

SYMPOSIUM BB

Characterization and Modeling of Domain Microstructures in Materials

November 27 – 29, 2000

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* Invited paper

SESSION BB1: DOMAIN-RELATED PHENOMENA
IN FERROIC MATERIALS I

Chairs: Avadh B. Saxena and Gerhard R. Barsch
Monday Morning, November 27, 2000
Room 201 (Hynes)

8:30 AM *BB1.1
STATICS AND DYNAMICS OF MARTENSITIC INTERFACES.
Gerhard R. Barsch, Penn State University, Dept of Physics and
Materials Research Laboratory, University Park, PA.

In this review an account of the contribution will be given that Landau-Ginzburg models have made so far to the understanding of the static and dynamic properties of domain walls and domain configurations in proper and improper ferroelastic martensites. Following a recapitulation of the conceptual and mathematical basis underlying the mesoscopic mean-field approach, theoretical results for the structure, energy and mobility of domain walls, twin bands and pretransformation modulations (tweed) and of their effect on elastic and phonon properties are discussed and compared with available experimental data. In addition, the relevance of these results and of the theoretical method for understanding the martensitic nucleation mechanism and the hysteresis in shape memory applications will be discussed. In conclusion, a listing of open questions will be presented.

9:00 AM *BB1.2
PREMARTENSITIC MICROSTRUCTURE IN FERROMAGNETIC
Ni-Mn-Ga SHAPE-MEMORY ALLOYS. Antoni Planes, Lluís
Mañosa, Teresa Castán, Eduard Vives, Univ Barcelona, Dept ECM,
Barcelona, Catalonia, SPAIN.

Ni-Mn-Ga alloys close to the stoichiometric Heusler composition are, at present, the only known ferromagnetic alloys which exhibit shape-memory properties associated with a thermoelastic martensitic transformation. The transformation takes place from a high-temperature $L2_1$ structure towards a tetragonal low-temperature phase. For several compositions, the martensitic transition is preceded by a transition towards a modulated microstructure resulting from the condensation of the $1/3(110)TA_2$ phonon. We will show that the coupling between vibrational and magnetic degrees of freedom is the key ingredient which gives rise to such a condensation. Experimental results including elastic constants, phonons and magnetic measurements will be presented that are aimed at (i) corroborating the existence of spin-phonon coupling and (ii) characterizing the premartensitic transition. These results will be discussed within the framework of a lattice model which contains structural and magnetic variables. Numerical simulations of the model show that some anomalies observed in the specific heat and magnetic susceptibility originate from large amplitude fluctuations. The main conclusion is that an adequate combination of magnetoelastic coupling and phonon softening is the essential feature for the premartensitic transition to occur.

9:30 AM *BB1.3
DOMAIN STRUCTURES AND INTERFACES IN Ni-Al
MARTENSITE: COMPARING HRTEM OBSERVATIONS WITH
CONTINUUM THEORIES. Dominique Schryvers, Philippe Boullay,
Pavel L. Potapov, EMAT, University of Antwerp, RUCA, Antwerp,
BELGIUM; John Ball, Mathematical Institute, University of Oxford,
Oxford, UNITED KINGDOM.

Domain structures and interfaces formed during the cubic-to-tetragonal martensitic transformation in Ni-rich Ni-Al are investigated experimentally using HRTEM. The effects of different materials preparation methods, including homogenised bulk, splat-cooled discs and melt-spun ribbons will be presented. The observed micro- and nanostructural configurations are interpreted in view of possible formation mechanisms. For interplate interfaces a distinction is made between cases in which the microtwins, originating from mutually perpendicular $\{110\}$ austenite planes, enclose a final angle larger or smaller than 90° , as measured over the boundary. Two different configurations, one with crossing microtwins and the other with ending microtwins producing a step configuration are described. The latter is related with the existence of microtwin sequences with changing volume fractions. Although both features appear irrespective of the materials preparation technique, rapid solidification seems to prefer the step configuration. Depending on the actual case, tapering, bending and tip splitting of the small microtwin variants is observed. Severe lattice deformations and reorientations occur in a region of 5-10 nm around the interface while sequences of single plane ledges gradually bending the microtwins are found up to 50 nm away from the interface. These structures and deformations are interpreted in view of the need to accommodate any remaining stresses. The measured parameters such as volume fraction, directions of rigid body rotations and relative angles between microtwins are compared with predictions of continuum theory assuming energy minimising configurations. For melt-spun material, local nanoscale

inhomogeneities yield different microstructures and result in a wide temperature range for the transformation process. The latter are documented by EDX, EELS, X-ray and DSC measurements.

