and D.G. Schlom, Penn State University, Dept of Materials Science and Engineering, University Park, PA.

The heteroepitaxial growth of oxides on silicon presents significant opportunities to harness the versatile superconducting, dielectric, pyroelectric, piezoelectric, and ferroelectric properties of multicomponent oxides while simultaneously exploiting the properties of the underlying semiconductor. Growth of epitaxial, crystalline structures in which the properties of the underlying silicon and overlying film both attain their full potential, however, is wrought with significant complications concerning compound stability and delicate oxidation considerations. These fundamental issues and resultant growth strategies have begun to be addressed with much of the recent work investigating the growth of $SrTiO_3$ on silicon. In this study, we describe the adaptation and development of these techniques to the heteroepitaxial growth of various multicomponent oxide systems on silicon.

9:30 AM O1.5
SILICIDE HETEROEPITAXY AND THE STABILITY OF THE CRYSTALLINE OXIDE/SEMICONDUCTOR INTERFACE. F.J. Walker, R.A. McKee and G.M. Stocks Oak Ridge National Laboratory.

We have considered the submonolayer reaction of alkaline earth metals with silicon and germanium and contrast the thermochemical reactions of these metals with that of the transition metals, oxygen and silicon as they influence the heteroepitaxial transition from a semiconductor surface to an overgrowing crystalline oxide. The thermodynamics and kinetics play an intricate balance in the practical realization of single crystal epitaxy. We present RHEED and XPS data against 1st principles calculations to demonstrate a singular path towards epitaxy on (001) Si and Ge. The physical and the electrical structure of the crystalline oxide/semiconductor interface depend critically on silicide heteroepitaxy at the monolayer level in these systems. We discuss these structures in terms of the valence and conduction band structure relating the fundamentals of three-component thermodynamic equilibrium to crystalline oxide heteroepitaxy on semiconductors.

9:45 AM O1.6
HfO_2 AND ZrO_2: COMPARISON OF STRUCTURES AND THERMODYNAMIC AND ELECTRONIC PROPERTIES BASED ON AB INITIO CALCULATIONS AND EXPERIMENT. Alexander A. Demkov, Physical Sciences Research Labs, Motorola, Inc., Tempe, AZ; and Alexandra Navrotsky, Thermochemistry Facility, Department of Chemical Engineering and Materials Sciences, University of California at Davis, Davis, CA.

The International Technology Roadmap for Semiconductors (ITRS) predicts that the strategy of scaling complementary metal-oxide-semiconductor (CMOS) devices will come to an abrupt end around the year 2012. The main reason for this will be the unacceptably high leakage current through the silicon dioxide gate with a thickness below 20 Å. Finding a gate insulator alternative to SiO_2 has proven to be far from trivial. Hafnium and zirconium dioxides and silicates have been recently considered as gate dielectrics with intermediate dielectric constants. Hafnia and ziconia are important ceramic materials as well, and their phase relations are rather well studied. There is also interest in hafnia as a constituent of ceramic waste forms for plutonium, based on its refractory nature and high neutron absorption cross section. We use a combination of the ab-initio calculations and calorimetry to investigate thermodynamic and electronic properties of hafnia and zirconia. We describe the cubic to tetragonal phase transition in the fluorite structure by computing the total energy surface for zone-edge

distortions correct to fourth order in the soft-mode displacement with the strain coupling renormalization included. We compare the two materials using some simple chemical concepts.

10:30 AM *O1.7
SILICIDE AND OXIDE HETEROEPITAXY ON SILICON - WHAT WE KNOW AND WHAT WE THINK. Rodney McKee, Oak Ridge National Laboratory, Oak Ridge, TN.

The complex issues of oxide heteroepitaxy on silicon are of paramount consideration as we attempt to formulate a comprehensive understanding of interface physics for crystalline oxides on semiconductors (COS). We have previously identified a "pathway" for layer-by-layer stabilization of a perfectly crystalline, commensurate interface between alkaline earth oxides and silicon. Here we will attempt to show how submonolayer silicide structures formed on silicon affect physical structure transitions, charge transfer processes, and the specifics of silicon-in-silicide bonding to oxide overlayers. These data ultimately relate to the fundamentals of Schottky barrier formation and the atomistic understanding of MOS capacitor physics in this new physical system of crystalline oxides on semiconductors. We will discuss our view of the implications for COS and these data for new functionality for oxide dielectrics on semiconductors.

11:00 AM *O1.8
CHALLENGES AND OPPORTUNITIES FOR EPITAXIAL OXIDES ON SILICON. R. Droopad, J. Ramdani, Z. Yu, L. Hilt, A. Demkov, C. Overgaard, J. Edwards, Y. Wei, J. Curless, K. Eisenbeiser, B. Ooms, Motorola Labs, Physical Sciences Research Laboratories, Tempe, AZ.

One of the main problems facing the semiconductor industry to the continuing reduction in the size of Si CMOS devices, is the scaling of the SiO_2 (high-k) gate dielectric. Presently SiO_2 is being used, but at thickness below 20 Å, it suffers from high tunneling leakage current and reliability problems. Alternative high-k materials to replace SiO_2 need to be developed as soon as possible. The alkaline earth oxides such as barium strontium titanate ($BaxSr_{1-x}TiO_3$) have a substantially higher dielectric constant than SiO2 and are ideal candidates for gate dielectrics. Because of the higher dielectric constant a physically thicker layer can yield an equivalent oxide thickness of $<$ 20 Å, thereby eliminating the leakage problems experienced with ultra-thin SiO_2. These oxides also exhibit ferroelectric behavior and their use as the gate dielectric on Si can be exploited in the realization of a single transistor memory element. In this presentation we will be discussing the challenges of integrating epitaxial oxides into semiconductor products, and ways to overcome some of these challenges. We will also discuss a number of other applications for epitaxial oxides, in addition to that of gate dielectrics.

11:30 AM *O1.9
EPITAXIAL GROWTH OF $Pb(Zr_{0.2}Ti_{0.8})O_3$ ON Si AND ITS NANOSCALE PIEZOELECTRIC PROPERTIES. A. Lin, X. Hong, V. Wood, A.A. Verevkin, and C.H. Ahn, Yale University, Department of Applied Physics, New Haven, CT; R.A. McKee, F.J. Walker, and E.D. Specht, Oak Ridge National Laboratory, Oak Ridge, TN.

Complex oxide materials are valued for their broad diversity of behavior, such as ferroelectricity, piezoelectricity, magnetism, and superconductivity. The ability to integrate these materials with mainstream Si processing opens several opportunities, including applications in microelectromechanical systems (MEMS) and microelectronics. Here, we consider the growth of ferroelectric and piezoelectric materials on Si. In particular, we discuss the epitaxial deposition of $Pb(Zr_{0.2}Ti_{0.8})O_3$ on (001) Si using an insulating, single-crystalline $SrTiO_3$ transition layer.[1] These structures have been grown by a combination of molecular beam epitaxy (using the crystalline-oxides-on-silicon technique developed by McKee and co-workers) and off-axis magnetron sputtering.[2] The surfaces of these films have a root-mean-square (RMS) roughness of $< 5\AA$, and piezoelectric microscopy measurements reveal a uniform piezoelectric response down to nanoscale levels, with a piezoelectric coefficient of $\sim$50 pm/V. The domain configuration of these structures is switchable down to sub-100-nm dimensions. We attribute the ability to write small, controlled nanofeatures to the epitaxial, smooth surfaces of these films. [1]A. Lin, X. Hong, V. Wood, A.A. Verevkin, C.H. Ahn, R.A. McKee, F.J. Walker, and E.D. Specht, Appl. Phys. Lett. **78**, 2036 (2001) [2]R.A. McKee, F.J. Walker, and M.F. Chisholm, Phys. Rev. Lett. **81**, 3014 (1998).

SESSION O2: OXIDE DEVICE ISSUES
Chairs: James N. Eckstein and Dave H.A. Blank
Monday Afternoon, November 26, 2001
Room 203 (Hynes)

1:30 PM *O2.1
EXCHANGE EFFECTS IN MAGNETIC OXIDE HETERO-STRUCTURES. A. M. Goldman, School of Physics and Astronomy, University of Minnesota, Minneapolis, MN.

We have studied exchange interactions in a variety of perovskite manganite heterostructures, including spin valve configurations, ferromagnet/paramagnet metallic superlattices, and ferromagnet/ antiferromagnet multilayers. These heterostructures were fabricated employing various combinations of the metallic ferromagnetic perovskite manganites, $La_{2/3}Ba_{1/3}MnO_3$ or $La_{23}Sr_{1/3}\%MnO_3$, the paramagnetic metal, $LaNiO_3$, and the insulating antiferromagnet, $La_{1/3}Ca_{2/3}MnO_3$. Heterostructures containing these epitaxially compatible compounds were grown by ozone-assisted molecular beam epitaxy on $SrTiO_3$ substrates using he block-by-block technique. Film growth was monitored *in situ* using reflection high energy electron diffraction. Structures were characterized using X-ray diffraction analysis. Oscillation of the indirect exchange coupling between magnetic layers as a function of metal spacer layer thickness was observed, and explained within the context of the Ruderman-Kittel-Kasuya-Yosida (RKKY) model employing an ab initio calculated band structure for $LaNiO_3$. The electrical resistance of metallic manganite layers in both multilayers and superlattices, appears to be determined by the magnitude of the vector sum of the effective exchange field and the external magnetic field.
This work was performed in collaboration with K.R. Nikolaev, I.N. Krivorotov, A. Yu Dobin, R. Wentzkovitch, and E.D. Dahlberg, and was supported in part by the University of Minnesota Materials Research Science and Engineering Center under Grant NSF/DMR-9809364.

2:00 PM *O2.2
MATERIALS ASPECTS OF DEVICES CONCEPTS BASED ON LAYERED OXIDES. Dave H.A. Blank, MESA Research Inst, Univ of Twente, Enschede, NETHERLANDS.

With the possibility to artificially create new materials, e.g. using block-by-block or layer-by-layer growth procedures, new device concepts become possible. Examples are field effect transistors, based on metal-insulators for practical applications, such as in active circuitry or novel memory storage-cells. In addition in the field of spin valve transistors the layered oxides will open new possibilities. Further, the oxygen permeability in most perovskites has already shown its applicability in fuel cells and oxygen separation and thin film membranes becomes available for practical use. In this contribution the challenges in all-oxide layered structures will be discussed. Attention will be paid to different device concepts based on layered oxide materials and the difficulties that have to be overcome, especially related to thin film growth.

2:30 PM *O2.3
NANOSCALE METAL OXIDE HETEROSTRUCTURES FOR CHEMICAL SENSORS. Xiaoqing Pan, Juan Dominguez, Wonwoo Kim, and Haiping Sun, Department of Materials Science & Engineering, The University of Michigan, Ann Arbor, MI.

A tunable, selective gas sensor has been a goal for a large variety of important environmental and industrial applications. Current sensor research trends have focused on arrays of complex materials with well-characterized adsorption properties, where temperature is the primary adjustable variable. In this work, we have investigated a new concept for selective gas sensors based on nanoscale epitaxial metal oxide heterostructures based on SnO_2 and TiO_2. High-quality epitaxial thin films of individual SnO_2 and TiO_2 as well as their heterostructures were grown on the $(10\bar{1}2)$ surface of sapphire substrate. We have systematically characterized the electrical transport properties of individual thin films of single layers and heterostructures as a function of chemical doping, film thickness, and temperature, and studied the chemical sensing properties of these films. Furthermore, we have fabricated simple model systems based on epitaxial single crystal films consisting of field effect transistor (FET) structure and studied the effect of bias applied across the p-n junction on the electrical and selective adsorption properties. The relationships between the film microstructures and chemical sensing performances will be discussed.

3:30 PM O2.4
EPITAXY AND OPTICAL PROPERTIES OF A_2CuO_3 (A: Sr, Ca) THIN FILMS WITH LARGE OPTICAL NONLINEARITY.
T. Manako[a], Y. Okimoto[b], M. Izumi[c], S. Shinomori[c], H. Kishida[c], H. Okamoto[c,f], T. Fukumura[d], M. Ohtani[e], M. Kawasaki[b,d,f], and Y. Tokura[b,c,f]; [a]NEC Corp., [b]Joint Research Center for Atom Technology (JRCAT), [c]University of Tokyo, [d]Tohoku University, [e]Tokyo Institute of Technology, [f]Correlated Electron Research Center (CERC).

Thin films of Sr_2CuO_3 (SCO) and Ca_2CuO_3 (CCO) were epitaxially grown using a pulsed laser deposition technique. The one-dimensional Cu-O chains responsible for the large optical nonlinearity of these

cuprates were aligned in one direction by using a $LaSrAlO_4$ (LSAO) (1 0 0) substrate and a proper buffer layer.
Owing to poor lattice matching between the film and substrate, the SCO films directly grown on substrates inevitably included a small portion of misoriented domains in which the Cu-O chains were aligned perpendicular to the film surface. CCO films, however, were easily grown in a single orientation with the Cu-O chains lying in the film plane. We found that single-oriented SCO film with high optical quality can be fabricated using a Sr_2TiO_4 buffer layer. This technique will be a key to fabricating ultra-fast all-optical switches with a waveguide structure.

3:45 PM O2.5
SCANNING TUNNELING MICROSCOPY STUDY OF POLARIZATION PHENOMENA IN EPITAXIAL FERROELECTRIC-BASED HETEROSTRUCTURES. O. Kuffer, I. Maggio-Aprile, O. Fischer, University of Geneva, Department of Condensed Matter Physics, Geneva, SWITZERLAND.

A scanning tunneling microscope (STM) was used to investigate ferroelectric polarization effects in all-perovskite field effect devices. The samples grown by off-axis radio-frequency magnetron sputtering were composed of a single crystalline ferroelectric film of $Pb(Zr_{0.2}Ti_{0.8})O_3$ (PZT) deposited on a conducting gate electrode substrate and covered with an ultra-thin channel of $La_{0.67}Ca_{0.33}MnO_3$ (LCMO), or with an ultra-thin highly resistive channel of $Sr(Ru_{0.37}Ti_{0.63})O_3$ (SRTO). Firstly, scanning tunneling potentiometry (STP) studies were used to detect on a local scale the ferroelectric field effect in the LCMO/PZT devices. The STP technique allowed to map the local electronic transport in the LCMO channel and to detect the local resistance variations induced by the ferroelectric polarization. Secondly, we performed scanning tunneling spectroscopy (STS) studies on the SRTO/PZT devices. The SRTO current-voltage (IV) characteristics as a function of the two PZT polarization states evidenced a transition from semiconducting to metallic behavior. Taking advantage of this phenomenon, we demonstrated the ability to use the STM to write single ferroelectric domains using pulses. The reading of the domains was performed by current imaging tunneling spectroscopy, i.e. by recording locally the current contrast at a suitable bias voltage. Using the STM/STS technique described here, we were able to write nanoscopic domains in the SRTO/PZT field effect devices with a size reaching 20nm.

4:00 PM O2.6
THEORETICAL STUDY OF STEPS ON THE $SrTiO_3$ (001) SURFACE. Xiaodong Zhang and A.A. Demkov Physical Sciences Research Labs, Motorola, Inc., Tempe, AZ.

Epitaxial perovskite oxides such as $SrTiO_3$(STO) have been suggested as alternative gate dielectrics in CMOS technology. When integrated with Si they also have interesting ferroelectric memory cell applications. Surfaces of STO are important during the crystal growth, in particular, stepped surfaces are important due to the high chemical activity of the step edges. Two possible terminations of the (001) STO surface result in SrO and TiO_2 terraces that could alternate being separated by steps of the half unit cell height. Alternatively, steps full unit cell high could separate terraces of the same termination. To further our understanding of the stepped STO surface and its role in the crystal growth we perform first principle simulations of the stepped (001) STO surface. Here we report on the half unit cell high steps. We first calculate surface energies of TiO_2 and SrO terminated STO surface. We find SrO terminated surface to have a lower free energy under most experimental conditions with the exception of the Ti-rich regime. We use the cleavage energy to calculate the step energetics. There are several possible terrace terminations resulting in different step edges. We find the step edge having both Sr and O atoms as the most favorable energetically. Furthermore, we find it is more energetically favorable to cover TiO_2 terminated surface by SrO and thus make an SrO terminated surface with the full unit cell high steps. This result is consistent with our prediction of the SrO terminated surface having the lower surface energy. Our results agree with the recent experimental study of the STO surface morphology, which shows the chemical treatment of STO surface results in single terminated surface [1]. We investigate the charge density and frontier orbitals for the STO surface with steps half unit cell high. We find that HOMO and LUMO states of a stepped surface are spatially separated by the step edge. We have calculated STM images of the stepped surface, our results indicate that half unit cell high steps may be hard to observe, and a careful correlation of the images obtained under opposite polarities is needed. 1. G. Koster, G. Rijnders, D.H.A. Bank, and H. Rogalla, Physics C 339, 215 (2000).

4:15 PM O2.7
FABRICATION OF AN OXIDE SUBSTRATE WITH A TUNABLE LATTICE CONSTANT. K. Terai, M. Lippmaa, T. Fujii, H. Koinuma, Department of Innovative and Engineered Materials, Tokyo Institute of Technology, Yokohama, JAPAN; P. Ahmet, T. Chikyo, National Institute for Materials Science, Tsukuba, JAPAN; M. Kawasaki, Institute for Materials Research, Tohoku Univ., Sendai, JAPAN.

The physical properties of oxide thin films can change dramatically due to in-plane strain imposed by the substrate crystal. Our aim is to fabricate a substrate with a tunable lattice constant, which could be used for growing strain-free films of materials for which lattice-matched substrates are not available. We start by growing a $BaTiO_3$ buffer layer on $SrTiO_3$ by pulsed laser deposition at a temperature of 650°C, followed by annealing at 1100°C or higher. The surface of this film is atomically smooth and has a clear step-and-terrace structure. Reciprocal-space mapping and high-resolution transmission electron microscopy (TEM) have shown that less than 200$\AA$-thick buffer layers are fully strained immediately after growth. A large number of dislocations are present in the films. The annealing treatment allows the $BaTiO_3$ buffer layer to relax fully, forming misfit dislocations at the $SrTiO_3/BaTiO_3$ interface. The buffer layer itself shows no dislocations in TEM images. This technique makes it possible to fabricate a stable surface with an in-plane lattice constant of 3.99$\AA$. The lattice constant can be tuned by depositing a thick layer of $Ba_{1-x}Sr_xTiO_3$ (BST) on the $BaTiO_3$ buffer before final in-situ annealing. As an example, we show for the case of $x = 0.5$ that the resulting BST surface is fully relaxed and cubic with a lattice constant of 3.95$\AA$. The slightly bunched step-and-terrace surface is an excellent starting point for further film growth. Any desired in-plane lattice constant between $SrTiO_3$ (a=3.905$\AA$) and $BaTiO_3$ (a=3.994$\AA$), covered by the BST system, can be prepared in this way.

4:30 PM O2.8
EPITAXIAL THICK FILM HETEROSTRUCTURES OF $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ RELAXOR FERROELECTRIC FILMS FOR HIGH FREQUECY MEDICAL TRANSDUCERS. S.D. Bu, D.M. Kim, C.B. Eom, Department of Materials Science and Engineering, University of Wisconsin-Madison, Madison, WI; J. Lettieri, D.G. Schlom, Department of Materials Science and Engineering, The Pennsylvania State University, University Park, PA; S.K. Streiffer, Materials Science Division, Argonne National Laboratory, Argonne, IL.

Single crystal relaxor ferroelectrics, such as $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PMN-PT) and $Pb(Zn_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PZN-PT), yield significantly higher electromechanical coupling coefficient than conventional polycrystalline ferroelectrics. A major challenge is to fabricate epitaxial PMN-PT thick films between epitaxial metallic oxides, and integrate them into microelectromechanical systems such as high frequency medical trasnsducers. We have grown 5-15 μm thick epitaxial PMN-PT films on both miscut (001) $SrTiO_3$ substrates and $SrRuO_3$ single crystal thin films of metallic oxide bottom electrode by magnetron sputtering. We found that the miscut angle of the $SrTiO_3$ substrate plays an important role in the growth of PMN-PT films with good crystalline quality. The effect of substrate temperature was studied by XRD on the growth of PMN-PT films, its relation with the second phase will be discussed. It also will be discussed the correlation with second phase and electrical properties such as piezoelectric coefficient and remnant polarization.

SESSION O3: POSTER SESSION
Chair: Harold Y. Hwang
Monday Evening, November 26, 2001
8:00 PM
Exhibition Hall D (Hynes)

O3.1
COMPLETE CONTROLS OF SUBSTRATE NANOSCALE SURFACE ROUGHNESS AND A-C ORIENTED YBCO THIN FILM GROWTH. Tamio Endo, Hideaki Kohmoto, Shin-ichi Iwasaki, Rajendra Kale, Tao Li, Mie Univ, Faculty of Engineering, Tsu, Mie, JAPAN.

Mechanism of a-axis and c-axis oriented YBCO thin film growth on MgO substrate is studied systematically employing IBS technique with supply of oxygen molecules either plasma. A ratio of c-oriented (a-oriented) phase increases (decreases) in an a-c mixed film with increasing substrate temperature (Ts). The a-phase ratio (Ra) increases with increasing oxygen partial pressure (Po). The crystal growth of a-phase is enhanced and improved by the plasma supply at low Ts, it can be grown at 450°C. "Surface migration" of deposited particles is promoted by higher thermal energy with increasing Ts, then the c-phase ratio (Rc) increases. With increasing Po, sputtered particle kinetic energy is more reduced by "collisions" with higher density of supplied oxygen species, then particle-energy assisted "surface migration" is retarded. This results in the increase in Ra. Wrong-sited Y and Ba atoms can be rearranged into higher order by the plasma energy on a growing surface of the a-phase at the low Ts. It causes "reduction of surface energy", then the a-phase growth is

enhanced and its crystallinity is improved by the plasma. To prove the proposal of "surface migration", surface roughness of MgO is controlled in nano-scale by irradiation of oxygen plasma before the deposition, then the film is in-situ deposited on it at Ts=600°C. With increasing plasma irradiation time, rms roughness (Sr) of the substrate surface increases and Ra (Rc) increases (decreases) from zero to 100%. With increasing the plasma intensity by raising plasma discharge current, Sr once decreases (smoothening) and then increases (roughening), and correspondingly once Rc increases to 100% and then Ra increases instead. Thus, the "surface migration" is certainly the crucial factor for the a-c oriented growth. We can produce a dot or line-shaped fine a-phase region in a matrix c-phase plane of YBCO thin film by irradiation patterning.

O3.2
ROOM-TEMPERATURE FABRICATION AND PHOTOVOLTAIC PROPERTY OF ALL EPITAXIAL MIS STRUCTURE OF TRANSPARENT CONDUCTING OXIDE/CeO_2/Si(111). J. Tashiro, A. Sasaki, K. Nakajima, S. Akiba, S. Satoh, M. Yoshimoto, Tokyo Inst. of Tech., Yokohama, JAPAN; P. Ahmet, T. Chikyow, Nanomaterial Lab.-COMET, Tsukuba, JAPAN.

Heteroepitaxial growth of insulating thin films on silicon substrates has attracted much interest in application such as silicon-on-insulator (SOI), gate insulator in LSI and the buffer layer in oxide electronics. We have reported the successful room-temperature heteroepitaxy of CeO_2 which has excellent lattice match with silicon onto Si(111) substrates using pulsed laser molecular beam epitaxy method (laser MBE) (Jpn. J. Appl. Phys., vol. 34 (1995) L688). Here we report the room-temperature fabrication of all epitaxial MIS structure of transparent conducting oxides (TCO)/CeO_2/Si(111) for application to solar cells. Conventional MIS-type solar cells utilize ultra-thin amorphous SiO_x insulating layer on silicon, but we introduce the epitaxial insulating layer to promote crystallization of TCO layer. The ultrathin CeO_2 film was deposited on the hydrogen-terminated Si(111) substrate at 20C. Reflection high energy electron diffraction (RHEED) pattern indicated the epitaxial growth. Then the 75nm-thick film of Al-doped ZnO was grown on CeO_2(111)/Si(111) at 20C. Epitaxial growth of ZnO layer was verified by RHEED. Sn-doped In_2O_3 (ITO) was also employed in the MIS structure. The photovoltaic property of the present MIS structured films was examined, showing the applicability to solar cells.

O3.3
FIRST PRINCIPLES CALCULATIONS OF DEFECT-INTERFACE INTERACTIONS IN STRONTIUM TITANATE. R. Astala and P.D. Bristowe, Department of Materials Science and Metallurgy, University of Cambridge, UNITED KINGDOM.

We have studied the properties of a $\Sigma = 3$ (111) grain boundary in strontium titanate using DFT plane-wave pseudopotential methods. The effect of heavy oxygen deficit in the grain boundary core and in the nearby bulklike region is examined. We have estimated the binding energy for the O vacancy and studied the atomic geometry and electron-level properties of the structures. Generally the incorporation of the vacancy resulted in only minor relaxations in the atomic geometry. In addition, we have extended our electronic structure calculations to heavy Nb doping of Ti sites in the grain boundary structure.

O3.4
STRUCTURE OF MgO/ZnO HETERO-JUNCTION WHISKERS. Hidetoshi Saitoh, Yusuke Okada, Shigeo Ohshio, Nagaoka Univ. Tech., Kamitomioka, Nagaoka, Niigata, JAPAN.

A chemical vapor deposition (CVD) technique operated under atmospheric pressure is available to synthesize various forms of metal-oxide crystals such as ZnO whiskers. In this study, growth of sodium chloride structure of MgO whiskers was attempted to confirm growth of the whisker with cubic system structure. MgO whiskers grew epitaxially on a single crystalline substrate of (0001) sapphire. The growth of the MgO whisker was strongly dependent on the growth temperature and the vapor rate of the raw material. The MgO whisker was also formed on the ZnO whisker, resulting the unique whiskers with a MgO/ZnO hetero-junction.

O3.5
NITROGEN DOPED ZnO FILMS DEPOSITED BY FILTERED CATHODIC VACUUM ARC. X.L. Xu, S.P. Lau, C.C. Seng, C.Y. Ng, J.S. Chen, B.K. Tay, Ion Beam Processing Lab, School of Electrical and Electronic Engineering, Nanyang Technological University, SINGAPORE.

ZnO film has caught much attention for potential applications in optoelectronic because of its high exciton energy (60mV) at room temperature. ZnO films can be doped with Al, In, Ga and N to obtain conductive transparency electrode and doped semiconductors. In this paper, we deposited nitrogen doped ZnO films by filtered cathodic vacuum arc technique. The films are preferably oriented to the (103) direction, and the XRD peak at (103) becomes broaden and shift slightly to the lower diffraction angle with increasing nitrogen flow rate. Surface morphology of the films was characterized by SEM and AFM. All the films displayed a wedgelike textured surface and the films roughness decrease with increasing nitrogen incorporation. The resistivity of the films increased from 5.86×10^{-3} to 9.96 Ωcm with increasing nitrogen to 2 sccm and slightly decreased with further increase nitrogen gas. The Raman peak at around 580 cm^{-1} is assigned to longitudinal optical phonon, which is not only influenced by crystal orientation and propagation directions but also the electro-optical coefficient and impurity, was enhanced with increasing nitrogen content. The composition and transmittance of films were investigated by XPS and optical absorption, respectively.

O3.6
STRUCTURE AND TRANSPORT PROPERTIES FOR $YBa_2Cu_3O_{7-\delta}$-$La_{2/3}Ca_{1/3}MnO_3$ BILAYERS. X.S. Wu[a,b], T.L. Kam[a], W.S. Tan[b], W.H. Tang[a], J. Gao[a]; [a]Department Of Physics, The University of Hong Kong, HONG KONG; [b]National Laboratory of Solid State Microstructures, Center for Advanced Studies in Science and Technology of Microstructures, Department of Physics, Nanjing University, Nanjing, CHINA.

The $La_{2/3}Ca_{1/3}MnO_3$ (LCMO)/$YBa_2Cu_3O_{7-\delta}$ (YBCO) heterostructural bilayers are prepared by dc magnetic sputtering technique on $SrTiO_3$ (STO) substrate. The structures, including the surfacial and interfacial roughness, the thickness of each layer, the interfacial diffusion, and so on have been characterized by X-ray diffraction (XRD), glacing incident X-ray reflectivity (GIXR), auger energy spectroscopy (AES), and atomic force microscopy (AFM). The surface for the bilayers is more smooth than that for thin film with one composition. The interfacial diffusion between YBCO and LCMO is observed. The transport properties for the prepared bilayers are measured by the standard four-probe method with or without applied magnetic field up to 8 Tesla. The nature of resistance for LCMO is screened by YBCO with the thickness more than about 25 nm, but the magnetoresistance (MR) effect is observed in the bilayers. MR increases with the decrease of the thickness of YBCO. The superconducting transtion temperature, Tc decreases with the decrease of the thickness of YBCO on STO with about 30 nm LCMO as buffer layer, and disappears at the thickness of about 4 nm. We believe that the variations of MR and Tc relate to the interaction between spin polarized electrons in the manganites and the cooper pair in the cuprates.

O3.7
MOVPE GROWTH AND CHARACTERIZATION OF SEMICONDUCTOR ZnO AND ZnCdO LAYERS WITH OPTICAL FUNCTIONS. Shizuo Fujita, Kyoto Univ, Int Innovation Center, Kyoto, JAPAN; Ken-ichi Ogata, Kyoto Univ, VBL, Kyoto, JAPAN; Sang-Woo Kim, Shigeo Fujita, Kyoto Univ, Dept of Electron Sci Eng, Kyoto, JAPAN.

In order to fully utilize the potential optical functions of ZnO-based semiconductors, solutions for three of the important issues are investigated. The first is the growth by metalorganic vapor-phase epitaxy (MOVPE) promising for practical mass production and artificial control of alloy composition. The second is the development of alloy materials for carrier-confinement device structures. Our research for this purpose is directed to ZnCdO layers which, from our recent work by MBE, are effective for exciton confinement in the Cd-rich region induced by remarkable phase-separation, similarly to the InGaN emission layers. The third is the growth on Si, as well as on sapphire, toward hybridization with Si-based devices. However, the nucleation of ZnO on Si is more difficult than on sapphire due to simultaneous oxidation of Si by the oxidizer source. First of all, among several oxidizer sources, it was found that NO_2, if grown with diethylzinc with sufficiently large VI/II ratio (e.g., 20000), is a promising source for high-quality ZnO layers, resulting in excitonic PL without noticeable deep-level emissions together with phonon replicas of them at low temperatures. Secondly, ZnCdO alloys were grown with dimethylcadmium, and Cd concentration was controlled upto a few percent without severe phase separation. The uniform PL was observed by a fluorescent microscopy. Thirdly, the growth on Si was achieved by inserting a ZnO nucleation layer on Si grown either by MOVPE with specific conditions, vacuum-evaporation, or sputtering. The main layer was then grown and resulted in excitonic PL lines both at room temperature and at low temperature. The nucleation layers also allowed the growth on GaAs and GaP, suggesting possible hybridization with various electrical and optical devices. Those findings for the solutions to grow high-quality ZnO-based semiconductor layers opened the possibility of developing a new material system for future novel optoelectronic devices.

O3.8
DEEP LEVEL TRANSIENT SPECTROSCOPY ANALYSIS OF AN ANATASE EPITAXIAL FILM GROWN BY METAL ORGANIC CHEMICAL VAPOR DEPOSITION. Takahira Miyagi[a,b], Tomoyuki Ogawa[b,c], Masayuki Kamei[b], Yoshiki Wada[b], Takefumi Mitsuhashi[b], Atsushi Yamazaki[a,b], Eiji Ohta[c], Tetsuya Sato[c]. [a]Waseda Univ, School of Science and Engineering, Tokyo, JAPAN; [b]Advanced Materials Laboratory, National Institute for Materials Science, Ibaraki, JAPAN; [c]Keio Univ, Faculty of Science and Technology, Kanagawa, JAPAN.

The deep level transient spectroscopy (DLTS) is an essential evaluation method for the semiconductor. However, even a single DLTS spectrum has not been reported for anatase-type TiO_2 to the best of our knowledge. Therefore, DLTS study of anatase-type TiO_2 material was performed for the first time. The anatase-type TiO_2 was epitaxialy grown on a conductive Nb-doped single-crystalline $SrTiO_3$ (100) substrate by metal organic chemical vapor deposition. The Shottky contact and ohmic contact for DLTS measurement were made by the Au electrode and the Nb-doped conducting $SrTiO_3$ substrate, respectively. According to the DLTS analysis, it was revealed that this anatase film had a characteristic deep level located at 0.96 eV below the bottom of the conduction band with large concentration (6.5×10^{16} /cm^3) and capture cross section (8.3×10^{-13} cm^2). Since this deep level has a large concentration combined with large capture cross section, the electrical properties of this film should be affected by this level and a further investigation of this level is necessary.

O3.9
POSSIBLE FORMATION OF INVERSION P-LAYER IN ZnO BASED METAL-INSULATOR-SEMICONDUCTOR HETEROSTRUCTURES. H. Saito, Electronic Components Laboratories, SHARP Corporation, Tenri, JAPAN; A. Tsukazaki, K. Tamura, C. Hirose, Dept. of Innovative and Engineered Materials, Tokyo Institute of Technology, Yokohama, JAPAN; H. Koinuma, Frontier Collaborative Research Center, Tokyo Institute of Technology, CREST, and COMET, Yokohama, JAPAN; M. Kawasaki, Insutitute of Material Research, Tohoku University, and COMET, Sendai, JAPAN.

Recentry, formation of inversion region with the metal-insulator-semiconductor (MIS) structure has became of great importance to device fabrication with new semiconductor materials[1,2]. ZnO, a wide-gap (3.37eV at room temperature) oxide semiconductor, is attractive one and potential candidate for ultraviolet optical device or invisible circuits. ZnO has much difficulty to controll p-type conduction by acceptor doping, however, it may be possible to generate more valuable devices by fabricating the p-layer and controlling its properties using the field-effect technique. Here we report on the investigation of inversion p-layer on ZnO MIS heterostructure. ZnO film was grown epitaxially on lattice-mattched $ScAlMgO_4(0001)$[3] substrate by laser MBE at growth temperature between 600 and 850 degrees. After growth we separated each device by etching and thinned the substrate by polishing and cleaving. Gate electrode was formed on the back of the thin $ScAlMgO_4$ substrate and source and drain electrode on the ZnO film. On the measurement of FET characteristics, deplation region cut off the drain current completely by -1,000V gate bias. The results of applying more bias to invert will be presented. [1] J.H. Schön et al., Science 289, 599 (2000) [2] I. Pallecchi et al., Appl. Phys. Lett. 78, 2244 (2001) [3] A. Ohtomo et al., Appl. Phys. Lett. 75, 2635 (1999).

O3.10
REACTIVE SPUTTER DEPOSITION OF EPITAXIAL CeO_2 ON (001) Ge. Mitesh Patel, Kyunghoon Kim, Mat Ivill, Hyung-Jin Bae, and David Norton, University of Florida, Department of Materials Science and Engineering, Gainesville, FL; John Budai and Matthew Chisholm, Oak Ridge National Laboratory, Oak Ridge, TN.

The formation of epitaxial oxides on semiconductors is of significant interest for field-effect devices as well as the integration of oxide functionality with conventional electronics. One particular challenge is in achieving epitaxial oxide growth using techniques that are amenable to large-area deposition. In this talk, we report on the epitaxial growth of (001) CeO−2 on (001) Ge using a hydrogen-assisted reactive sputter deposition method. A cerium metal target was used to achieve nucleation and epitaxy on (001) Ge, with water vapor serving as the oxidizing species. In order to control the formation of native GeO−2 on the substrate surface prior to and during nucleation of the CeO−2 film, hydrogen is introduced during the nucleation process. X-ray diffraction data of deposited CeO−2 show that the film is completely epitaxial with respect to the substrate. In addition to describing the effects of nucleation conditions on epitaxy, we will also discuss surface morphology and electronic properties of the deposited oxide films, including dielectric response of MOS-type structures. These structures should prove useful in understanding the synthesis and electronic properties of crystalline oxide/semiconductor interfaces.

O3.11
HETEROEPITAXIAL TRANSPARENT CONDUCTING OXIDE FILMS FOR ORGANIC LIGHT-EMITTING DIODES. H. Kim, J.S. Horwitz, R. Kim, Z.H. Kafafi, and D.B. Chrisey, Naval Research Laboratory, Washington, DC.

Transparent conducting oxide thin films such as indium tin oxide (ITO) and aluminum doped zinc oxide (AZO) were grown by pulsed laser deposition (PLD) on various single crystal substrates including yttria-stabilized zirconia (YSZ), sapphire, and magnesium oxide. The structural, electrical and optical properties of these films were investigated as a function of substrate deposition temperature and background gas pressure. Films were deposited using a KrF excimer laser (248nm, 30 ns FWHM) at fluences of 1 - 2 J/cm^2. Films were deposited at substrate temperatures ranging from 25°C to 600°C in oxygen pressures ranging from 1 to 100 mTorr. X-ray diffraction (XRD), scanning electron microscopy (SEM) and atomic force microscopy (AFM) were used to characterize the structure and morphology of the deposited films. UV/VIS/NIR spectroscopy and Hall effect measurements were used to characterize the optical and electrical properties. Heteroepitaxial ITO films (310 nm thick), grown by PLD on single crystalline YSZ substrate at 300°C and 10 mTorr of oxygen, show a resistivity as low as $1.\times 10-4\ \Omega$·cm with a visible transmittance of 90%. The properties of these films grown on various single crystal substrates will be compared and the use of these films as transparent anode electrodes for organic light-emitting diodes (OLEDs) will be discussed.

O3.12
EFFECTS OF EPITAXIAL STRAIN ON DOPING IN $YBa_2Cu_3O_{7-x}/PrBa_2Cu_3O_{7-x}$SUPERLATTICES. M. Varela, Univ. Carlos III, SPAIN; D. Arias, Z. Sefrioui, C. Leon, Univ. Complutense de Madrid, SPAIN; S.J. Pennycook, Oak Ridge National Lab, Oak Ridge, TN; C. Ballesteros, Univ. Carlos III, SPAIN; J. Santamaria, Univ. Complutense de Madrid, SPAIN.