10:30 AM *BB1.4
DOMAINS AND MULTISCALE MICROSTRUCTURE IN ELASTIC
MATERIALS. Turab Lookman, Los Alamos National Lab., Los
Alamos, NM.

Martensitic structural transitions, especially those with unit cells related by continuous deformations, exhibit a rich variety of temperature/stress induced domains. We present a unified understanding of twins, tweed, nucleation pathways and stress loading for the martensitic transition, from a mesoscopic (Ginzburg-Landau) free energy entirely in strain variables. The strain description demands that the elastic compatibility constraint be satisfied. The important consequence of this is a long-range, anisotropic interaction in the order parameter strain components that mediates different elastic textures under different thermodynamic conditions. For instance, local stress inhomogeneities can effect structures on a global scale. The formalism is readily applied to all major crystal systems in 2D and 3D. We will illustrate it for (a) structural transitions of the square and triangular lattices in 2D and (b) cubic to tetragonal and cubic to trigonal transformations in 3D. We show how these ideas are generalized to ferroelectrics, magnetoelastics and many other related materials where strain is a secondary order parameter. The experimental observation and role of various microstructures on material specific functionalities will be emphasized. Collaborators: K. Rasmussen, A. Saxena, A.R. Bishop, R.C. Albers (Los Alamos) and S.R. Shenoy (ICTP, Trieste).

11:00 AM BB1.5
X-RAY AND TEM INVESTIGATIONS ON SINGLE CRYSTALS OF
Ni₂MnGa SHAPE MEMORY ALLOY. Christina Wedel, Kimio
Itagaki, Tohoku University, Institute for Advanced Materials
Processing, Sendai, JAPAN; Boris Wedel, DOWA Mining Co. Ltd.,
Central Research Laboratory, Hachioji, JAPAN.

The ferromagnetic Heusler alloy Ni₂MnGa and the non-stoichiometric alloys from the same phase have recently attracted attention. They are very promising materials for several applications, since the magnetic properties make it possible to control the martensitic transition with a magnetic field. Several research projects focused on this subject. The weak point in the research of this material was the confusion about the crystal structure. So far it was known that Ni₂MnGa undergoes a thermoelastic martensitic transformation from a f.c.c. $L2_1$ -ordered cubic phase to a tetragonal phase, but the structure of this low temperature phase has so far only been reported in an incomplete and often wrong way. X-ray diffraction methods and TEM studies were used to investigate the phase transformation and the resulting domain structure. The single crystals were prepared by the Czochralski method. Several crystals with different compositions and therefore with varying temperatures of the martensitic transition T_{M_s} were used. For the alloys with T_{M_s} below room temperature a cooling device was used for both experimental techniques.

The basic phase transition occurs between the Heusler type and an ordered superstructure of the AuCu type. In this superstructure the c axis of the AuCu type is doubled. Besides the undisturbed domains of this type, the TEM investigations show domains with statistically distributed stacking faults and others with long range ordered structures. The modulation of the structure associated with the premartensitic state was also observed.

11:15 AM BB1.6
PHASE FIELD MODEL AND COMPUTER SIMULATION OF
MARTENSITIC TRANSFORMATION IN POLYCRYSTALS OF
Fe-Ni ALLOYS. A. Artemev, Carleton University, Dept. of
Mechanical and Aerospace Engineering, Ottawa, Ontario, CANADA;
Y. Jin and A.G. Khachatryan, Dept. of Ceramic and Materials
Engineering, Rutgers University, NJ.