The critical temperature of ultrathin (1-5 unit cells) $YBa_2Cu_3O_{7-x}$ layers is known to decrease when the thickness is reduced. Although several explanations have been proposed for this reduction (dimensionality, proximity effect, strain, charge transfer, etc.), the exact mechanism remains unknown. In this communication we examine this problem in high quality [$YBa_2Cu_3O_{7-x}$(YBCO) N / $PrBa_2Cu_3O_{7-x}$/(PBCO)M] 1000Å superlattices, with N ranging between 1 and 12 unit cells and M=5 unit cells, grown by high oxygen pressure sputtering [1]. Intracell structure has been analyzed by x ray diffraction (XRD) and transmission electron microscopy (TEM) [2]. We have found significant epitaxial strain for YBCO thickness below 4 unit cells. To investigate possible effects of the intracell strains in doping, we have systematically changed the doping level adjusting the oxygen content during sample cool down. Transport measurements and high spatial resolution electron energy loss spectroscopy (EELS) in a scanning transmission electron microscope conclusively show that the strained layers are underdoped, probably as a result of a hindered charge transfer arising from a reduced Ba-CuO chains distance in the strained layers. [1] M. Varela, Z. Sefrioui, D Arias, M. Navacerrada, M. Lucia, M.A. Lopez de la Torre, G. Loos C. Leon, and J. Santamaria. Phys. Rev. Lett. 83, 3936 (1999) [2] M. Varela, W. Grogger, D Arias, Z. Sefrioui, C. Leon, C. Ballesteros, K.M. Krishnnan, and J. Santamaria. Phys. Rev. Lett. 86, 5156 (2001). Acknowledgements: Work supported by CICYT MAT99 1706E.

O3.13
GROWTH MODE CHANGE OF ZnO HETEROEPITAXY AT EARLY STAGE BY A ULTRATHIN ZnO NUCLEATION LAYER. In Woo Kim, Hyun Seung Kim, and Seok Joo Doh, Department of Materials Science and Engineering, POSTECH, Pohang, KOREA; Jung Ho Je, Department of Materials Science and Engineering, POSTECH, Pohang, KOREA and Materials Science Division, Argonne National Laboratory, IL; Tae Sik Cho, Department of Materials Science and Engineering, Sangju National University, Sangju, KOREA; Byung Mook Weon, Gong-Seog Park, and Soo-Deok Han, Display Device Research Lab., LG Electronics Inc., KOREA.

A growth mode change of ZnO/sapphire(001) heteroepitaxy by a ultrathin (40 $\AA$) ZnO nucleation layer was studied in in-situ, real-time synchrotron x-ray scattering experiments. In early growth stage at high temperature of 500°C, the growth mode of ZnO changes to 2-dimensional (2D) layer mode by introducing the ultrathin ZnO nucleation layer, instead of 3D island mode on bare sapphire substrate. Even though the growth mode eventually changes to 3D island mode, the initial 2D layer mode enables not only the surface roughness to improve significantly, but also the multi-oriented grain growth at high temperature to be suppressed, sustaining the early stage epitaxial growth. Finally we discuss the structural evolution of ZnO on sapphire at high temperature from the viewpoint of wetting in nucleation.

O3.14
PHOTOLUMINESCENT PROPERTIES OF ZnMgO AND ZnCdO THIN FILMS GROWN BY METAL-ORGANIC VAPOR PHASE EPITAXY. W.I. Park, S.W. Jung, G.-C. Yi, H.M. Jang, Pohang Univ. of Science and Technology (POSTECH), Dept of MS&E, KOREA (SOUTH); S. Hong, T. Joo, Pohang Univ. of Science and Technology (POSTECH), Dept of Chemistry, KOREA (SOUTH).

ZnO, a wide-gap semiconductor oxide, has attracted considerable attention due to its large exciton binding energy (∼60 meV) and bond strength, which might make reliable high efficiency photonic devices based on ZnO. Recently it has also been reported that ZnMgO was grown with maximum Mg incorporation up to 36 at.% without phase separation and that the room temperature luminescence energy in this film blue-shifted from 3.3 to 4.0 eV. Since ZnMgO containing MgO over 4 at.% is in a thermodynamically metastable state, which indicates that the solubility limit of Mg in ZnO depends on growth mechanisms as well as growth conditions. Meanwhile, current research on the growth of ZnO-related alloys is restricted to pulsed laser deposition and molecular beam epitaxy. Despite the epitaxial growth of high quality ZnO and related alloys using these methods, they might have disadvantages in mass production, due to high cost and low throughput. In this talk, we demonstrate that metal-organic vapor phase epitaxy (MOVPE), which has great advantages in terms of large area deposition and atomic composition control feasibility, is an excellent technique for the epitaxial growth of high quality ZnO and related alloy films. In addition, the structural and optical characterizations of ZnMgO and ZnCdO thin films will be reported. By increasing Mg content up to 47 at.%, the c-axis constant of ZnMgO films decreased from 0.521 nm to 0.515 nm and no significant phase separation in the ZnMgO films was observed as determined by x-ray diffraction measurements. Furthermore, the near-band-edge (NBE) emission peak position showed blue shifts of 100, 430, and 570 meV with Mg content levels of 9, 21, and 47 at.%, respectively. Time-integrated and time-resolved photoluminescent properties of the ZnMgO and ZnCdO alloy films will also be discussed.

O3.15
METAL-ORGANIC VAPOR PHASE EPITAXIAL GROWTH AND PHOTOLUMINESCENT PROPERTIES OF ZnO/ZnMgO QUANTUM WELLS. W.I. Park, S.W. Jung, S.J. An, G.-C. Yi, Pohang Univ. of Science and Technology (POSTECH), Dept of MS&E, KOREA (SOUTH).

ZnO, a wide-gap semiconductor oxide, has attracted considerable attention due to its large exciton binding energy (∼60 meV) and bond strength, which might make reliable high efficiency photonic devices based on ZnO. Moreover, as reported by Othomo et al., the fundamental bandgap energy of this material increases from 3.3 eV to 4.0 eV by alloying ZnO with MgO, depending on Mg content, which might be used practically for fabrications of ZnO/ZnMgO heterostructure light emitters as well as ultraviolet photodetectors. With the introduction of a low dimensional double heterostructure, the modified density of states confines both excitons and photons, and facilitates greater efficiency of the emission process more efficient. Meanwhile, current research on the growth of ZnO-related alloys and quantum structures has been conducted using mainly pulsed laser deposition and molecular beam epitaxy. In this talk, we demonstrate that metal-organic vapor phase epitaxy (MOVPE), which has great advantages in terms of large area deposition and atomic composition control feasibility, is an excellent technique for the epitaxial growth of high quality ZnO/ZnMgO quantum well structures. For the fabrication of quantum wells, ZnO and ZnMgO films were epitaxially grown on $Al_2O_3(00\bullet1)$ substrates. Mg up to 47 at.% was incorporated into the films and the near-band-edge (NBE) emission peak position showed blue shift of 570 meV. Furthermore we fabricated ZnO/ZnMgO multiple quantum well structures and observed the blue shift from photoluminescence spectra of the ZnO layers in the quantum wells, which presumably results from the quantum confinement effect.

O3.16
THE ELECTRONIC PROPERTIES OF A GRAIN BOUNDARY IN Sb DOPED ZnO. J.M. Carlsson, B. Hellsing, Experimental Physics, Chalmers and Göteborg University, Gothenburg, SWEDEN; H.S. Domingos, P.D. Bristowe, Department of Materials Science and Metallurgy, University of Cambridge, Cambridge, UNITED KINGDOM.

The electronic properties of a Σ=13 [0001] tilt grain boundary in ZnO have been investigated using *first-principles* calculations. The structural units present in the undoped grain boundary are found to be consistent with those observed using electron microscopy. Doping the grain boundary with antimony reveals a strong driving force for segregation. Analysis of the electronic densities of states, bond populations and Mulliken charges shows that antimony acts as a donor dopant and that the boundary region contains bonds that are metallic in character.

O3.17
TRANSPORT PROPERTIES UNDER PRESSURE OF EPITAXIAL $SrRuO_3$ THIN FILMS. Francoise Le Marrec, Albin Demuer, Didier Jaccard, Jean-Marc Triscone, DPMC, University of Geneva, SWITZERLAND; Min Ku Lee, Chang-Beom Eom, University of Wisconsin-Madison, WI.

Measurements of electrical resistivity and Curie temperature have been performed on epitaxial oxide thin films (down to 350 Å) under hydrostatic pressures up to 25 GPa and temperatures down to 1.25 K. For these experiments, we chose the ferromagnetic metallic oxide $SrRuO_3$ which is chemically very stable and use the Bridgman anvils technique to produce pressure. The films were grown on vicinal (001) $SrTiO_3$ substrates in situ using 90^o degree off-axis sputtering.$SrRuO_3$ is an itinerant ferromagnet with a bulk Curie temperature of 160 K. Magnetic ordering manifests itself as a kink in the resistivity versus temperature curve. At atmospheric pressure, our thin films display a Curie temperature of about 150 K. This reduced Curie temperature is related to the lattice strains induced by lattice mismatch between the film and the substrate. Under pressure, our first results show that the room temperature resistivity of the materials is not modified whereas the Curie temperature is decreasing almost linearly at a rate of 4.5 K/GPa, a rate smaller than what is observed for bulk materials. At 16 GPa the measured Curie temperature is about 80 K.

O3.18
VARIATION IN MATERIALS PARAMETERS FOR HEXAGONAL $BaFe_{12}O_{19}$ FILMS ON (111) Mgo AS A FUNCTION OF OXYGEN PRESSURE. S.D. Yoon and C. Vittoria, Department of Electrical and Computer Engineering, Northeastern University, Boston, MA; S.A. Oliver, Center for Subsurface Sensing and Imaging Systems, Northeastern University, Boston, MA.

This research examines the material characteristics and magnetic properties of hexagonal M-type barium ferrite ($BaFe_{12}O_{19}$) films on (111) magnesium oxide substrates deposited by pulsed laser ablation deposition. All the films were deposited with identical deposition parameters except for oxygen pressure, which used 300mTorr to 10mTorr. All the films have excellent (0001) orientation, where x-ray diffraction data showed the lattice constant of the film grown at 300mTorr c=23.23$\AA$ increased to c=23.30$\AA$ for the film grown at 20mTorr. The films grown with higher oxygen pressure show rougher surfaces than lower oxygen pressure films from SEM surface morphology measurement. Saturation magnetization values ($4M_s$) for the films were obtained from hysteresis loops and torque magnetometry measurement. The uniaxial anisotropy field (H_a) of each film was also obtained by torque magnetometry and ferrimagnetic resonance measurement (FMR). The $4M_s$ and H_a values for deposited higher than 50mTorr films have values near bulk values where $4M_s \approx$ 4.80kOe and $H_a \approx$ 18.4kOe. Films deposited with oxygen pressure between 50 and 20mTorr obtained $4M_s \approx$ 3.43kOe and $H_a \approx$ 15.23kOe, which are lower than its bulk values. The narrowest FMR linewidths for as-produced films was obtained for the 10mTorr film where ΔH=0.39kOe. Films grown with the oxygen pressure higher than 200mTorr showed two resonance modes which is $\Delta H_1 \geq$ 0.90kOe and $\Delta H_2 \geq$ 1.10kOe at 59GHz. The FMR linewidths for this film decreased to ΔH_1=0.52kOe and ΔH_2=0.45kOe after annealing at 1000°C for 1 hour. These results indicate that the films grown at higher oxygen pressures have magnetic parameters near its bulk value but also show larger magnetic losses than films grown at lower oxygen pressures.

O3.19
TEMPERATURE DEPENDENCE OF THE GROWTH MODE OF ZnO/SAPPHIRE(001) THIN FILMS. Hyun Seung Kim, Seok Joo Doh, In Woo Kim, Jung Ho Je, POSTECH, Department of Materials Science and Engineering, Pohang, KOREA; Tae Sik Cho, Sangju National Univ, Department of Materials Science and Engineering, Sangju, KOREA.

We studied the temperature dependence of the growth mode of ZnO/sapphire(001) thin films in a radio-frequency magnetron sputtering, using *in-situ*, real-time synchrotron x-ray scattering. At low temperatures ($\leq$ 300°C), the ZnO film grows by highly strained two-dimensional (2D) epilayers in initial growth stage, followed by the growth of strain relieved three-dimensional (3D) islands on top of the 2D layers. At high temperatures ($\geq$ 400°C), however, the growth mode changes to Volmer-Weber mode, in which 3D islands directly grow on bare substrate. We discuss the temperature dependence of the growth mode of ZnO/sapphire films in viewpoints of adatom mobility, growth kinetics, strain energy, and interfacial energy in nucleation.

O3.20
DEFECTS IN EPITAXIAL $SrRuO_3$ FILMS ON (001) $SrTiO_3$

SUBSTRATE: MISFIT DISLOCATIONS AND IMPURITY PHASE PRECIPITATION. Sang Ho Oh, Chan Gyung Park, Pohang Unvi Sci & Tech (POSTECH), Dept of Materials Science and Engineering, Pohang, KOREA.

Deviation in either chemical composition or lattice parameters of epitaxial oxide films from bulk states can largely influence on the physical properties of heteroepitaxial structures. Off-stoichiometry in film composition can cause secondary phase precipitation, which may be expected or unexpected from equilibrium phase diagram, and lattice misfit between two materials can introduce defects such as misfit dislocation and twin at the interface to accommodate lattice misfit stress. In the present study, we report the transmission electron microscopy (TEM) study on the coherent metallic Ru precipitation and misfit dislocations in ion-beam sputtered epitaxial $SrRuO_3$ films prepared with stoichiometric $SrRuO_3$ target. Metallic Ru precipitates imbedded in primary phase of $SrRuO_3$ have regular "needle-like" shapes and are oriented to $< 110 >$ directions of substrate with densities up to $4.2 \times 10^{12}/cm^2$. The excess Ru incorporation of around 2 ~7%, which was induced by the difference in sputter yield between Sr and Ru, could cause a eutectoid solidification and resulted in the two-phase composite structure made of single-crystalline $SrRuO_3$ matrix and Ru precipitates. Epitaxial relationship between Ru precipitates and $SrTiO_3$ (STO) revealed to be $(011)_{Ru}//(002)_{STO}$ and $[100]_{Ru}//[110]_{STO}$. This type of in-plane arrangement is somewhat similar with $\{111\}_{pc}$ denotes pseudo-cubic symmetry of $SrRuO_3$) twin occurring at Sr-O planes of $SrRuO_3$ in order to accommodate oxygen deficiency or lattice misfit. The misfit dislocations in $SrRuO_3/SrTiO_3$ heteroepitaxial structure were analyzed by large angle convergent electron diffraction (LACBED) as well as diffraction contrast imaging methods. Above critical film thickness (~9.4 nm), the interface between $SrRuO_3$ and $SrTiO_3$ revealed the square network of misfit dislocations along $a_{pc} < 100 >$ directions with Burgers vectors of pure edge character $a_{pc} < 010 >$. Besides the square network of edge dislocations, pure screw dislocations along $a_{pc} < 110 >$ directions of $SrTiO_3$ substrate were observed to form along growth steps, due to the orthorhombic nature of $SrRuO_3$.

O3.21
MOCVD OF EPITAXIAL IRON OXIDE THIN FILMS. K. Shalini, Anjana Devi, and S.A. Shivashankar, Materials Research Centre, Indian Institute of Science, Bangalore, INDIA; Lehrstuhl für Anorganische Chemie II, Ruhr-Universität Bochum, Bochum, GERMANY.

Growth of high-quality single-crystal thin films of iron oxides such as α-Fe_2O_3, γ-Fe_2O_3, and Fe_3O_4 has received considerable interest because of technological applications in heterogeneous catalysis and magnetic recording, and as sensors. It is well known that with the increase in temperature, oxidation of Fe_3O_4 to α-Fe_2O_3 through a metastable γ-Fe_2O_3 occurs. Epitaxial films of these materials allow detailed study of the relevant properties of these oxides as a function of crystalline orientation. We have investigated the growth and epitaxy of thin films of these oxides by MOCVD using a novel metalorganic iron complex, tris(t-butyl-3-oxobutanoato)iron(III), as the precursor. The complex is a stable crystalline solid that sublimes above 120°C, thus being considerably more volatile than tris(2,4-pentadionato)iron(III). We have found that single crystal MgO(100) provides an excellent template for the growth of Fe_3O_4, as it has <1% lattice mismatch with the half unit cell dimension of Fe_3O_4 of the inverse spinel structure. We have also deposited strongly oriented α-Fe_2O_3 on single crystalline α-Al_2O_3 substrates as both of these are isostructural, though there is a considerable lattice mismatch of 5.8% between the two oxides. Under similar conditions of growth, on glass, polycrystalline Fe_3O_4 forms at lower temperatures and smaller oxygen flow rates and, as the temperature and the oxygen flow rates are increased, the phase changes to α-Fe_2O_3. Epitaxial growth has also been achieved on $SrTiO_3(100)$ and $NdGaO_3(100)$. The orientational relationship in epitaxy in the case of each substrate, and the quality of epitaxy, have been analyzed by phi-scan and rocking-curve x-ray diffraction. Film morphology has been studied by SEM and AFM.

O3.22
TRANSPARENT AND CONDUCTING Al DOPED ZnO THIN FILMS GROWN BY PULSED LASER DEPOSITION ON ALUMINA SUBSTRATES. P. Bhattacharya, J. Mass and R.S. Katiyar, Department of Physics, University of Puerto Rico, San Juan, PR.

Zinc oxide holds considerable promise as an optical transparent conducting material due to its wide band gap (3.3 eV) and can be heavily doped with group III dopants. Thin films of Al (1-5%) doped ZnO were deposited on (0001) oriented alumina (Al_2O_3) substrate by using KrF excimer laser ablation. The substrate was heated to 750°C and the film deposition was carried out with 1mTorr of oxygen pressure. The deposited films with different Al concentrations were found to be c-axis oriented and highly transparent (80%) in the visible range. X-ray diffraction study also showed that the films were with an internal stress as determined from the shift of (0002) peak. Undoped as well as 1% and 2% Al doped films showed a decrease of c-axis value, however, increase of Al concentration from 3 - 5% increased the c-axis value in comparison to the bulk ZnO. The lowest resistivity was obtained for 1% Al doped ZnO with a carrier concentration of $4x10^{19}$ and mobility 50 cm^2/V-s. The resistivity was increased with the increase of Al concentration above 1% mainly due to the decrease in mobility. The resistivity was almost unchanged with the variation of temperature from 140-400 K which reflects the Al doped ZnO films were degenerate. The optical bandgap of Al doped ZnO films was increased with increase of carrier concentration due to Burstein-Moss band filling effect. The effect of stress on electrical properties of Al doped ZnO films will be discussed. This work is supported in parts by DEFG029ER75764 and NASA-NCCC5-518 grants.

O3.23
MAGNETOTRANSPORT IN EPITAXIAL COLOSSAL MAGNETORESISTANCE THIN FILMS ON BARIUM TITANATE. Darren Dale, Dept. Mat. Sci. and Eng., Cornell University, Ithaca, NY; Aaron Fleet, Joel Brock, Dept. Applied and Eng. Physics, Cornell University, Ithaca, NY; Yuri Suzuki, Dept. Mat. Sci. and Eng., Cornell University, Ithaca, NY.

Epitaxial $La_{0.5}Sr_{0.5}MnO_3$ films on $BaTiO_3$ (BTO) and $SrTiO_3$ (STO) substrates provide model systems in which the effect of strain on magnetism and transport of colossal magnetoresistance materials can be systematically studied. Films are grown by pulsed laser deposition. The bulk LSMO has a 3.6% and 1.2% lattice mismatch with the BTO and STO substrates, respectively. Structural studies of LSMO films with thickness varying from 50 to 9000 $\AA$ indicate that lattice relaxation occurs for all films grown on BTO. Lattice relaxation occurs in films greater than 3000 $\AA$ thick grown on STO. Thickness dependence studies of the Curie temperature and saturation magnetization confirm the importance of strain on the magnetics. Upon varying temperature, the surface unit cell of the BTO substrate and thus the strain state of an LSMO film is altered. At 190K and 290K where BTO undergoes structural transitions, the magnetic and transport properties of the LSMO film exhibit hysteretic behavior attributed to the structural transition of the substrate. Hysteretic behavior of LSMO magnetism and transport is also observed between 190 to 290K where BTO is in its orthorhombic phase. This behavior correlates with the partial relaxation and nonreversible structural behavior of the LSMO film. We will also show the effects of poling BTO on the magnetics and transport of the overlying LSMO.

O3.24
EPITAXIAL THIN FILMS OF Co_3O_4 DEPOSITED ON SINGLE CRYSTAL MgO{100} BY LOW PRESSURE, LOW TEMPERATURE MOCVD. Anil U. Mane[a], K. Shalini[a], A. Wohlfart[b], A. Devi[b] and S.A. Shivashankar[a]; [a]Materials Research Centre, Indian Institute of Science, Bangalore, INDIA; [b]Lehrstuhl für Anorganische Chemie II, Ruhr-Universität Bochum, GERMANY.

The growth of highly oriented or $\{hetero-\}$ epitaxial thin films of transition metal oxides on suitable substrates usually requires that the growth temperatures be rather high or that energy be injected to the growth surface through, for example, the bombardment of the substrate with energetic particles during the growth process. We have grown epitaxial thin films of Co_3O_4 on different single crystal substrates at temperatures as low as 400°C by low pressure MOCVD using cobalt$\{II\}$ acetylacetonate as the precursor, and oxygen as the reactant gas. Highly oriented films of Co_3O_4 are formed on MgO{100}in the temperature range ~400-550°C, and a total reactor pressure of ~100 Pa-5 kPa. Films grown on MgO{100} at 410°C show a rocking curve FWHM of 0.44°, which is nearly as small as the FWHM of perovskite-type oxide films grown on $SrTiO_3$ {100} by laser ablation at ~750°C. The close lattice match between MgO and Co_3O_4 which is responsible for the observed high degree of epitaxy is reflected also in the sharply-lowered activation energy $\{E_a\}$ for film growth in the low temperature region; E_a is reduced from 101 kJ/mol for polycrystalline films grown on glass to 9.5 kJ/mol for epitaxial films on MgO{100} *grown simultaneously*. Rapid thermal annealing of Co_3O_4 on MgO{100}for 3 sec at ~725°C lowers the FWHM even further to only 0.06°, representing epitaxy of high quality. Phi-scan and pole figure analysis shows cube-on-cube epitaxy of Co_3O_4 on MgO, just as on $SrTiO_3$. The films have been characterized also by SEM and AFM, which show the epitaxial morphology clearly.

O3.25
ATOMICALLY ABRUPT INTERFACES AND LAYERS IN PEROVSKITE HETEROSTRUCTURES. Harold Y. Hwang, Akira Ohtomo, David A. Muller, John L. Grazul, Bell Laboratories, Lucent Technologies, Murray Hill, NJ.

Recent progress in heteroepitaxial oxide thin film growth has raised

the possibility of novel structures and interfaces between materials with very different physical properties. Because many of the scattering and coherence length scales are in the nanometer regime, understanding the degree of atomic precision at interfaces experimentally achievable is highly relevant. We have grown a number of perovskite thin film interfaces and multilayers by pulsed laser deposition. Scanning transmission electron microscopy studies and transport properties were used to examine the chemical and electronic profile at interfaces under variations in growth conditions. Using Z-constrast imaging, we demonstrate extreme delta-doping at the atomic level in perovskite heterostructures.

O3.26
STRUCTURAL, TRANSPORT AND MAGNETIC PROPERTIES OF $La_{0.79}Sr_{0.135}Ca_{0.075}MnO_3$ THIN FILMS AND $La_{0.79}Sr_{0.135}Ca_{0.075}MnO_3/La_{0.45}0.45Sr_{0.55}MnO_3$ BILAYERS. Bogdan Dabrowski, Northern Illinois University, Dept of Physics, DeKalb, IL; Piotr Przyslupski, Stanislaw Kolesnik, Northern Illinois University, Dept of Physics, DeKalb, IL/Institute of Physics, Polish Academy of Science, Warsaw, POLAND; Yasuo Ito, Northern Illinois University, Dept of Physics, DeKalb, IL.

Substituted $La_{1-x}Sr_xMnO_3$ (LSMO) compound shows an enhanced CMR effect at moderate field close to the room temperature due to a structural transition from orthorhombic to rhombohedral phase for x$\sim$0.17 doping level. Motivated by the results from polycrystalline ceramic samples we have performed similar experiments on epitaxial thin films. We will present results of our work on the structure, microstructure, transport and magnetic properties of sputtered $La_{0.79}Sr_{0.135}Ca_{0.075}MnO_3$ (LSCMO) films and LSCMO/LSMO heterostructures on $SrTiO_3$(100), $LaAlO_3$(100) and $NdGaO_3$(110) single crystal substrates. Both LSCMO thin films and LSCMO/LSMO bilayers were prepared by means of dc high pressure sputtering method, in-situ at argon pressure of 1 mbar. Magnetotransport data show a magnetoresistance peaked around 300 K, similar to the observation in polycrystalline samples. However, structural phase transformation is suppressed as a result of film/substrate lattice mismatch. Relationship between magnetotransport property and the film/substrate microstructure observed by the transmission electron microscopy will be discussed. Preliminary efforts to fabricate a spin valve-like heterostructure LSCMO/LSMO will be also presented.

O3.27
MICROSTRUCTURAL STABILITY AND DEFECT CHEMISTRY OF EPITAXIAL (La,Sr)$CoO_{3-\delta}$ THIN FILMS FOR ELECTROCHEMICAL APPLICATIONS. Susanne Stemmer, Department of Mechanical Engineering and Materials Science, Rice University, Houston, TX; Allan J. Jacobson, Department of Chemistry, University of Houston, Houston, TX; X. Chen, A. Ignatiev, SVEC, University of Houston, Houston, TX.

Thin films of complex oxides have several potential technological advantages for electrochemical devices, such as solid oxide fuel cells (SOFCs), oxygen transport membranes and gas sensors. They allow lower operating temperatures, thus reducing costs and materials degradation problems and the integration with electronic devices. Epitaxial thin films are also of scientific interest because they can be used to study the electronic and oxygen transport properties of materials, interfaces and surfaces along controlled orientations and in structurally highly perfect materials. $La_{0.5}Sr_{0.5}CoO_{3-\delta}$ (LSCO) is a good electronic and oxygen conductor, making LSCO interesting for a variety of thin film electrochemical applications. Oxygen nonstoichiometry and arrangements of oxygen vacancies in the microstructure all influence LSCO film properties, including the electronic and ionic conductivity, oxygen surface exchange and thermal mismatch with the substrate. This paper presents investigations of epitaxial LSCO thin films grown on $LaAlO_3$ single crystals, with the goal to establish the influence of thermal and chemical stresses on the oxygen vacancy concentration and arrangement in the films. The microstructure of LSCO thin films with well-characterized oxygen surface exchange properties was characterized by high-resolution transmission electron microscopy and electron diffraction before and after annealing treatments under different oxygen partial pressures. Structural models that explain the superstructure contrast observed in HRTEM in both films with cation shifts associated with oxygen vacancies are proposed and verified by HRTEM image simulations. The as-deposited film showed weak, short-range ordering of oxygen vacancies within nanometer-sized domains. The annealed film showed long-range order and a strong anisotropy in ordering, with the oxygen-deficient planes aligned in the plane of the film. We will discuss implications on the properties of thin film electrochemical devices. A model of the relationship between film stresses and the nature and concentration of oxygen vacancies in these films will be presented.

O3.28
EPITAXIAL GROWTH OF $BiMnO_3$ FILMS ON (001) $SrTiO_3$ THROUGH EPITAXIAL STABILIZATION. N.J. Murphy, J. Lettieri, D.G. Schlom, Penn State University, Dept of Materials Science and Engineering, State College, PA; A.M. Santos and T. Cheetham, University of California, Santa Barbara, CA.

Although limited information concerning the perovskite oxide $BiMnO_3$ exists, it is believed that $BiMnO_3$ is simultaneously both ferroelectric and ferromagnetic and may represent one of the simplest known multiferroic systems. As a result, this material is an intriguing material from a physics and device application viewpoint. The dearth of information on $BiMnO_3$ is in part due to the inability to form this phase at atmospheric pressure. Previous research, however, has demonstrated the synthesis of polycrystalline samples at high pressures (over 40,000 atm). In this study we use epitaxial stabilization to grow this metastable phase at low pressures. By studying these films we hope to gain insight into the very rare multiferroic character that is derived from its unique magnetic, electric, and physical structure relationship. Issues concerning growth regimes, phase stability, and orientation relationships will be discussed.

SESSION O4: ARTIFICIAL SUPERLATTICES
Chairs: David Vanderbilt and Harold Y. Hwang
Tuesday Morning, November 27, 2001
Room 203 (Hynes)

8:30 AM O4.1
EPITAXIAL GROWTH AND OPTIMIZATION OF TRANSPARENT CONDUCTING OXIDE MULTILAYER. Min Yan, Melissa Lane, John Ireland, Carl R. Kannewurf, and Robert P.H. Chang, Materials Research Center, Northwestern University, Evanston, IL.

Several transparent conducting oxide (TCO) compounds in the ternary alloy system of CdO-In_2O_3-SnO_2 exhibit encouraging electrical and optical properties. Furthermore, it is believed that there are some metastable phases and solution ranges not available in the bulk form but can be obtained as thin films. In our research, we synthesized and examined the electrical, optical and structural properties of thin films grown on MgO (111), Si (111) and Corning 1737 glass substrates with different compositions deposited in a multi-target pulsed laser deposition (PLD) system. Epitaxial growth of both pure and doped CdO and $CdIn_2O_4$ thin films has been achieved on MgO (111) substrates using PLD. A maximum conductivity of 42,000 S/cm with mobility of 609 $cm^2/V{\cdot}s$ is achieved when the CdO epitaxial film is doped with 2.5% Sn. Pure CdO epitaxial film has a bandgap of 2.4 eV and it increases with doping and reaches a maximum of 2.87 eV when the doping level is 6.2%. Both grain boundary scattering and ionized impurity scattering are found to contribute to the mobility of CdO films. For as deposited 2.5% Sn doped $CdIn_2O_4$, the conductivity achieved is 1,800 S/cm with a mobility of 22 $cm^2/V{\cdot}s$. It has a bandgap energy of 3.2 eV. Multilayered-film structure is then designed to achieve both high conductivity and high transmittance in the visible range: the 2.5% Sn doped CdO is used as the conductive layer and the 2.5% Sn doped $CdIn_2O_4$ is used as the wide bandgap layer. The optimized multilayer has an effective conductivity of 20,600 S/cm and an average transmittance larger than 85% in the range of 400-700 nm. Detailed results of the structural and basic solid state properties of these TCO films will be discussed.

8:45 AM O4.2
ELECTRONIC INTERACTION BETWEEN FERROMAGNETISM AND SUPERCONDUCTIVITY IN YBaCuO / LaCaMnO SUPERLATTICES. Hanns-Ulrich Habermeier, Georg Cristiani, Max-Planck Institut FKF, Stuttgart, GERMANY.

The physical properties of the perovskite-type oxide $RuSr_2GdCu_2O_8$ have been recently discussed in the view of a simultaneous occurrence of ordering mechanisms leading to superconductivity and ferromagnetism in different parts of the unit cell. In order to explore some peculiarities of these compounds such as presence or absence of the Meissner state we have prepared superlattices of oxides that are known to be either ferromagnetic [$La_{.67}Ca_{.33}MnO_3$] or superconducting [$YBa_2Cu_3O_7$]. Superlattices of different periodicity serve as model systems for the understanding of the peculiarities of the $RuSr_2GdCu_2O_8$ system and are used to compare their properties with those of our single phase epitaxially grown $RuSr_2GdCu_2O_8$ thin films. The YBCO/LCMO superlattices have been grown by pulsed laser deposition with individual layer thickness ranging from 4 to 200 unit cells for the $YBa_2Cu_3O_7$ and 10 to 500 unit cells for the $La_{.67}Ca_{.33}MnO_3$. The films are characterized by X-ray diffraction analysis, Raman spectroscopy, susceptibility and transport measurements. Whereas simple heterostructures [single layer $La_{.67}Ca_{.33}MnO_3$ and single layer $YBa_2Cu_3O_7$ 50 nm thickness each] reproduce the intrinsic properties of the constituent material rather well [Curie temperature 250K superconducting transition at T = 70K]

there are some novel effects emerging due to the coupling between the layers observed in the superlattices. Superlattices with individual thickness of the constituent materials of 4 nm e.g. show a reduced Curie temperature of 120K and a superconducting transition temperature of 60K. Lowering the temperature a reentrant normal state occurs at T = 25K. Switching off the electronic interlayer coupling by the introduction of insulating $SrTiO_3$ spacer layers leads to the intrinsic critical temperatures. The experimental findings are discussed within the frame of a phenomenological model based on ferromagnetic interlayer coupling and superconducting proximity effect.

9:00 AM O4.3
ELECTRONIC STRUCTURE OF METAL Ti/INSULATOR MgO SUPERLATTICE. K. Koga, K. Akai, K. Oshiro, T. Kado[a], and M. Matsuura, Faculty of Engineering, Yamaguchi University, Tokiwadai, Ube-city, JAPAN. [a]The National Institute of Advanced Industrial Science and Technology, Hiro, Kure-city, JAPAN.

MBE method has been used to obtain Metal Ti/Insulator MgO superlattice by one of the present author (T. Kado). In the present work, electronic band structure of Metal Ti/Insulator MgO superlattice is calculated by the full-potential linearlized augmented plane wave (FLAPW) with the generalized gradient approximation (GGA). The atomic configuration in the superlattice interface is determined from the comparison of calculated total energies. The calculated result shows that the Ti atom is in contact with the O atom at the interface. Based on this atomic configuration at the interface, electronic band structure of the superlattice is calculated. It is found that there are metallic states in a Ti layer within the band gap in an insulator MgO layer and then the metallic states constitute the quasi two-dimensional electronic states in a metallic Ti layer. Band diagram of Ti/MgO superlattices is shown. Also, calculated electronic band structure is used to calculate a tunneling current, i.e, a tunneling of an electron between two metallic Ti layers through the thin MgO layer under an electric field.

9:15 AM O4.4
RHEED-BASED COMPOSITION CONTROL FOR THE GROWTH OF Sr-Ti-O, Ba-Ti-O, Ba-Ru-O, AND Sr-Ru-O SYSTEMS BY MBE. J.H. Haeni, J. Lettieri, and D.G. Schlom, Dept of Materials Science and Engineering, Penn State University, University Park, PA; W. Tian, X.Q. Pan, Dept of Materials Science and Engineering, University of Michigan, Ann Arbor, MI.

The controlled synthesis of a wide variety of homologous oxide crystal systems and superlattices offers tremendous potential for tailoring the ferroelectric, dielectric and superconducting properties of these materials. The primary obstacle in the growth of high quality, phase pure members of a homologous series, or superlattices with sharp interfaces, is composition control. By monitoring the reflection high-energy electron diffraction (RHEED) intensity oscillation we have developed a method for precise composition control of systems containing Sr-Ti-O, Ba-Ti-O, Ba-Ru-O, and Sr-Ru-O. Using this technique we have grown the first five members of the $Sr_{n+1}Ti_nO_{3n+1}$ Ruddlesden-Popper Homologous Series, the first two members of the $Sr_{n+1}Ru_nO_{3n+1}$ Ruddlesden-Popper Homologous Series, and atomically abrupt $SrTiO_3/BaTiO_3$ superlattices. Structural and electronic characterization including in situ RHEED and four-circle x-ray diffraction analysis, high-resolution TEM images, dielectric and transport properties of these films we be presented.

9:30 AM O4.5
TRANSPORT PROPERTIES ASSOCIATED WITH LATTICE AND CHARGE MODULATION STRUCTURES IN $SrTiO_3/LaTiO_3$ SUPERLATTICES. Akira Ohtomo, David A. Muller, John L. Grazul, and Harold Y. Hwang, Bell Laboratories, Lucent Technologies, Murray Hill, NJ.

$La_{1-x}Sr_xTiO_3$ has been intensively studied in bulk, exhibiting a broad evolution of physical properties from a doped semiconductor to a Mott antiferromagnetic insulator as the formal titanium valence is varied from 4^+ to 3^+. In order to study the consequences of artificial charge modulation over varying distances, we have grown superlattice films consisting of $SrTiO_3$ and $LaTiO_3$ layers using laser molecular-beam-epitaxy which enables us to create sharp heterointerfaces and to control titanium valence between 3^+ and 4^+. As-grown films show metallic behavior dominated by carriers in oxygen deficient layers. These carriers are removed by annealing in oxygen, allowing measurements of the "intrinsic" magnetotransport properties of our structures. However, above a critical thickness of $LaTiO_3$ layer, oxygen annealing creates a partially-occupied doubling of the LaO layer (similar to those in layered perovskites such as $Bi_4Ti_3O_{12}$), resulting in formation of new superlattice structures. This stacking fault can be controlled coherently over many superlattice periods. Below the critical thickness, Ti valence modulated structures could be formed.

9:45 AM O4.6
MAPPING THE ELECTRONIC STRUCTURE OF ATOMIC-SCALE CHARGE MODULATIONS IN PEROVSKITE SUPERLATTICES. David A. Muller, Akira Ohtomo, John L. Grazul, Harold Y. Hwang, Bell Labs, Lucent Technologies, NJ.

How thin can a bulk material be before its bulk electronic properties are lost? We address this question using electron energy loss spectroscopy (EELS) to measure composition, valence and excited states on the atomic scale. Pulsed laser deposition is used to grow perovskite superlattices where the cation composition can be controlled with unit-cell precision. By growing quantum wells of varying thickness and examining the local bonding with EELS, we determine the critical length-scales required for bulk-like behavior. The spectroscopy is performed in a scanning transmission electron microscope where electrons scattered through a thin, cross-sectioned sample undergo energy losses characteristic of the different elements in the sample and their local environment. By scanning the electron beam across a quantum well and recording energy-loss spectra at each point in the scan, a composition and electronic-structure profile can be built up, atom-column by atom-column. Doing so, we observe differences in the degree of charge modulation from one to four unit-cell thick multilayers.

10:30 AM *O4.7
COMPOSITIONAL INVERSION SYMMETRY BREAKING IN FERROELECTRIC PEROVSKITES. David Vanderbilt, Department of Physics and Astronomy, Rutgers University, Piscataway, NJ.