A three-dimensional phase field model of the martensitic transformation in polycrystalline materials was developed. The evolution of phase field functions is described by a kinetic equation of the Ginzburg-Landau type, which explicitly takes into account the contribution of the transformation-induced coherency strain by using the phase field micromechanics of a structurally inhomogeneous coherent system. This model was used to simulate the fcc to bcc transformation in polycrystals of Fe - 31.0 at. % Ni. Simulations were performed for a wide range of undercoolings and under both constrained and unconstrained conditions for an average deformation in the systems. The effect of applied stress on the transformation and structure evolution in a material after the transformation was studied. Microstructures with polytwinned martensite structures were obtained. Simulation has shown that systems with low misorientation angles between grains have the same type of transformation as

single-crystal systems do. A large value of the volumetric transformation effect in the Fe - 31.0 at. % Ni system resulted in significant fractions of the residual parent phase in constrained systems after the transformation. The volume fraction of the residual parent phase in these systems depended strongly on the undercooling. The stress vs. strain curves with the hysteresis effect were obtained by the simulation of the structure evolution under applied stress.

11:30 AM **BB1.7**

EFFECT OF ELASTIC INTERACTION ON THE FORMATION OF COMPLEX MULTI DOMAIN MICROSTRUCTURAL PATTERN DURING A COHERENT HEXAGONAL TO ORTHORHOMBIC TRANSFORMATION. Y.H. Wen, Y. Wang, Department of Materials Science and Engineering, The Ohio State University, Columbus, OH; and L.Q. Chen, Department of Materials Science and Engineering, The Pennsylvania State University, University Park, PA.

The formation and temporal evolution of domain structures during a hexagonal to orthorhombic transformation is studied using computer simulations based on a continuum diffuse-interface phase-field approach. All essential driving forces for the domain formation and evolution are taken into account, including bulk chemical free energy, domain wall energy, and elastic strain energy. Two different types of transformation, i.e., congruent ordering leading to a single orthorhombic phase and precipitation resulting in a two-phase mixture, are considered. The influence of an applied strain on the transformation process is also studied. The various domain configurations observed from the computer simulations show excellent agreement with existing and concurrent experimental observations in a number of alloy systems undergoing hexagonal to orthorhombic transformations. It is shown that, even with the assumption of isotropic domain wall energy and isotropic elastic modulus, the anisotropic elastic interactions alone caused by the non-dilatational strains can reproduce all the interesting domain structures observed experimentally. It is also demonstrated that many of the specific domain configurations are actually formed during domain coarsening after the phase transformation has completed.

11:45 AM **BB1.8**

MICROSCOPIC SIMULATIONS OF PHASE SEPARATION IN ALLOYS WITH VARYING MISFIT BETWEEN PRECIPITATES AND MATRIX. Richard Weinkamer¹, Himadri Gupta², Peter Fratzl¹ and Joel L. Lebowitz². ¹Erich Schmid Institute of Materials Science, Austrian Academy of Sciences, and Metal Physics Institute, University of Leoben, AUSTRIA. ²Departments of Mathematics and Physics, Rutgers University, NJ.

Long-range elastic interactions resulting from a lattice misfit between matrix and precipitates are known to influence crucially the shape and arrangement of the precipitates and the coarsening kinetics of the microstructure during phase separation [1]. Employing a microscopic model, we have performed Monte Carlo simulations to study elastic effects on the phase separation process in a binary alloy with face-centered cubic structure, where elastic interactions are represented by springs connecting nearest neighbor atoms [2]. In this paper we compare the results of simulations, where a varying misfit between precipitates and matrix were chosen. The salient feature concerning the microstructure of the model alloy is the regular arrangement of the precipitates with increasing lattice misfit leading to a mesolattice formed by the precipitates. Deviations from a spherical precipitate shape cause the emergence of an interconnected precipitate structure even for low volume fractions. In the case of a large lattice misfit we observe a slowing down of the coarsening process, which we interpret as the prevalence of new coarsening mechanisms which have to enable coarsening without destroying the mesolattice. The movement of mesoscopic defects of the mesolattice was identified as such a new mechanism.

[1] P. Fratzl, O. Penrose and J.L. Lebowitz, J. Stat. Phys. 95, 1429 (1999).

[2] P. Fratzl and O. Penrose, Acta Metall. Mater. 43, 2921 (1995).

SESSION BB2: DOMAIN-RELATED PHENOMENA IN FERROIC MATERIALS II

Chairs: Takaaki Tsurumi and Yet-Ming Chiang
Monday Afternoon, November 27, 2000
Room 201 (Hynes)

1:30 PM ***BB2.1**

DOMAIN CONFIGURATIONS OF FERROELECTRIC SINGLE CRYSTALS AND THEIR PIEZOELECTRIC PROPERTIES. Satoshi Wada, Takaaki Tsurumi, Tokyo Inst. of Technology, Dept of Metallurgy & Ceramics Science, Tokyo, JAPAN.