Ternary cubic perovskite compounds of the form $(A_{1/3}A'_{1/3}A''_{1/3})BO_3$ and $A(B_{1/3}B'_{1/3}B''_{1/3})O_3$, in which the differentiated cations form an alternating series of monolayers, are studied [N. Sai, N. Meyer, and D. Vanderbilt, Phys. Rev. Lett. **84**, 5636 (2000)] using first-principles methods. Such compounds are representative of a possible new class of materials in which ferroelectricity is perturbed by compositional breaking of inversion symmetry imposed intentionally during atomic layer-by-layer growth. For isovalent substitution on either sublattice, the ferroelectric double-well potential is found to persist, but becomes sufficiently asymmetric that minority domains may no longer survive. The strength of the symmetry breaking is enormously stronger for heterovalent substitution, so that the double-well behavior is completely destroyed. Possible means of tuning between these behaviors may allow for the optimization of desirable materials properties such as the piezoelectric response.

11:00 AM *O4.8
NON-LINEAR ELECTRONIC RESPONSE FROM DIELECTRIC SUPERLATTICES WITH COMPOSITIONALLY BROKEN INVERSION SYMMETRY. James N. Eckstein, Maitri Warusawithana, Dario Farias, Bruce Davidson, University of Illinois at Urbana-Champaign, Dept of Physics, Urbana, IL; Charles Ahn, Yale University, Dept of Applied Physics.

Three wave mixing processes such as second harmonic generation, sum or difference frequency generation and the linear electro-optic effect rely on the second order electronic susceptibility, chi-2. While many other factors are important in practice, finding a way to increase chi-2 would reduce the size of devices using such mixing or reduce the power required to operate them. In order to possess such a susceptibility, a material should exhibit broken inversion symmetry. This allows a dielectric response different for oppositely directed electric fields. Currently, ferroelectric or similar materials are used for this application. The spontaneously broken inversion symmetry gives rise to a non-zero chi-2, which also depends on the curvature of the frequency dependent polarization response. Higher response can be obtained operating near the curie temperature, but in this case the material is unstable to repoling. In order to increase chi-2 we have investigated assembling complex oxide ferroelectric superlattices having broken inversion symmetry designed into the molecular layer stacking sequence. Such superlattices effectively bias the sample, which has a non-zero polarization even in the paraelectric phase. This provides another materials parameter, namely the stacking architecture, which can be chosen to increase the aggregate chi-2 of the sample. Initial experiments show about a factor of three greater reflection second harmonic generation from a thin film with broken inversion symmetry compared with one having inversion symmetry.

11:30 AM *O4.9
FIRST-PRINCIPLES STUDY OF METAL/FERROELECTRIC AND INSULATOR/FERROELECTRIC HETEROSTRUCTURES. Philippe Ghosez, Magali Zimmer, Univ. of Liège, Dept of Physics, Liège, BELGIUM.

Heterostructures composed of thin ferroelectric oxide films in contact with metallic or insulating neighboring compounds present an interest for various technological applications. The use of the functionalities of

the ferroelectric material in ultra-thin, eventually nanosized, devices raises fundamental questions concerning the evolution of the ferroelectric properties with thickness and the influence of the neighboring phases on this size dependence. This includes questions concerning the role of the interface chemistry as well as the influence of the mechanical and electrical boundary conditions imposed by the neighboring phases. In order to address some of these problems, we report first-principles calculations, performed within the density functional theory, on metal/ferroelectric ($SrRuO_3/BaTiO_3$) and insulator/ferroelectric ($BaO/BaTiO_3$) heterostructures. We discuss the properties of both kinds of structure and compare the results with what is observed for free standing slabs.

SESSION O5/C6: JOINT SESSION
EPITAXIAL FERROELECTRIC FILMS
Chairs: Darrell G. Schlom and Stephen K. Streiffer
Tuesday Afternoon, November 27, 2001
Room 210 (Hynes)

1:30 PM O5.1/C6.1
FERROELECTRIC DOMAIN STRUCTURE OF $SrBi_2Nb_2O_9$ AND $SrBi_2Ta_2O_9$ EPITAXIAL FILMS. M.A. Zurbuchen, G. Asayama, J. Lettieri, Y. Jia, L.Q. Chen, and D.G. Schlom, Department of Materials Science and Engineering, The Pennsylvania State University, University Park, PA; S.K. Streiffer, Materials Science Division, Argonne National Laboratory, IL; X.Q. Pan, Dept of Materials Science and Engineering, The University of Michigan, Ann Arbor, MI.

Using two different techniques, dark field transmission electron microscopy (TEM) and FFT analysis of high-resolution TEM images, we have for the first time analyzed the ferroelectric domain structure of $SrBi_2Ta_2O_9$ and $SrBi_2Nb_2O_9$ epitaxial thin films, grown on both (001) and (111) $SrTiO_3$ substrates. Ferroelectric domains are non-laminar, with an average diameter of 50 nm. Unlike the domain walls in many other ferroelectrics, the domain walls in $SrBi_2Nb_2O_9$ and $SrBi_2Ta_2O_9$ are often highly curved, particularly in $SrBi_2Nb_2O_9$. This is believed to arise due their extremely low ferroelastic distortion; $SrBi_2Nb_2O_9$ has a ferroelastic distortion three orders of magnitude less than that of PZT. This results in a much weaker role of preferred crystallographic orientation for domain walls. A brief overview of the techniques, which can be applied in a straightforward manner to many types of low-distortion ferroelectric films, will also be given. The resolution using the dark field TEM technique is anisotropic and is limited by drift and specimen thickness; however, with the high-resolution TEM technique it is possible to resolve ferroelectric domain structure detail of 2.5 nm.

1:45 PM O5.2/C6.2
PREPARATION AND CHARACTERIZATION OF a- AND b-AXES-ORIENTED EPITAXIALLY GROWN $Bi_4Ti_3O_{12}$-BASED THIN FILMS WITH LONG-RANGE LATTICE MATCHING. Takayuki Watanabe and Hiroshi Funakubo, Dept of Innovative and Engineered Materials, Tokyo Institute of Technology, Kanagawa, JAPAN; Keisuke Saito, Application Laboratory, Analytical Department, Philips Japan, Ltd., Kanagawa, JAPAN.

Thin films of bismuth layer-structured ferroelectrics (BLSFs) have been investigated for a ferroelectric random access memory (FeRAM) application because of its good ferroelectric properties, especially high fatigue endurance. We have systematically investigated the dependence of the ferroelectricity on the film orientation and the m-number using various kinds of epitaxially grown BLSFs films. However, the characterization of the ferroelectricity along [100] direction, polar axis, is limited only for bulk single crystals. Especially there is no reports on the characterization of epitaxially grown films. This is due to the difficulty of the long range lattice matching between the long c-axis of BLSFs and the substrates. In the present study, we focused on an epitaxially grown conductive oxide films with rutile-type structure as bottom electrodes for the preparation of a- and b-axes-oriented BLSFs. We prepared Nd- and V- substituted $Bi_4Ti_3O_{12}$ [$(Bi,Nd)_4(Ti,V)_3O_{12}$] film with c-axis lattice constant of approximately 3.3nm. a- and b-axes-oriented epitaxial $(Bi,Nd)_4(Ti,V)_3O_{12}$ films were successfully deposited on $(101)RuO_2//(012)Al_2O_3$ and $(101)IrO_2//(012)Al_2O_3$ substrates at 600°C by metalorganic chemical vapor deposition. By X-ray pole figure measurement with a fixed 2Theta angle corresponding to (117)BNTV peak, fourfold dense poles located around psi angle = 56° indicating the epitaxial growth of a- and b-axes-oriented BNTV films on each substrates;
$(100)(010)(Bi,Nd)_4(Ti,V)_3O_{12}//(101)RuO_2//(012)Al_2O_3$ and $(100)(010)(Bi,Nd)_4(Ti,V)_3O_{12}//(101)IrO_2//(012)Al_2O_3$. Finally, a large remanent polarization above $20\mu C/cm^2$ was observed.

2:00 PM O5.3/C6.3
GROWTH AND CHARACTERIZATION OF FERROELECTRIC La-SUBSTITUTED $Bi_4Ti_3O_{12}$ EPITAXIAL THIN FILMS BY PULSED LASER DEPOSITION. Ho Nyung Lee, Dmitri N. Zakharov, and Dietrich Hesse, Max-Planck-Institut für Mikrostrukturphysik, Halle/Saale, GERMANY.

La-substituted $Bi_4Ti_3O_{12}$ (BLT) thin films have been received much attention due to their good ferroelectric properties and high fatigue endurance as well. Like $SrBi_2Ta_2O_9$ (SBT), BLT has an anisotropic crystallographic structure and anisotropic ferroelectric properties. There are several reports on either polycrystalline or randomly oriented BLT films. However, few works on the epitaxial growth of $Bi_4Ti_3O_{12}$ films are reported to date and these are mostly concerned only with the growth of *c*-axis-oriented films. Ferroelectric epitaxial BLT thin films having (001), (118), and (104) orientations (in orthorhombic indices) have been grown by pulsed laser deposition on conducting $SrRuO_3$-covered (001)-, (011)-, and (111)-oriented $SrTiO_3$ or Nb-doped $SrTiO_3$ substrates, and on buffered Si substrates. Due to the crystallographic anisotropy of BLT, epitaxial films of different crystallographic orientations display different properties along the direction normal to the film plane. This anisotropy is being investigated by x-ray diffraction, transmission electron microscopy, and electrical measurements, using epitaxially grown films of (001), (118), and (104) orientations. Heteroepitaxial films of BLT(118)/$SrRuO_3$(110)/YSZ(100) and BLT(104)/$SrRuO_3$(111)/MgO(111)/YSZ(100) are grown *in situ* on Si(100) substrates. Investigations of crystallographic orientation relationships and structural and electrical properties will be presented. The results will be compared to those obtained from non-doped $Bi_4Ti_3O_{12}$ epitaxial thin films having the same orientations, to characterize the effect of La substitution on the ferroelectric properties.

2:15 PM *O5.4/C6.4
SURFACE, THICKNESS, AND STRAIN EFFECTS ON STRUCTURAL INSTABILITIES IN EPITAXIAL $PbTiO_3$ FILMS. Brian Stephenson, Stephen Streiffer, Jeffrey Eastman, Orlando Auciello, Anneli Munkholm, Materials Science Div, Argonne National Laboratory, Argonne, IL; Carol Thompson, Marian Aanerud, Dept of Physics, Northern Illinois University, DeKalb, IL.

As in many perovskites, competing ferroelectric and antiferrodistortive instabilities are present in $PbTiO_3$. In the bulk, the ferroelectric instability is stronger. However, surface, strain, and film thickness effects can either augment or suppress such structural instabilities and the associated property enhancements. We present results from *in situ* x-ray scattering studies during and after organo-metallic vapor phase epitaxy of $PbTiO_3$ on $SrTiO_3$, carried out at the Advanced Photon Source. We find that, under an equilibrium vapor pressure of PbO, the $PbTiO_3$ (001) surface reconstructs to a $c(2 \times 2)$ structure consisting of a single antiferrodistortive layer with oxygen cages counter-rotated 10 degrees about the Ti ions. We have also investigated the ferroelectric phase transition in thin (< 400 Å), coherently-strained films. We find that not only is T_C elevated above the bulk value, as predicted, but in addition the ferroelectric phase forms as 180-degree stripe domains in these films, with a thickness-dependent T_C. This results from a fascinating competition between strain, polarization, and electric field that determines the minimum free energy configuration. This work is supported by the State of Illinois under HECA, and the U.S. Department of Energy, BES-DMS under contract W-31-109-ENG-38.

3:15 PM *O5.5/C6.5
SIZE EFFECTS ON POLARIZATION IN EPITAXIAL FERROELECTRIC THIN FILMS. H. Kohlstedt, N.A. Pertsev, R. Waser, Forschungszentrum Jülich, Institut für Festkörperforschung, Jülich, GERMANY; A.G. Zembilgotov, State Technical Univ of St. Petersburg, St. Petersburg, RUSSIA.

Extrinsic and intrinsic size effects on the spontaneous polarization of epitaxial single-domain ferroelectric films are discussed. The former includes the dead-layer effect and the misfit-strain one, while the latter is due to spatial correlations of the ferroelectric polarization. The competition between different effects may lead to a nonmonotonic scaling of polarization with the film thickness. The dead-layer effect is known to be associated with the presence of nonferroelectric subsurface layers. It is supposed that in perovskite ferroelectrics the depolarizing-field effect on the static properties may be neglected due to their finite conductivity. Experimental data suggest that the structure of subsurface layers depends on the electrode material. The use of electrodes with a perovskite structure is expected to reduce the dead-layer effect. The extrinsic size effect is also attributed to the thickness dependence of the film in-plane lattice strain S. This dependence appears via the strain relaxation caused by the generation of misfit dislocations. A nonlinear thermodynamic theory, which allows for the mechanical film/substrate interaction, is used to describe the variation of the film polarization with the misfit strain S in the heterostructure. The intrinsic effect of the film surfaces is taken into account via the concept of extrapolation length L. Thermodynamic calculations are performed for lead titanate and

barium titanate (BTO) films grown on compressive substrates (S < 0). The negative intrinsic size effect (L > 0) is found to be reduced in strained films. It is shown that, well below the transition temperature, the film mean polarization may be larger than the bulk polarization even in an ultrathin epitaxial layer. This increase is caused by the in-plane compression the film lattice, which overlaps the negative surface effect. The theory explains the increase of polarization in very thin (10-20 nm) BTO films grown on strontium titanate.

3:45 PM O5.6/C6.6

NANOSCALE CONTROL AND SWITCHING DYNAMICS IN EPITAXIAL $Pb(Zr_{0.2}Ti_{0.8})O_3$ THIN FILMS. P. Paruch, T. Tybell and J.-M. Triscone, University of Geneva, DPMC, Geneva, SWITZERLAND.

Atomic force microscopy (AFM) was used to precisely manipulate and study individual ferroelectric domains in high quality $Pb(Zr_{0.2}Ti_{0.8})O_3$ thin films. Nanoscale control of domain size was achieved by varying the strength and duration of the voltage pulses used to polarize the material, permitting the creation of sub-20nm width lines and ultra-high density arrays reaching $\sim$30 Gbit/cm^2. The domains were individually accessible, fully reversible, and all, including those written with $\sim$100ns pulses, remained completely stable for the 7-day duration of the experiment. Understanding the relation between domain size and writing parameters, and the resulting local control, are potentially important for applications such as random access ferroelectric memories and ultra-high density storage. The AFM approach developed also allowed us to investigate the fundamental physics of domain interactions and switching behaviour in ferroelectrics. For sub $\sim 20\mu$s writing times, domain size was observed to saturate at $\sim$40nm diameter, limited, we believe, by the size of the AFM tip itself. For longer writing times, domain size was found to depend linearly upon the writing voltage and logarithmically upon the writing time. This indicates a field activated domain growth mechanism, via nucleation-assisted domain wall motion. The activation field increases with decreasing film thickness. For the thinnest films investigated, 200Å, the value is $\sim$200 times larger than the experimentally measured coercive field in thick films. P. Paruch, T. Tybell, J.-M Triscone, to appear in Appl. Phys. Lett. July 23, 2001. "Nanoscale control of ferroelectric polarization and domain size in epitaxial $Pb(Zr_{0.2}Ti_{0.8})O_3$ thin films"; T. Tybell, C.H. Ahn, J.-M. Triscone, Appl. Phys. Lett. 75, 856 (1999). "Ferroelectricity in thin perovskite films"; T. Tybell, C.H. Ahn, J.-M. Triscone, Appl. Phys. Lett 72, 1454 (1998). "Control and imaging of ferroelectric domains over large areas with nanometer resolution in atomically smooth epitaxial $Pb(Zr_{0.2}Ti_{0.8})O_3$ thin films".

4:00 PM O5.7/C6.7

FUNCTIONAL BEHAVIOUR OF THIN FILM DIELECTRIC SUPERLATTICES. J.M. Gregg, M.H. Corbett, D. O'Neill, G. Catalan, R.M. Bowman, Queens University Belfast, Department of Pure and Applied Physics, Belfast, N. Ireland, UNITED KINGDOM.

Pulsed laser deposition has been used to fabricate thin-film capacitor structures in which the dielectric layer is a superlattice. The properties of two superlattice systems were investigated as a function of superlattice wavelength (λ) - one based on barium strontium titanate and the other on lead-based relaxor electroceramics. In both systems the dielectric constant was significantly enhanced at stacking wavelengths of a few unit cells. However, the dielectric enhancement seen in the barium strontium titanate superlattices was found to be due to Maxwell-Wagner effects, whereas in the relaxor superlattices Maxwell-Wagner behaviour was not evident; rather, in relaxors, the dielectric enhancement was associated with the onset of polar coupling around λ - 10nm.

4:15 PM O5.8/C6.8

STRUCTURAL AND FERROELECTRIC PROPERTIES OF $BaTiO_3$ AND $Ba_xSr_{1-x}TiO_3$ THIN FILMS. J. Schubert, O. Trithaveesak, Ch. Buchal, Institut für Schichten und Grenzflächen, Forschungszentrum Jülich GmbH, Jülich, GERMANY; J. Rodríguez Contreras, K. Szot, H. Kohlstedt, Institut für Festkörperforschung, Forschungszentrum Jülich GmbH, Jülich, GERMANY.

The ferroelectric properties of $BaTiO_3$ and $Ba_xSr_{1-x}TiO_3$ thin films are of current interest for future electronic devices. We have prepared epitaxial $BaTiO_3$ (BTO) and $Ba_{0.5}Sr_{0.5}TiO_3$ (BST) thin films using pulsed laser deposition. Films of thicknesses between 10 nm and 200 nm were grown on epitaxial $SrRuO_3$ (SRO) films on $SrTiO_3$(100) substrates (STO) or directly on STO. The SRO was prepared ex-situ by high-pressure on-axis sputtering. The structure of the films was characterized by X-ray diffraction, Rutherford backscattering spectrometry/channeling and atomic force microscopy (AFM) measurements. X-ray diffraction indicates that the BTO and BST thin films grow c-axis oriented on STO and on SRO/STO. The c-axis is elongated in the thinner films. A small mosaicity of $\Delta\omega < 0.2°$ is observed. Minimum yield values of less than 2% are typical for BST thin films down to a thickness of 10 nm. AFM measurements reveal very smooth surfaces of order 0.3 nm RMS roughness for a 60 nm thick BST-film on STO as well as on SRO/STO. Pt/BST/SRO and SRO/BTO/SRO trilayers were patterned to capacitors. Ferroelectric hysteresis loops have been obtained for BST film thicknesses ranging from 200 nm down to 50 nm and for BTO down to 10 nm. A 12 nm thick BTO film on SRO bottom electrode using SRO top electrode shows a remanent polarization of 26 μC/cm^2 ($2P_r$) and an coercitive field of 720 kV/cm ($2E_c$) (measured at f=40 kHz). Generally the remanent polarization P_r decreases while the coercitive field E_c increases towards thinner BTO film thickness. 100 nm thick BTO films showed no fatigue for 10^8 cycles.

4:30 PM O5.9/C6.9

IN-SITU GROWTH OF COMPOSITIONALLY GRADED OXIDE FILMS BY PLD. H.M. Christen, H.Y. Zhai, and D.H. Lowndes, Oak Ridge National Laboratory, Solid State Division, Oak Ridge, TN.

Rapid sequential deposition of sub-monolayer amounts of different materials is an effective way to form alloys between multiple constituents without requiring a post-annealing process to promote interdiffusion. This technique is readily implemented in pulsed laser deposition, where the firing of the laser can be synchronized with the exchange of multiple targets, and where a small set of targets is sufficient to obtain films with various stoichiometries. Using appropriate automation techniques, any arbitrary composition profile perpendicular to the substrate surface can be obtained, including continuously graded profiles and periodic structures with smooth or abrupt composition variations. Differently graded materials can exhibit interesting of strain profiles that may prove useful for the stabilization of metastable phases. Results will be shown for the case of alloys between dissimilar ferroelectrics as well as between conducting and insulating perovskites. Applications to oxide electronics, mode-discriminating optical waveguides, and buffer layers for the integration of dissimilar materials will be discussed, including the development of compositionally graded buffer layers for the growth of high-Tc superconductors on metal tapes. Research sponsored by the US Department of Energy under contract DE-AC05-00OR22735 with the Oak Ridge National Laboratory, managed by UT-Battelle, LLC, and by the DOE Office of Energy Efficiency and Renewable Energy, Office of Power Technologies Superconductivity Program.

4:45 PM O5.10/C6.10

PULSED LASER DEPOSITION OF $Ba_{1-x}Sr_xTiO_3$, (x = 0.4) THIN FILMS ON $LiNbO_3$ X-CUT AND Z-CUT SUBSTRATES: STABILIZATION OF THE RHOMBOHEDRAL PHASE AT ROOM TEMPERATURE. Daniel M. Bubb, J.S. Horwitz, S.B. Qadri, R.M. Stroud, W. Chang, S. Kirchoefer, and D.B. Chrisey, Naval Research Laboratory, Washington, DC.

Single phase, epitaxial thin films of $Ba_{1-x}Sr_xTiO_3$, (x = 0.4) have been grown on x-cut and z-cut $LiNbO_3$ substrates by pulsed laser deposition. X-ray diffraction studies show that the films are epitaxial and assume the rhombohedral symmetry of the substrate. To our knowledge, this is the first such observation of rhombohedral $Ba_{1-x}Sr_xTiO_3$ thin films at room temperature. Rutherford Backscattering measurements have been performed on the films and demonstrate that the films have the correct stoichiometry. Transmission electron microscopy (TEM) images of the film/substrate interface will be presented as an aid in understanding the stabilization of the rhombohedral phase. Interdigitated capacitors have been placed on the films and the microwave dielectric response has been extensively investigated. We will compare these results with earlier structure/property studies of cubic/tetragonal $Ba_{1-x}Sr_xTiO_3$ thin films on MgO and $LaAlO_3$ substrates.

SESSION O6/C8: JOINT POSTER SESSION
EPITAXIAL FERROELECTRIC FILMS
Chairs: Sanjeev Aggarwal and Masaru Shimizu
Tuesday Evening, November 27, 2001
8:00 PM
Exhibition Hall D (Hynes)

O6.1/C8.1

FERROELECTRIC $BaTiO_3$ THIN FILM OPTICAL WAVEGUIDE MODULATORS. A. Petraru, M. Siegert, J. Schubert and Ch. Buchal, Institut für Schichten und Grenzflächen (ISG1-IT), Forschungszentrum Jülich, GERMANY.

High quality $BaTiO_3$ epitaxial thin films on MgO substrates have been grown by pulsed laser deposition. Both, c-axis and a-axis $BaTiO_3$ orientation were studied. For growing the c-axis films the substrate temperature was approx. 800°C and the laser energy 1100 mJ/pulse. In the case of the a-axis films the substrate temperature was 50°C higher and the laser energy 300 mJ/pulse. Mach-Zehnder

optical waveguide modulators with a fork angle of 1.7 deg. have been fabricated by ion beam etching. The waveguides are of the ridge type, the $BaTiO_3$ thickness is 1 μm, the ridge step 50 nm, the width 2 μm. Light was coupled into the waveguides from optical fibers. The $BaTiO_3$ waveguide propagation losses are 1-2 dB/cm. Electrodes of 3 mm length were deposited besides the waveguides. Electro-optic modulation has been demonstrated with V_π= 6.3V @ 632nm wavelength and V_π = 9.5V @1550nm wavelength for the a-axis samples and with V_π= 7V @ 632nm wavelength and V_π= 15V @1550nm for the c-axis samples. Theoretical modelling of the Mach-Zehnder modulators for both crystalline orientations of the $BaTiO_3$ films gave the Pockels coefficients r_{51} =30 pm/V for the c-axis film and an effective Pockels coefficient r_{eff} = 734 pm/V for the a-axis films at 632 nm wavelength.

O6.2/C8.2

EQUIVALENT CIRCUIT INTERPRETATION OF DIELECTRIC DISPERSION IN FERROELECTRIC SUPERLATTICE CAPACITOR. Mufei Xiao, CCMC-UNAM, San Ysidro, CA.

We present an alternative explanation to the dielectric dispersion at low frequency in ferroelectric superlattice capacitors. The dispersions were recently observed and attributed to the Maxwell-Wagner effects in [D. O'Neill, R.M. Bowman, and J.M. Gregg, Appl. Phys. Lett. 77 (2000)1520]. We alternatively explain the observations based on an equivalent circuit. For small stack periodicities that are less than a few Angstroms, the model reaches similar conclusions that the Maxwell-Wagner effects contribute to the dielectric loss at low frequencies. For larger stack periodicities, the model interprets correctly the experimental observations. Finally, the model appears useful for understanding dielectric responses in relaxor ferroelectrics, and thus suggests that stacking various ferroelectric thin films may produce relaxor responses.

O6.3/C8.3

ELECTROMECHANICAL RESPONSES OF SUB-MICRON RELAXOR STRUCTURES. Robert Bowman, Carles Morros, Gustau Catalan, Niall Donnelly and Marty Gregg, School of Maths & Physics, Queen's University of Belfast, UNITED KINGDOM.

The large electromechanical response of relaxor compounds makes these materials interesting for numerous applications. Recent advances in film fabrication have lead to creation of high quality thin films and much work is underway to quantify and understand the relaxor behaviour in thin films. Dielectric and electromechanical measurements at a microscopic level and very small volumes are an additional aid. Using pulsed laser deposition we fabricate single layers of PMN and PMN-PT and superlattices of relaxor compositions such as PZN-BT/PMN. Piezoresponse microscopy using a modified AFM allows the mapping and quantification of electromechanical behaviour on the surface of the films. In PMN layers we measure an effective d_{33} of around 300pm/V in fields of 0.25MV/cm. In 90 x 6nm period superlattices of 0.2PZN-0.8BT/PMN strains of 0.4% in 0.25MV/cm are seen. In this paper we will describe the properties of such films and superlattices when they have been laterally confined by the creation of sub-micron ($\leq$500nm) features such as mesas and strips; in addition to the confinement by reducing thickness of the grown film. The relaxor films are fashioned using a focused ion beam microscope (FIB); a FEI Strata using a Ga^+ beam is used to define the elements. Detailed dielecric and electromechanical measurements are preformed on either bare surfaces of patterned films or metalised top layers, with either ex-situ noble metal coating or FIB deposited Pt. HRTEM is also used to correlate measurements with microstructure.

O6.4/C8.4

INITIAL GROWTH STAGES OF $BaTiO_3$ THIN FILMS ON Nb-DOPED $SrTiO_3$ SUBSTRATES. A. Visinoiu, M. Alexe, H.N. Lee, C. Harnagea, D.N. Zakharov, A. Pignolet and D. Hesse, Max Planck Institute of Microstructure Physics, Halle, GERMANY.

The growth mechanism of epitaxial $BaTiO_3$ films on vicinal Nb-doped $SrTiO_3$ (Nb:$SrTiO_3$) substrates has been studied. (100)-oriented $BaTiO_3$ thin films and nanostructures have been grown on (100)-oriented Nb:$SrTiO_3$ substrates with well-defined terraces by pulsed laser deposition. As initial stages of film growth, self-organized $BaTiO_3$ nanostructures 5 to 10 nm in height and 100 nm in lateral size have been formed. Substrate surfaces consisting of TiO_2-terminated steps have been prepared by chemical etching in buffered HF solution and 10 min thermal annealing at 1200°C. Under our conditions, barium titanate preferred to grow with an island growth mechanism. The lattice mismatch of 2.28 % along the a direction between $BaTiO_3$ (a = 3.994 Å) and $SrTiO_3$ (a = 3.905 Å) strongly influences the epitaxial relationship between film and substrate. The size of the individual $BaTiO_3$ islands has been controlled by the variation of laser repetition rate and overall number of laser shots. X-ray diffraction and high-resolution transmission electron microscopy have been performed in order to check the orientation and crystallinity of the $BaTiO_3$ films and structures. Macroscopic measurements of dielectric properties of 500 nm thick films show a relative dielectric constant ϵ_r of 200, a remnant polarization P_r of 0.72 μC/cm^2, and a coercive field E_c of 27 kV/cm for a maximum applied field of 100 kV/cm. Local piezoelectric measurements confirmed the ferroelectric behavior, as well as the presence of an initial imprint in $BaTiO_3$ thin films on Nb:$SrTiO_3$ substrates.

O6.5/C8.5

ORIENTATION-CONTROLLED EPITAXIAL GROWTH OF $SrBi_2Ta_2O_9$ THIN FILMS. Ho Nyung Lee, Dmitri N. Zakharov, Stephan Senz, Alain Pignolet, and Dietrich Hesse, Max-Planck-Institut für Mikrostrukturphysik, Halle/Saale, GERMANY; Peter Reiche and Reinhard Uecker, Institut für Kristallzüchtung, Berlin, GERMANY.

$SrBi_2Ta_2O_9$ is a well known ferroelectric material due to its high fatigue endurance, which is related to its bismuth layered perovskite structure. Moreover, the ferroelectric properties of $SrBi_2Ta_2O_9$ strongly depend on its crystallographic orientation because the spontaneous polarization is directed along the a axis of the highly anisotropic orthorhombic system. In order to study its basic ferroelectric properties, we have grown epitaxial thin films having different orientations on various single crystalline substrates of different orientations including silicon. Epitaxial $SrBi_2Ta_2O_9$ thin films having their c axis tilted 0°, 47°, 56°, and 90° from the normal to the film plane have been grown by pulsed laser deposition on complex oxide single crystal substrates. For the substrates, we have chosen lattice-matched substrates, such as (001)-, (011)-, and (111)-oriented $SrTiO_3$, (100)-oriented $SrPrGaO_4$, and (110)-oriented $SrLaGaO_4$, as well as non-lattice-matched Si(100) substrates. For the latter, heteroepitaxial buffer layers of $SrRuO_3$(110)/YSZ(100) and $SrRuO_3$(111)/MgO(111)/YSZ(100) have been deposited *in situ* on Si(100) substrates in order to grow (116)- and (103)-oriented $SrBi_2Ta_2O_9$ epitaxial thin films, respectively. A comparative analysis of the electrical properties for the various crystallographic orientations will be presented.

O6.6/C8.6

TEMPERATURE-DEPENDENT DIELECTRIC RESPONSE AND TUNABILITY OF $KTaO_3/KNbO_3$ SUPERLATTICES. Jennifer Sigman, David Norton, Rajiv Singh, and Josh Howard, Univ. of Florida, Dept. of Materials Science and Engineering, Gainesville, FL; Hans Christen, John Budai, Elliot Specht, Pam Fleming, and Lynn Boatner, Oak Ridge National Laboratory, Oak Ridge, TN.

In recent years, significant effort has focused on the manipulation of material properties through reduced dimensionality. Understanding size effects in ferroelectrics is useful in delineating local versus collective phenomenon in polarization behavior. In this talk, we will report on the temperature-dependent dielectric properties of $KTaO_3/KNbO_3$ superlattice structures grown by pulsed-laser deposition. This is an interesting system to consider as it provides a nearly lattice-matched perovskite multilayer structure in which the end compounds range from a paraelectric ($KTaO_3$) to a ferroelectric ($KNbO_3$) with T_c = 704 K. Previous work has shown that the structural transition associated with ferroelectricity is modified by strain and layer thickness in superlattice structures. In this talk, we will present temperature-dependent capacitance measurements using interdigitated capacitor structures fabricated on $KTaO_3/KNbO_3$ superlattice films. The peak in the dielectric response indicate the Curie temperature for a given superlattice structure. These measurements will then be compared with structural data obtained from temperature dependent x-ray diffraction. Comparison of superlattice dielectric response is also made with alloy thin film structures in which the composition corresponds to the average Ta/Nb ratio in the superlattices. We will also discuss relatively large voltage tunability of the dielectric response as a function of temperature both below and above the Curie temperature.

O6.7/C8.7

GROWTH MECHANISM OF PZT THIN FILMS EPITAXIALLY GROWN ON $SrRuO_3/SrTiO_3$ BY MOCVD. H. Nonomura, H. Fujisawa, M. Shimizu and H. Niu, Dept. of Electronics, Himeji Inst. Tech., Hyogo, JAPAN.

Investigation of the growth mechanism of PZT thin films is very important because very thin films with high quality are required for the realization of ferroelectric random access memories (FeRAMs) with high integration and low voltage operation. In this paper, the growth mechanism of MOCVD-PZT thin films epitaxailly grown on $SrRuO_3/SrTiO_3$ was investigated using scanning probe microscopy (SPM) and transmission electron microscopy (TEM). $SrRuO_3$ films were prepared at 550°C on $SrTiO_3$(100) by rf magnetron sputtering. Then PZT films were prepared on $SrRuO_3/SrTiO_3$ at 540°C by MOCVD using $(C_2H_5)_3PbOCH_2C(CH_3)_3$, $Zr(O\text{-}t\text{-}C_4H_9)_4$,

$Ti(O\text{-}i\text{-}C_3H_7)_4$ and O_2. In order to observe the initial growth stage of PZT films, they were grown for various deposition time, 10-120 sec. From SPM observations, at early initial growth stage, two dimensional layer growth was observed. Afterward three dimensional islands were observed and these gradually grew until finally they covered the surface. Surface roughness measured by SPM drastically increased after the appearance of PZT islands. These results suggest that the growth mechanism of PZT on $SrRuO_3/SrTiO_3$ is the Stranski-Krastanov growth mode.

O6.8/C8.8

STRUCTURE AND DIELECTRIC PROPERTIES OF EPITAXIALLY GROWN PMN THIN FILMS ON (100)MAGNESIUM OXIDES. Y. Yamada, M. Shimoda, R. Suzuki, T. Matsuda, A. Unno, K. Wasa, Yokohama City Univ, Faculty of Science, Yokohama, JAPAN.

Thin films of [Pb(Mg1/3, Nb2/3)]O3 (PMN) were grown on the (100)MgO substrates by rf-magnetron sputtering. The crystal structure and the dielectric properties were studied.The substrate temperature during the sputtering deposition was 600°C. The sputtered thin films were rapidly cooled to a room temperature after the deposition. The thickness of the PMN thin films were 200 to 400nm. The XRD analyses showed that the crystalline (100)PMN thin films of perovskite strucure were epitaxially grown on the MgO substrates without a pyrochlore phase. The in-plane XRD analysis indicated that the PMN thin films showed 3-dimensional epitaxy. A cross sectional TEM (transmission electron microscopy) image suggested that the PMN thin films were continuous single crystal-like structure. The dielectric properties of the PMN thin films were evaluated in the sandwich structure, Au/PMN/(100)Pt/ (100)MgO,with epitaxial (100)Pt thin films as a base electrode and vacuum evaporated Au thin films as a top electrode. A typical room temperature dielectric constant was 600 to 800 at 100kHz with a loss factor D=0.01 and a small frequency dispersion. The dielectric constant increased with a decrease of the temperature as expected. The PMN thin films showed P-E hysteresis curve at a room temperature with Pr=4 micro-Coulomb/cm^2 and Hc=125kV/cm.The Pr increased with the decrease of the temperature and Pr=15 micro-Coulomb/cm^2 was observed at 0°C.

O6.9/C8.9

FABRICATION AND PROPERTIES OF EXPITAXIAL LITHIUM NIOBATE THIN FILMS BY COMBUSTION CHEMICAL VAPOR DEPOSITION (CCVD). Yong Dong Jiang, Jake McGee, Todd A. Polley, Robert E. Schwerzel, Andrew T. Hunt, MicroCoating Technologies, Chamblee, GA.

Lithium niobate has been widely investigated because it has a great number of useful physical properties: excellent ferroelectricity and piezoelectricity, high electrooptic coefficient, and large photorefractive effect. These characteristics make lithium niobate an attractive material for electrooptic and acousto-optical applications such as waveguides, modulators, second harmonic generators, and transducers. In addition, erbium-doped lithium niobate can be used as active material for waveguide lasers. In this study, expitaxial lithium niobate thin films were deposited on c-sapphire (α-Al_2O_3) by the low-cost, open-atmosphere CCVD technique developed by MicroCoating Technologies, Inc. It was found that the process parameters, such as solution concentration and deposition temperature, play a critical role in determining the growth behavior of the lithium niobate thin films. In order to improve the quality and performance of lithium niobate, the effects of dopants and seed layers were investigated as well. XRD 2-Theta scans and pole figure analysis were utilized to determine the identities and relative ratios of the orientations present in the deposited films. Film surface morphology was examined using optical microscopy and SEM. Surface roughness was measured using optical profilometry, and film thickness and index of refraction were determined by spectroscopic ellipsometry.

O6.10/C8.10

STRUCTURAL AND FERROELECTRIC PROPERTIES OF $PbZr_{0.52}Ti_{0.48}O_3/SrRuO_3$ THIN FILMS ON $SrTiO_3$ SUBSTRATES. J. Rodriguez Contreras, U. Poppe, K. Szot, C.L. Jia, H. Kohlstedt, R. Waser, Forschungszentrum Jülich, Institut für Festkörperforschung, Jülich, GERMANY; J. Schubert, Forschungszentrum Jülich, Institut für Schichten und Grenzflächen, Jülich, GERMANY.

Lead based ferroelectric materials are currently the most promising candidate for use in non-volatile ferroelectric random access memory (FeRAM). We have deposited $PbZr_{0.52}Ti_{0.48}O_3/SrRuO_3$ (PZT/SRO) heterostructures on $SrTiO_3$ (STO) substrates using high-pressure on-axis sputtering. X-ray diffraction indicates that both PZT and SRO layers are c-axis oriented and have exact parallel orientations with the STO (100) substrate. No secondary phase contribution, e.g. pyrochlore phase in PZT films, has been observed. The PZT films possess a small mosaicity ($\Delta\omega < 0.1°$, $\Delta\Phi < 1°$). Whereas in the case of SRO films these values are $\Delta\omega < 0.1°$ and $\Delta\Phi < 0.4°$. Atomic Force Microscopy reveals very smooth surfaces in the order of 0.25 nm and 0.65 nm (RMS roughness) for SRO and PZT films respectively. High Resolution Transmission Electron Microscopy shows very homogenous interfaces without defects in 10 nm and 4 nm thick PZT films on SRO. The stoichiometry has been verified by Rutherford Backscattering Spectrometry. As can be seen by channeling measurements the SRO bottom electrode improves significantly the crystalline quality of the PZT film on top. Pt/PZT/SRO as well as SRO/PZT/SRO trilayers were patterned to form capacitors. Ferroelectric hysteresis loops has been obtained for PZT film thickness ranging from 90 nm down to 12 nm. For example a 23 nm thick PZT film on SRO bottom electrode using Pt top electrode shows a remanent polarization of 105 $\mu C/cm^2$ ($2P_r$) and an coercitive field of 1024 kV/cm ($2E_c$) (at 10 kHz). Generally the remanent polarization P_r decreases while the coercitive field E_c increases towards thinner PZT film thickness.