Piezoelectric property of ferroelectric single crystals significantly depends on their domain configurations. The domain configurations

depend on a crystal symmetry and a crystallographic directions. Recently, in [001] oriented 3m PZN-PT single crystals, ultrahigh piezoelectric constant over 2,500pm/V and hysteresis-free strain vs. electric field behavior were reported, and their origin was assigned to an engineered domain configuration. It is also expected that the engineered domain configuration is useful for FRAM application from the viewpoint of the improvement in fatigue property. In this study, a relationship between domain configuration and piezoelectric property was investigated as a function of crystallographic direction and crystal symmetry using BaTiO₃ and other ferroelectric single crystals. Especially, it is important to reveal this relationship along the crystallographic directions related to the engineered domain configuration. For example, in 4mm ferroelectric crystal, two kinds of engineered domain configurations are expected along <111> and <110> directions. In [111] oriented 4mm BaTiO₃ crystal, piezoelectric constant over 200pm/V and hysteresis free strain vs. electric field behavior were observed while [001] oriented 4mm BaTiO₃ crystal showed the piezoelectric constant below 100pm/V and large hysteretic strain vs. electric field behavior even if its polar direction was <001> directions. Coercive electric field in [111] oriented 4mm BaTiO₃ crystal was smaller than that in [001] oriented 4mm BaTiO₃ crystal. This means that the engineered domain configuration in BaTiO₃ crystal affected domain kinetics. These studies were extended to other symmetries (mm2 and 3m) of BaTiO₃, PZN-PT and KNbO₃ crystals.

2:00 PM ***BB2.2**

SYNCHROTRON X-RAY SCATTERING STUDY OF NANODOMAINS IN PMN-PT SINGLE CRYSTALS. Andrei Tkachuk, Haydn Chen, Paul Zschack, Dept of MS&E and Frederick Seitz Materials Research Laboratory, University of Illinois, Urbana, IL.

A systematic study of the temperature dependence of superlattice reflections has been carried out using an anomalous x-ray scattering technique in single crystals of PMN-PT Pb(Mg,Nb)O₃-PbTiO₃ for the first time. Two types of nanodomains were found: 1) chemically ordered nanodomains, and 2) polar regions formed by short-range correlated ionic displacements. Each type of nanodomains gives rise to superlattice reflections at different locations in the reciprocal space. The focus of the present study is to investigate the behavior of these nanodomains as a function of temperature, leading to an improved understanding of the microstructure origin and the underlying physical principles responsible for the relaxor ferroelectric properties. The anomalous scattering technique was utilized, with the synchrotron x-ray energies tuned close to the PbL(III) absorption edge so as to highlight the contribution of Pb species to the scattering intensities. This experimental method was chosen because local dipoles, originated from Pb displacements, are most important to the observed ferroelectric behavior. From the temperature dependence of the superlattice peak intensity corresponding to polar regions borne out from in-phase rotation of oxygen octahedra (i.e. alpha spots at the face-centered locations), we found the freezing temperature of correlated atomic displacements is 200K. From the line profile analysis, the estimated nanodomain size is about 4-5 nm. Those polar domains disappeared above the freezing temperature. In contrast, chemically ordered nanodomains, which gives rise to F spots at the body-centered locations exhibited little temperature dependence in its size. These ordered domains were found even at 800K. However, a noticeable change in intensity, and its size, was observed near 200K. We believe the degree of short-range order remains unchanged due to its relatively low thermal energy. But the out-of-phase oxygen octahedra rotation and its associated Pb displacements can cause intensity to vary at the same F spots. It is thus concluded that observed intensity changes are related to octahedral oxygen rotations. Our results confirm the dipolar glass model for the PMN relaxors, where intrinsic disorder due to off-stoichiometric composition and competing interactions are both present.

2:30 PM ***BB2.3**

STRUCTURE AND DYNAMICS OF FERROELECTRIC DOMAIN WALLS. V. Gopalan, S. Kim, Pennsylvania State University, State College, PA; A. Itagi, Carnegie Mellon University, Pittsburgh, PA; T.E. Mitchell, P. Swart, A. Saxena, Los Alamos National Lab, Los Alamos, NM.