O6.11/C8.11

EFFECT OF A-SITE SUBSTITUTION ON THE MAGNETIC AND DIELECTRIC BEHAVIORS OF FERROELECTRIC $RMnO_3$ (R:YTTRIUM OR RARE EARTH ION) THIN FILMS. N. Fujimura, H. Sakata, D. Ito, T. Yokota and T. Ito, Department of Applied Materials Science, College of Eng, University of Osaka Prefecture, Sakai, Osaka, JAPAN.

Ferroelectric materials with magnetic properties might have several advantages when the materials use for memory devices. Hexagonal yttrium and rare earth manganites form an interesting class of materials known as ferroelectro-magnet in which the ferroelectric and magnetic orders coexist at low temperatures. We have demonstrated that $YMnO_3$ thin films show ferroelectric and antiferromagnetic behaviors and demonstrate a relationship between carrier concentration and magnetic properties of $YMnO_3$ films. However, little interaction was observed between the magnetic spin and dipole moment. This paper describes that the effect of A-site substitution on the magnetic properties of $YMnO_3$ and also demonstrates that the relationship between magnetic ordering and ferroelectric or transport property.

O6.12/C8.12

STRUCTURAL VARIANT CONTROL IN EPITAXIAL $SrBi_2Nb_2O_9$ THIN FILMS THROUGH GROWTH ON VICINAL SUBSTRATES. J. Lettieri, G. Asayama, Y. Jia, and D.G. Schlom, Penn State University, Dept of Materials Science and Engineering, University Park, PA.

The inherent low symmetry common to many multicomponent oxides often results in complicated structural variants or growth twins during heteroepitaxial growth. A number of these materials also undergo phase transformations during cooling after growth that can contribute to a very complex domain structures. Given the anistropy of these low symmetry oxides and the defects associated with variant boundaries, variant control is an important part of isolating and maximizing properties of heteroepitaxial oxides. Such is the case with the ferroelectric $SrBi_2Nb_2O_9$ whose highly anisotropic polarization makes variant control in heteroepitaxial films crucial. In this study we show structural variant control of (103) oriented $SrBi_2Nb_2O_9$ films on through growth on vicinal (111) $SrTiO_3$ substrates. The orientation relationships and resultant growth based on x-ray diffraction data will be discussed.

O6.13/C8.13

INVESTIGATION OF NUCLEATION AND GROWTH OF EPITAXIAL BARIUM TITANATE THIN FILMS BY MOVPE. D.J. Towner, B.W. Wessels, Department of Materials Science and Engineering, Northwestern University, Evanston, IL; J. Ni, T.J. Marks, Department of Chemistry, Northwestern University, Evanston, IL.

$BaTiO_3$ is being considered for thin film light wave devices. Epitaxial thin films with atomically smooth surfaces and interfaces are required for these applications. We have been developing low pressure metal-organic vapor phase epitaxy to meet this need. A two dimensional nucleation and growth process is required to obtain the requisite surface roughness. The fluorinated MOVPE precursor $Ba(hfa)_2$:PEB has been utilized as the barium source due to its low melting point (71°C), high volatility, and long-term stability. Optimization of the film quality is achieved through a controlled nucleation process, with temperature and reactant partial pressure as the variable parameters. Near atomically smooth $BaTiO_3$ films (as low as 0.89 nm for 275 nm thick) with excellent crystalline quality (rocking curve FWHM as low as 0.29 degrees) have been grown on MgO (100) substrates.

O6.14/C8.14
GROWTH AND ELECTRICAL PROPERTIES OF Fe DOPED (Ba,Sr)TiO_3 THIN FILMS DEPOSITED BY PULSED LASER DEPOSITION. Yoshiyuki Yonezawa, Megumi Kato, Yoshinori Konishi, Nobuhiro Okuda, Hideaki Matsuyama, Shinji Ogino, Noboru Furusho, Fuji Electric Corporate Research and Development, Ltd. Yokosuka, JAPAN; Toyohiro Chikyow, National Institute for Materials Science (NIMS), Tsukuba, JAPAN; Masashi Kawasaki, Institute for Materials Research, Tohoku University, Sendai, JAPAN; Hideomi Koinuma, Frontier Collaborative Research Center, Tokyo Institute of Technology, Yokohama, JAPAN.

(Ba,Sr)TiO_3 is a good candidate for gate oxides and integrated capacitors because of its high dielectric constant and absence of lead content. Consequently, direct epitaxial growths of BST on Si substrate have been studied extensively. However, there are problems with the formation of SiO_2 interlayer and leakage current due to stoichiometrical deviation. In particular, thin films deposited by Pulsed Laser Deposition require more than 100mtorr of oxygen partial pressure to get the required high break down voltage. To suppress the leakage current we tried to dope transition metals into the target, such as Mo, Mn, Cr, W and Fe. The target was prepared by sintering mixed powders of $SrTiO_3$, $BaTiO_3$ and 1mol% transition metal oxide. Nb-doped:$SrTiO_3$ was used for the substrate and the Pulsed Laser Deposition was used for the film fabrication. Substrate temperature was 600°C; oxygen partial pressure of 5mtorr and a post annealing process was performed under 1atm oxygen pressure. After the deposition film qualities were studied by atomic force microscopy(AFM), scanning electron microscopy (SEM) and x-ray diffraction (XRD). Pt upper electrodes were deposited on the films and the electrical properties were measured. Among the transition metals, the Fe-doped film had a significant effect on suppressing the leakage current. Subsequently, we changed the amount of Fe doping from 0.1mol% to 6%. Then we fabricate the film and compare the electrical properties. As a result, with post annealing, the sample with 4% Fe showed the lowest leakage current among those. While without post annealing, the sample with 6% Fe doped showed the lowest leakage current. As for the dielectric constants, it decreased as the doping increased. But at most only 20% of reduction was observed. XAFS results will be presented for estimating the local structure of the Fe ions.

O6.15/C8.15
FEMTOSECOND PULSED LASER DEPOSITION OF $BaTiO_3$ THIN FILMS. W. Tian[a], P.A. Van Rompay[b], Z.Y. Zhang[b], P.P. Pronko[b], J. Lettieri[c], D.G. Schlom[c], and X. Q. Pan[a]; [a] Department of Materials Science and Engineering, The University of Michigan, Ann Arbor, MI; [b]Center for Ultrafast Optical Science and Department of Electrical Engineering and Computer Science, The University of Michigan, Ann Arbor, MI; [c]Department of Materials Science and Enginerring, Penn State University, University Park, PA.

Considerable attention has recently focused on the use of femtosecond pulsed lasers for the fabrication of oxide thin films. The high intensity pulse of a femtosecond laser ($\sim 10^6$ order of magnitude higher than nanosecond laser), regardless of wavelength, is thought to have advantages over nanosecond laser for fabricating oxide thin films at relatively lower substrate temperature. We employed a femtosecond pulsed laser, with wave-length centered at 780 nm and pulse width 100 - 150 fs, to deposit $BaTiO_3$ thin film on the (001) $SrTiO_3$ substrate. The repetition rate of the laser is 10Hz. Processing parameters including target composition, oxygen partial pressure, substrate temperature, substrate-target distance, and diameter of the focused laser beam were varied to optimize the growth conditions. X-ray diffraction, Rutherford backscattering spectroscopy (RBS), x-ray photoelectron spectroscopy (XPS), TEM, and AFM were used to characterize the structure, microstructure, surface morphology, and chemical composition of the films. At the nominally optimum growth condition, x-ray diffraction studies indicate the formation of nearly phase-pure and epitaxial $BaTiO_3$ thin films, however, RBS and XPS studies reveal an overall off-stoichiometry of the films from the desired $BaTiO_3$. Moreover, the thin films grown by femtosecond laser exhibit much rougher surface than that of the films grown by nanosecond Excimer laser under similar growth conditions, which is most likely due to the non-stoichiometry nature of the films. Thus, the applicability of infrared (IR) femtosecond laser into complex oxide thin film deposition needs further investigation.

SESSION O7: HIGH-TEMPERATURE SUPERCONDUCTORS
Chairs: Jean-Marc Triscone and Xiaoxing Xi
Wednesday Morning, November 28, 2001
Room 203 (Hynes)

8:30 AM O7.1
HETEROEPITAXY OF $YBa_2Cu_3O_{7-\delta}/Pr_{1-x}Ca_xMnO_3$ ON $Al_2O_3(0001)$ SUBSTRATE FOR SUPERCONDUCTING MAGNETOSTATIC WAVE DEVICES. Hiroaki Nishikawa, Shigeki Hontsu, Masafumi Terakago, Shigenobu Mine, Kinki Univ, Faculty of Biology-Oriented Science and Technology, Wakayama, JAPAN; Masaya Nakamori, Kinki Univ-Technical College, Mie, JAPAN; Hitoshi Tabata, Tomoji Kawai, Osaka Univ, The Institute of Scientific and Industrial Research, Osaka, JAPAN.

We have noted heterostructure of high-temperature superconductor (HTS)/oxide magnetic materials because it is useful for "magnetic field controllable" superconducting microwave devices such as tunable microwave devices and, especialyy, magnetostatic wave devices. Previous investigations have used $Y_3Fe_5O_{12}$ (YIG) as the magnetic material of excellent magnetic and dielectric properties at the microwave frequency region. The problem was the large lattice mismatch of YIG to HTS. Thus, the HTS/YIG structure has been prepared by the contact of those materials. In order to realize the well-defined HTS/oxide magnetic material heterostructure, we have looked for a magnetic material which has excellent microwave properties and grow epitaxially with $YBa_2Cu_3O_{7-\delta}$ (YBCO). In this study, we show the possibilities of heteroepitaxy with the YBCO for various magnetic materials on $Al_2O_3(0001)$ substrate. The Al_2O_3 is one of the most ideal material as the substrate for the microwave devices because of its low loss tangent, high chemical stability and low cost. In this system, the magnetic materials also play a role as the buffer layer to prevent the interface between YBCO and Al_2O_3 from interdiffusion. The (0001) surface of Al_2O_3 is chosen since the ion configuration of that plane (close packing of oxygen) is similar to that of (113) plane for YBCO (close packing-like of yttrium, barium and oxygen). The single layered thin films and heterostructure with the YBCO of various magnetic materials such as perovskite type manganite $Pr_{1-x}Ca_xMnO_3$ (PCMO) have been prepared on $Al_2O_3(0001)$ and $SrTiO_3(111)$ substrates by pulsed laser deposition technique. The $SrTiO_3(111)$ substrate is used for the investigation of the [113] oriented YBCO properties. The YBCO/$SrTiO_3(111)$ shows the superconductivity with the T_c of 89 K. Furthermore, the T_c of the YBCO grown on the PCMO buffered $Al_2O_3(0001)$ is 88 K. This means that we can get high quality YBCO/PCMO heterostructure on $Al_2O_3(0001)$ substrate.

8:45 AM *O7.2
INVESTIGATIONS OF MICROSCOPIC MAGNETIC PROPERTIES OF HIGH-TEMPERATURE SUPERCONDUCTORS AND MAGNETIC SYSTEMS WITH POSITIVE MUONS. Hugo Keller, Physik-Institut der Universität Zürich-Irchel, Zürich, SWITZERLAND.

The muon-spin rotation (μSR) technique is a unique and powerful microscopic magnetic probe for the study of magnetic properties of superconductors and magnetically ordered systems. In many cases this rather exotic method has provided important information on the magnetic properties of these materials, which are hardly obtained with any other experimental technique. In the μSR method the spin-polarized positive muon is used as a sensitive *microscopic* probe of internal magnetic fields in the *bulk* of a specimen. The internal fields can be observed by means of muon-spin precession or relaxation. In particular, the complex vortex structure in high-temperature superconductors can be explored with this technique. As an example, μSR experiments on the vortex phase diagram (flux-lattice melting, 3D-2D crossover, vortex fluctuations) of single-crystal BSCCO are described. So far μSR studies have been performed using muons with energies in the MeV range, which require rather thick samples. Recently, a novel μSR technique using low-energy muons was developed at the Paul Scherrer Institute (PSI), Switzerland [Morenzoni *et al.*, J. Appl. Phys. **81**, 3340 (1997)]. These ultra-slow muons of tunable energy (between 10 eV and 30 keV) can be implanted at very small and controllable depths in the sample to be investigated. This opens completely new possibilities to study microscopic magnetic properties of thin magnetic (antiferromagnetic) and superconducting films, as well as of multi-layer superconductors and multi-layer magnetic systems near the surface of the sample with a few nanometers' resolution. First results obtained with this new technique on a thin YBCO film are presented.

9:15 AM *O7.3
EPITAXIAL SUPERCONDUCTING OXIDE FILMS: STRAIN ENHANCED PAIRING AND EPITAXY PRODUCED NEW MATERIALS. Michio Naito, Hisashi Sato, Shin-ichi Karimoto, Hideki Yamamoto, and Akio Tsukada NTT Basic Research Laboratories, Atsugi-shi, Kanagawa, JAPAN.

Heteroepitaxy provides positive effects on superconducting oxide films. One example is T_c enhancement by epitaxial strain. On $LaSrAlO_4$ substrates, compressive epitaxial strain enhances T_c of $(La,Sr)_2CuO_4$ to 44 K and T_c of $(La,Ba)_2CuO_4$ to 47 K along with complete removal of the 1/8 anomaly. As means of understanding these phenomena microscopically, we point out two empirical trends

that we have found in hundreds of either compressively or expansively strainded LSCO and LBCO films. The first is that a larger c-axis lattice constant gives higher T_c, and the second is that smaller residual resistivity gives higher T_c. As regards the latter, our recent magnetotransport results indicate that the residual resistivity is dominantly governed by Kondo scattering due to Cu^{2+} spins. These results suggest the following scenario: spins are poison to high-T_c as well as to low-T_c. New materials produced by heteroepitaxy is another example of the positive effects. Since the discovery of high-T_c superconductors, most of the efforts to search for new superconductors have been made by a conventional or high-(oxygen)-pressure bulk synthesis. At present, however, the discoveries of new superconductors by bulk synthesis are getting fewer and fewer. So new synthetic routes are required as the next strategic material search. Since 1996, we have started material search by molecular beam epitaxy (MBE). The key features of MBE synthesis are low-temperature synthesis, hetero-epitaxy, atomic layer control, contamination-free environments, and strong uniform oxidation with ozone. This approach has produced new superconductors such as $Sr_2CuO_{4-\delta}$ (T_c~70 K), $Ba_2CuO_{4-\delta}$ (T_c~90 K), $PbSr_2CuO_{5\delta}$ (T_c~40 K), and $(La,Ce)_2CuO_4$ (T_c~30 K).

9:45 AM O7.4
EPITAXIAL STRAIN EFFECTS ON SUPERCONDUCTING AND TRANSPORT PROPERTIES OF $La_{2-x}Sr_xCuO_{4+\delta}$ THIN FILMS. X.X. Xi, X.H. Zeng, Weidong Si, Z.M. Stum, A. Sukiassyan, Department of Physics, Pennsylvania State University, University Park, PA.

We have studied the epitaxial strain effects on superconducting and transport properties of pulsed-laser-deposited $La_{2-x}Sr_xCuO_{4+\delta}$ thin films. The epitaxial strain is controlled by using $SrLaAlO_4$ buffer layers of different thicknesses on $SrTiO_3$ substrates. The La_2CuO_4 films are insulating under tensile epitaxial strain, but when the strain is sufficiently compressive they become superconducting. The epitaxial-strain-induced insulator-superconductor transition suggests a significant interplay between strain and oxygenation in the films. The Hall coefficient of films with Sr concentration over the range of $0 \leq x \leq 0.30$ was studied. We found that in undoped thin films, compressive epitaxial strain leads to lower Hall coefficient, thus higher carrier density. This is consistent with that compressive strain help the insertion of interstitial oxygen. The down turn in the Hall coefficient, which has been suggested to indicate a transition to the static strip phase, has not been observed in $La_{2-x}Sr_xCuO_{4\delta}$ films with $x = 1/8$, with different epitaxial strain, oxygenation, and Nd doping. This may suggest that the static stripes are suppressed in the thin films.

10:30 AM *O7.5
SPIN INJECTION AND COHERENT TRANSPORT AT DOUBLE SUPERCONDUCTOR-FERROMAGNETIC INTERFACES (F/S/F). Denis Feinberg, R. Mélin, CNRS Grenoble, FRANCE; Guy Deutscher, Univ. of Tel Aviv, ISRAEL; G. Falci, Univ. of Catane, ITALY; F.W.J. Hekking, Univ. of Grenoble, FRANCE.

At a good superconductor-metal contact, Andreev reflections convert a Cooper pair current into a normal current. This proceeds through reflection of an electron into a hole with opposite spin. Therefore this mechanism tends to be blocked if the metal is ferromagnetic. We show here that multiple F/S interfaces offer new current channels: for instance the Cooper pair current can be coherently shared between neighbouring F electrodes if they have antiparallel spin polarizations (SP). On the other hand, if the SP's are parallel, elastic cotunneling from a F electrode to the other can occur through the super-conductor. These coherent effects decay on the superconductor's coherence length. As a consequence, mesoscopic F/S/F interfaces may open a variety of applications ranging from SP measurement at the atomic scale (Spin STM) to spin injection. The progress in nanofabrication and increasing control of ferromagnetic nanoparticles thus offers wide perspectives, using such F/S hybrid structures.

11:00 AM *O7.6
MBE GROWTH OF CUPRATE MULTILAYERS FOR HIGH-Tc ELECTRONICS. I. Bozovic, G. Logvenov, J. Sveklo, B. Narimbetov, I. Belca, M. Verhoeven, L. Caputo and E. Goldobin, Oxxel GmbH, Technologiepark Universitaet, Bremen, GERMANY.

We have used shuttered molecular beam epitaxy (MBE) to deposit thin films of cuprate superconductors (including (La,Sr)CuO and several BiSrCaCuO phases) and other complex oxides, on $SrTiO_3$ and $LaSrAlO_4$ substrates. Heteroepitaxy has been studied by characterizing in detail hundreds of bare substrates, buffer layers, and HTS films, in situ using RHEED and TOF-ISARS (time-of-flight ion scattering and recoil spectroscopy, which enables monitoring of the chemical composition of surface layers), and ex situ by AFM and XRD. Under optimal deposition conditions, the HTS films emerge essentially atomically smooth (rms surface roughness down to 0.1 -0.2 nm), except for occasional steps due to slightly vicinal substrates. This has enabled growth of various multi-layered hetero-structures and superlattices, and fabrication of HTS trilayer (sandwich) junctions. Apart from practical interest, these devices allow one to study fundamental problems such as the existence (or not) of the so-called Giant Proximity Effect. By virtue of atomic layering and/or epitaxial stabilization, it has been possible also to synthesize some metastable phases which cannot be produced by the conventional bulk methods, such as $Bi_2Sr_2Ca_3Cu_4O_{12}$ (superconducting, with Tc above 60 K) and $SrMnO_3$ in the perovskite structure. In the case of (La,Sr)CuO, the data allow for an assessment of the direct dependence of Tc on the epitaxial strain in contrast to the effect of chemical doping by incorporation of extra oxygen.

11:30 AM O7.7
ELECTROSTATIC MODULATION OF THE HALL RESPONSE IN THIN $NdBa_2Cu_3O_{7-\delta}$ FILMS. S. Gariglio, D. Matthey, J.-M. Triscone, DPMC, University of Geneva, Geneva, SWITZERLAND; C.H. Ahn, Dept. of Applied Physics, Yale University, New Haven, CT.

We have used the ferroelectric field effect in hetero-structures based on superconducting $NdBa_2Cu_3O_{7-\delta}$ and ferroelectric $Pb(Zr_{0.2}Ti_{0.8})O_3$ to electrostatically modulate in a reversible and nonvolatile fashion the carrier density of $NdBa_2Cu_3O_{7-\delta}$ superconducting films. Reversing the ferroelectric polarization induces a constant relative change in the resistivity and inverse Hall constant of 9% and -6%, respectively, at all temperatures above the superconducting transition. These results suggest a resistivity and Hall constant that are both inversely proportional to a temperature independent mobile carrier density. The cotangent of the Hall angle displays a T^2 temperature dependence with a slope that depends on the polarization direction. This behavior can be explained by a modification of the effective mass induced by the change in the carrier density.

11:45 AM O7.8
FERROELECTRIC FIELD EFFECT INDUCED MODULATION OF THE CRITICAL TEMPERATURE AND PENETRATION DEPTH IN THIN $NdBa_2Cu_3O_{7-\delta}$ FILMS. Daniel Matthey, Stefano Gariglio, J.-M. Triscone, Dept of Condensed Matter Physics, Univ of Geneva, SWITZERLAND; C.H. Ahn, Dept. of Applied Physics, Yale Univ, New Haven, CT.

In epitaxial ferroelectric $Pb(Zr,Ti)O_3$ (PZT) / $NdBa_2Cu_3O_{7-\delta}$ (NBCO) heterostructures grown by RF magnetron sputtering, we have used the ferroelectric field effect to modulate the carrier density of thin superconducting oxide films. In lithographically patterned structures, reversing the polarization field of the ferroelectric PZT layer allows a non-volatile and reversible modification of the critical temperature of the superconducting film. Using the resistivity versus temperature data, we performed a detailed analysis of the superconducting transitions (for the two polarization states) of an 80 Å thick film, following the 2-D Coulomb gas scaling approach of Minnhagen[1], which has been used to describe vortex fluctuations in oxide superconductors above T_c. Using this model, we extracted the mean field transitions T_{co} and the Kosterlitz-Thouless critical temperatures T_{KT}. For the film investigated, the T_{co} values are 78.2 K and 78.6 K for the two polarization states, respectively, the higher T_c being for the polarization direction that adds holes to the channel. The corresponding Kosterlitz-Thouless transitions are 58.2 K and 59.3 K. We estimated the zero temperature penetration depths from these data and from measurements of the activation energies in the liquid vortex phase, using a 2-dimensional model for vortex dynamics. The change in penetration depth induced by the polarization reversal will be discussed and correlated with the modification observed in the inverse Hall constant[2].
[1] For a review, see P. Minnhagen, Rev. of Mod. Phys. 59, 1001 (1987).
[2] S. Gariglio et al., this conference.

SESSION O8: MAGNETIC OXIDES
Chairs: Allen M. Goldman and Hitoshi Tabata
Wednesday Afternoon, November 28, 2001
Room 203 (Hynes)

1:30 PM *O8.1
HETEROEPITAXY OF LAYERED STRUCTURES: THE CASE STUDY OF HEXAGONAL BARIUM FERRITE. L.V. Saraf[a], S.M. Bhagat[b] and R. Ramesh[a,b], [a]Department of Materials Science and Engineering, University of Maryland, College Park, MD; [b]Department of Physics, University of Maryland, College Park, MD.

Multicomponent oxides present materials systems rich in structural chemistry and with a broad range of functional responses. Creation of novel, multilayered heterostructures using combinations of these materials continues to be a strong focus of research worldwide, with a strong emphasis on perovskite structures as prototypical building blocks. The layered ferrites, with a hexagonal crystal structure present

another class of materials with technologically valuable properties, especially for microwave applications and in magnetic recording. Creation of highly single crystalline model thin film systems out of these materials is a central theme of our research. Hexagonal barium ferrite (BaM) films on (0001) oriented Al_2O_3, reflect the strong effect of lattice mismatch induced strains. The use of a nonferromagnetic buffer dramatically alters the ferromagnetic and resonance properties of the film. The narrowest FMR lines and largest number of spin wave resonances are obtained when the field is perpendicular to the film plane of 0.3 and 0.46 μm films. To quantitatively account for variations with field angle (θ) we assume (i) a strain induced anisotropy energy with $\cos^4\theta$ dependence and (ii) that the film should be pictured as a mosaic whose tiles are tilted out of the film plane. The effect of substrate induced lattice strains on the structural and magnetic properties are also studied in which the broadening of the resonance absorption is observed in the presence of strain.
This work is supported by the Office of Naval Research

2:00 PM O8.2
ROOM TEMPERATURE SPIN(CLUSTER)-GLASS AND ITS PHOTO-CONTROL IN SPINEL FERRITE FILMS. Yuji Muraoka, Hitoshi Tabata, Hiromasa Saeki, Tomoji Kawai, Osaka Univ, ISIR-Sanken, Osaka, JAPAN.

Room temperature spin-glass states have been found in (Mg,Fe)Mg,Fe,Ti_2O_4, (Co,Zn)FeO_4 and (Ru.Al)FeO_4 spinel ferrite thin films. Although spin-glass has been regarded for a long time as a low temperature phenomenon, below 50 K, room temperature spin-glass has been achieved by the fine-tuning of spin states through the control of composition, random oxygen deficiencies and the stress induced by the film/substrate lattice mismatch in hetero-epitaxyal films. All of which give rise to effects that enhance the exchange interaction of spins in the ferrite films. We have demonstrated the change of magnetic state by means of light-irradiation from spin-glass to a ferri-magnet over a wide range of temperatures up to 300 K. The direct photo-excitation of spins with photon energy around 2.0 eV, which corresponds to the spin excitation energy for Fe ions, is the most effective for realizing photo-induced magnetization.

2:15 PM O8.3
STUDY OF FERRITE THIN FILMS AND FERRITE BASED SPIN FILTER DEVICES. Guohan Hu, Y. Suzuki, Cornell Univ, Dept of Materials Science and Engineering, Ithaca, NY; V.G. Harris, Naval Research Laboratory, Washington, DC.

Epitaxial spinel structure ferrite thin films have been studied because of their unique magnetic and transport properties. Our studies on highly magnetostrictive $CoFe_2O_4$ (CFO) films grown on MgO and on $CoCr_2O_4$ buffered $MgAl_2O_4$ substrates demonstrate that microstructure, such as anti-phase boundaries, cation distribution and lattice mismatch strains, plays a critical role in determining the magnetic properties of the films. In particular, by annealing the CFO films and hence varying the cobalt cation distribution among the tetrahedral and octahedral sites and the strain state of the films, we have been able to tune the symmetry and the magnitude of the magnetic anisotropy. Based on our studies of CFO films, we have fabricated a magnetoresistive tunnel device on (110) oriented $SrTiO_3$ substrate, which is composed of a highly spin polarized (LaSr)MnO_3 (LSMO) electrode, a highly insulating CFO tunnel barrier and a normal metal counter electrode. The magnetic tunnel barrier provides a spin filter effect depending on the relative orientation of the spins in the LSMO and CFO layers. We observe tunneling behavior in junctions with barriers up to 100$\AA$ thick and spin dependent magnetoresistive behavior.

2:30 PM O8.4
STRUCTURE AND MAGNETIC PROPERTIES OF SPINEL $CoFe_2O_4$ ON (111) MgO. S.D. Yoon, C. Vittoria, Department of Electrical and Computer Engineering, Northeastern University, Boston, MA; S.A. Oliver, Center for Subsurface Sensing and Imaging Systems, Northeastern University, Boston, MA.

Highly oriented films of (111) cobalt ferrite ($CoFe_2O_4$) were deposited on single crystal (111) magnesium oxide substrates by pulsed laser deposition (PLD), and analyzed for their crystallographic and magnetic properties. All the $CoFe_2O_4$ films were grown under identical deposition parameters except for the substrate temperature, which was varied from 400 to 925°C. All films have excellent (111) orientations with cubic lattice parameters matched that of bulk $CoFe_2O_4$ for the film grown at 700°C, and was smaller for films deposited at lower and higher temperatures. This variation in lattice constant was used to deduce the film strain, which affects the film magnetic properties through magnetostriction. Films grown at substrate temperatures T_s-d500°C have smaller lattice dispersions than films grown at T_s<500°C. Hysteresis loop and torque magnetometer results show that the magnetic behavior was different for measurements out of the film plane and in the film plane, for films deposited at T_s-d700°C. Films grown at lower temperatures have magnetically isotropic. A small in-plane uniaxial anisotropy was noticed for the T_s=925°C sample. The coercive force along the film normal increased from H_c=0.42kOe at 925°C to H_c=3.03kOe at 400°C. The increases in H_c and lattice dispersion for low temperature films imply that these samples are oriented polycrystalline films with reduced grain size. The measured anisotropy values are lower than those seen on (100) and (110) MgO substrates [1]. [1] Y. Suzuki, G. Hu, R.B. van Dover, R.J. Cava, JMMM 191 (1999) 1.

3:15 PM *O8.5
STABILIZATION OF METASTABLE RARE-EARTH MANGANESE OXIDES AS THIN FILMS. Paul Salvador, Andrew Francis, Arati Bagal, Oscar Castillo, Balasubramanian Kavaipatti, Carnegie Mellon University, Dept of Materials Science and Engineering, Pittsburgh, PA.

Although heteroepitaxial growth of complex oxides is now well known as an approach to synthesizing metastable materials, phase stability during heteroepitaxial growth of specific systems is still only partially understood. The $RE$$MnO_3$ system, where RE is a rare earth or yttrium, is ideal for developing a better fundamental understanding of this complicated problem. $RE$$MnO_3$ materials are single phase across the entire RE series and they adopt one of two possible structures, the other structure being metastable. Compounds that contain the smaller rare-earth cations form in a hexagonal crystal structure and have interesting ferroelectric properties. Compounds of the larger rare-earth cations form in perovskite-related structures that have interesting magnetic properties. In this work, the factors that lead to the stabilization of the metastable phase as a heteroepitaxial film are investigated for the entire rare-earth series. We first consider the various thermodynamic factors that lead to phase selection during nucleation of an epitaxial thin film, including the formation energy of both phases as a function of rare-earth cation, the various interfacial energies, and the misfit strain energy. By varying the type and crystallographic orientation of the substrate, one can influence the overall interfacial and/or the strain energy contributions to phase stability, thereby favoring metastable phase formation. $RE$$MnO_3$ compounds across the entire RE series have been deposited by pulsed laser deposition on a wide range of substrates, including $SrTiO_3$, $LaAlO_3$, $NdGaO_3$, MgO, $RE$$MnO_3$, Dy_2CuTiO_6, Pt, SiC, and GaN. By doing so, the contributions of each thermodynamic factor can be assessed with respect to its efficacity for metastable film formation. Particular attention is paid to the synthesis and characterization of the larger rare-earth compounds in their metastable hexagonal structure because they have not been synthesized to date by any method. Structural and physical property characterization of these films will also be discussed.

3:45 PM O8.6
SELF-ORGANIZATION OF $SrRuO_3$ THIN FILMS THROUGH SWITCHING OF SURFACE TERMINATION. Guus Rijnders and Dave H.A. Blank, Low Temperature Div, MESA Research Institute & Dept. of Applied Physics, Univ of Twente, Enschede, THE NETHERLANDS; Junghoon Choi and Chang-Beom Eom, Dept of Materials Science and Engineering, Univ of Wisconsin-Madison, Madison, WI.

Using a unique growth monitoring system, we were able to study the growth of $SrRuO_3$ on TiO_2-terminated $SrTiO_3$. The system used is reflection high energy electron diffraction (RHEED) operating at unusual high deposition pressure. The experimental data show that during the initial growth not the expected RuO_2 but SrO becomes the termination layer. This change in surface termination is accompanied by a change in growth mode, attributed to a change in mobility of ad-atoms. This mode can be described as a combination of step-flow and 2-D layer by layer, where islands are created and subsequently amalgamate by coalescence with the advancing step edges. During the initial growth this leads to roughening of the step edges. This roughening continues until a steady state is reached in the step density, in such way that the average distance of the deposited atoms to a step edge becomes comparable to the migration length. Therefore, this transition depends on the average terrace width of the vicinal substrate as well as the temperature during deposition and can be seen as a self-organization of the surface morphology. This change in growth mode will lead to single domain films, a requisite for the fabrication of ferromagnetic tunnel junctions.

4:00 PM O8.7
MAGNETORESISTANCE IN COLOSSAL MAGNETORESISTANCE THIN FILMS AND TRILAYERS. Lisa Berndt, Dept of Applied and Engineering Physics, Cornell Univ, Ithaca, NY; Yuri Suzuki, Dept of Materials Science and Engineering, Cornell Univ, Ithaca, NY.

The correlation of the electronic and magnetic properties of colossal magnetoresistance (CMR) materials with structural properties has been the focus of research over the last few years. We have focused on

tensilely strained epitaxial thin films of the CMR material $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) on (001) and (110) oriented $SrTiO_3$ substrates. Our work relating biaxial strain states to magnetic anisotropy has shown that the magnetic anisotropy of CMR films is dominated by the strain or magnetocrystalline anisotropy depending on the orientation of the film. Trilayer films were made with two LSMO layers separated by of a 20 - 100 angstrom thick layer of $La_{0.7}Ca_{0.3}MnO_3$ (LCMO), which is insulating and paramagnetic at room temperature. LCMO was chosen for the middle layer in order to reduce interface strain. Room temperature magnetotransport measurements of trilayer films suggest that the LSMO layers exhibit exchange or dipole coupling depending on the thickness of the LCMO layer.

4:15 PM O8.8
STRAIN RELEASE OF $La_{0.67}Ca_{0.33}MnO_3$ GROWN ON $SrTiO_3$. Z.Q. Yang, R. Hendrikx, J. Aarts, Kamerlingh Onnes Laboratory, Leiden University, THE NETHERLANDS; Y. Qin, H.W. Zandbergen, National Center for High-Resolution Electron Microscopy, Delft University of Technology, THE NETHERLANDS.

Films of ferromagnetic metallic $La_{0.67}Ca_{0.33}MnO_3$ (L) have a wide range of possible applications, both due to their 'Colossal Magneto-Resistance' properties and their large spin polarization. When grown epitaxially on $SrTiO_3$ (S), there is by now convincing evidence that the strain imposed by the substrate leads to appreciable lowering of the Insulator-Metal transition temperature. The issue is clowded, however, by the fact that 'bulk-like' properties can also be found, depending on deposition technique and conditions. Here we elucidate several of these points, using sputter-deposited films and combining electron microscopy, AFM and transport data. Firstly, we show that strain is present and can be released very effectively by first depositing a thin (5 - 50 nm) template layer of $YBa_2Cu_3O_x$ (Y); the IM-transition returns to its bulk value, as do both the in-plane and out-of-plane lattice parameter. We also compare three-layer samples S/L/Y/L, with a four-layer sample S/Y/L/Y/L. The three-layer sample clearly shows two differently strained L-layers with different properties, while the four-layer sample has two fully equivalent L-layers. Secondly, we show the effects of different growth rates. Fast growth leads to substantially higher IM transition temperatures, but this is not due to strain release by dislocations. The in-plane lattice parameters are still those of the substrate. However, fast growth leads to rougher surfaces than slow growth, indicating relaxation at the growing surface. It may be speculated that the accompanying inhomogeneities in the film would be favourable for phase separation phenomena.

4:30 PM O8.9
PROPERTIES OF CHARGE-ORDERED $Pr_{0.5}Ca_{0.5}MnO_3$ FILMS ON $SrTiO_3$. Z.Q. Yang, R.W.A. Hendrikx, J. Aarts, Kamerlingh Onnes Laboratory, Leiden University, THE NETHERLANDS; H.W. Zandbergen, National Center for High-Resolution Electron Microscopy, Delft University of Technology, THE NETHERLANDS.

In bulk $Pr_{0.5}Ca_{0.5}MnO_3$ (PCMO), the transition from a paramagnetic insulator to the charge-ordered (CO) state by the ordering of the Mn^{3+} and Mn^{4+} ions leads to a strong orthorhombic distortion of the lattice unit cells with two elongated axes and one compressed axis. For the bulk material the melting magnetic field of the CO states is around 30 T at 5 K. Using X-ray diffraction, magnetization, transport measurement, and PCMO films grown on $SrTiO_3$ by sputtering, we show that at small thickness (25 nm) the very strained films still have high melting fields, of the order of 20 T. With increasing film thickness, the strain relaxes but this does not lead to more bulk-like behavior (as found in e.g. $La_{0.67}Ca_{0.33}MnO_3$ on STO). The CO melting field of an almost strain-free film has reduced to 8 T at 5 K, with a strong increase in the hysteretic behavior. Apparently, the square symmetry imposed by the substrate hinders the formation of the orthorhombic structure and the CO-state, in a manner similar to what was found for chromium-doped PCMO which shows ferromagnetic metal behavior. As a result, the relaxation of the substrate-induced strain promotes the formation of the ferromagnetic domains, and suppresses rather than enhances the CO state.

4:45 PM O8.10
PHASE SELECTIVE CHEMICAL VAPOR DEPOSITION OF CHROMIUM OXIDES FROM THE DECOMPOSITION OF CHROMYL CHLORIDE. Young-Nam Cho, William J. DeSisto, University of Maine, Dept. of Chemical Engineering and Laboratory for Surface Science and Technology; Sofian Kanan, Carl Tripp, Dept. of Chemistry and Laboratory for Surface Science and Technology, Orono, ME.

Ultrathin films of highly spin-polarized materials are central to many magneto-electronic devices. An efficient and controlled chemical vapor deposition (CVD) process for depositing highly spin-polarized, metastable chromium dioxide (CrO_2) on single crystal TiO_2 substrates has been developed using chromyl chloride (CrO_2Cl_2) as a precursor. This precursor is a liquid at room temperature with a vapor pressure adequate for CVD using conventional precursor handling equipment. The films were grown at atmospheric pressure in a horizontal reactor. The films were shiny, black, conducting and approximately 200 nm thick. Thin, epitaxial films of metastable CrO_2 in (100) and (001) orientation have been stabilized by (100) and (001) TiO_2 single crystal substrates as evidenced by x-ray diffraction θ-2θ scans. X-ray diffraction ϕ-scans on the CrO_2 (110) reflection indicated the expected two-fold symmetry, with no evidence of misaligned material. The growth of epitaxial CrO_2 proceeded in either pure O_2 or Ar atmospheres at temperatures between 360 and 400°C. In contrast, thin epitaxial films of the more stable Cr_2O_3 in (006) orientation, were formed when using (0001) Al_2O_3 single crystal as a substrate material over the same deposition temperature range. At temperatures above 400°C, (110) Cr_2O_3 was formed on (100) TiO_2. A crossover from kinetically limited growth to mass transfer limited growth occurred at approximately 400°C for both substrates. The chemically driven substrate selectivity was examined using Infrared and Raman spectroscopy, and correlated to the thin film deposition experiments. Additional materials characterization consisted of surface morphology, electrical and magnetic measurements, and spin-polarization measurements.