This talk addresses classical issues of domain wall structure and motion under external driving forces with reference to lithium niobate and lithium tantalate ferroelectrics. Recent studies reveal many new aspects of these ferroelectrics, such as a large sensitivity of physical properties to small amounts of nonstoichiometry, regions of strain, electric fields and optical birefringence at the 180 deg domain walls extending over many microns, local pinning and bending motion of a domain wall well below the coercive field required for domain motion, and time dependent mobility of a wall at a fixed driving field. The talk will revisit very fundamental questions in ferroelectrics, such as, what is internal structure of a domain wall? and what are the intrinsic and extrinsic factors governing domain wall motion?

3:30 PM *BB2.4

NONDESTRUCTIVE OBSERVATIONS OF POLAR DOMAIN STRUCTURE AND ITS KINETICS USING SGH INTERFERENCE MICROSCOPE. Yoshiaki Uesu, Noritaka Kato, Haruyuki Mohri, Hirotohi Shibata, Department of Physics, Waseda University, Tokyo, JAPAN.

Optical second harmonic generation (SHG) becomes an important technique for observing various kinds of domain structure in ferroelectrics/1,2/ and anti-ferromagnetics/3/, polar molecule aggregation state/4-6/ and poling state in organic crystals. We have proposed an interference SHG microscope which enables us to observe domain structures with opposite polarity/7,8/. So far the non-destructive observation has been believed to be impossible, as the states with opposite polarity have same optical properties. Our method is to exploit the nature of the SH waves generated in anti-parallel domains, i.e. they have equal amplitude but opposite phase. If they are mixed with other homogeneous SH wave, they interfere constructively in one domain, while destructively in another. Thus the domains with opposite polarity become visible. We applied successively the SHG interference microscopy to the observations of domain structures in BaTiO₃/1/, periodically inverted domains in LiTaO₃ quasi-phase matching devices for wavelength converters /9/ and J-aggregate domain structure in merocyanine dye molecules/4,5/. In this paper, a special emphasis will be put on the non-destructive observations of domain reversal process in LiNbO₃ and 2D phase transition between J-aggregates accompanied by the thermochromism. References:

- [1] Y. Uesu, S. Kurimura, Y. Yamamoto: Appl. Phys. Lett. 66 (1995) 2165.
- [2] M. Floersheimer et al: Appl. Phys. B, B67 (1998) 593-599.
- [3] M. Fiebig, D. Froehlich, G. Stuyterman, R.V. Pisarev: Appl. Phys. Lett. 66 (1995) 2906.
- [4] N. Kato, K. Saito, Y. Uesu: Thin Solid Films, 338 (1998) 257.
- [5] N. Kato, K. Saito, Y. Uesu: Chem. Phys. Lett. 312 (1999) 115.
- [6] M. Floersheimer et al: Adv. Mater. 9 (1997) 1056, *ibid.*, 9 (1997) 1063.
- [7] Y. Uesu: J. Korean Phys. Soc., 32 (1998) S461.
- [8] Y. Uesu, N. Kato: Phys. Solid State, 41 (1999) 688.
- [9] S. Kurimura, Y. Uesu: J. Appl. Phys. 81 (1997) 369.

4:00 PM BB2.5

DOMAIN STRUCTURES AND PHASES IN RELAXOR FERROELECTRIC CRYSTALS. Chi-Shun Tu, L.-F. Chen, C.-L. Tsai, Fu Jen University, Dept of Physics, Taipei, TAIWAN; V. Hugo Schmidt, Montana State University, Dept of Physics, Bozeman, MT.

The domain structures and phase transitions have been observed as a function of temperature for relaxor ferroelectric single crystals (PbMg_{1/3}Nb_{2/3}O₃)_{1-x}(PbTiO₃)_x (PMN-xPT) for x=0.24,0.31,0.34, by using polarized light microscope. In the region of diffuse phase transition where corresponds a frequency-dependent behavior in dielectric permittivity, PMN-24%PT crystal shows complex domain configurations with the coexistence phase of trigonal (pseudo-cubic) and cubic states. Beside the coexistence phase of tetragonal and cubic orderings in the higher temperature region, domain structures of both PMN-31%PT and PMN-34%PT crystals exhibit another coexistence phase of tetragonal and trigonal (pseudo-cubic) states in a wide temperature range below 380 K. It was also found that different individual domains in the same phase have different transition temperatures. This phenomenon suggests an inhomogeneous distribution of Ti⁴⁺ concentration in the crystals. Such a fluctuation is believed to result from an unequal occupation of the B-site by the competitive ions Mg²⁺, Nb⁵⁺ and Ti⁴⁺ while lattice structure establishing. With light scattering and dielectric results, a phase diagram will be proposed.

4:15 PM BB2.6

EFFECT OF OXYGEN VACANCIES ON DOMAIN STRUCTURE-PROPERTY RELATIONSHIP OF MODIFIED PZT FERROELECTRIC CERAMICS. Qi Tan, Honeywell Electronic Materials, Spokane, WA.

Oxygen vacancies were known to be very effective in changing electrical/ferroelectric properties and domain structures in hard PZTs. They are also the major cause for fatigue and imprint of thin film devices. Understanding the characteristics, distribution, and role of oxygen vacancies in PZT materials would provide valuable information for the enhancement of ferroelectric properties. Although there are a large number of publications addressing the effect of oxygen vacancies from various perspectives, a general picture of oxygen vacancies in terms of their characteristics and generation mechanism has not yet been explored. This study investigated the origins of oxygen vacancies in terms of three different classes: lower-valent substitution; higher-valent substitution; and reduced oxygen atmosphere. Creation of oxygen vacancies due to lower-valent substitution has been extensively studied primarily because of "hard"

PZT application. Information on domain structure and their relations with ferroelectric properties are limited. This study observed a special domain structure and the distribution of oxygen vacancies in lower-valent modified PZTs. Higher valent substitution is usually not thought to be a cause of oxygen vacancies. However, our study revealed such a possibility when taking ionic size of substituents into account. We found domain structures and the corresponding ferroelectric properties in PZT modified by small high-valent substituents to be comparative to those in lower-valent substituted PZT. Domain structure and ferroelectric properties of PZT materials processed in reduced oxygen atmosphere have been rarely investigated. Our work revealed fine structure within nanodomains in oxygen deficient PZT due to vacuum hot-pressing. The elimination of oxygen vacancies resulted in the recovery of normal domain structure and ferroelectric properties.

4:30 PM BB2.7

THE ORIGIN OF THE FERROELECTRIC EFFECT IN AURIVILLIUS COMPOUNDS. Donaji Y. Suárez, Ian M. Reaney, William E. Lee, Department of Engineering Materials, The University of Sheffield, UNITED KINGDOM.

Bi containing, layered perovskites based on Aurivillius compounds are finding applications in non-volatile piezoelectric random access memories and as high temperature piezoelectric sensors. The parent compound, Bi₄Ti₃O₁₂, is one of a family of oxides which has the general formula, Bi₂A_{n-1}Ti_nO_{3n-1}, where the A site can be occupied by different cations (e.g. Sr, Ba, Ca and Pb). The crystal structure comprises perovskite type slabs which are separated and sheared along 1/2(111) by rock salt structured Bi₂O₂²⁻ layers. The number of perovskite, ABO₃, units in the slabs depends on the concentration of the A-cation. It is known that these materials undergo ferroelectric and cell-doubling octahedral tilt phase transitions on cooling Superlattice reflections and planar defects arising from tilting of octahedra around the c-axis have been identified in a large number of Aurivillius compounds. The intensity of superlattice spots in electron diffraction patterns at room temperature has been qualitatively shown to diminish as the tolerance factor of the perovskite blocks increases. The paraelectric to ferroelectric phase transition temperature (T_c) was measured for all the compounds and plotted against the number of perovskite units inbetween Bi₂O₂²⁻ layers and the tolerance factor. No correlation of T_c with the number of perovskite units was observed, but T_c decreased over the range of samples as the tolerance factor increased. It is proposed that the onset of octahedral tilting and T_c are related in the Aurivillius phases.

4:45 PM BB2.8

DOMAIN STRUCTURE AND POLARIZATION BEHAVIOR OF FERROELECTRIC SURFACES BY SCANNING PROBE MICROSCOPY. Dawn A. Bonnell, Sergei V. Kalinin, Dept. Mat. Sci. Eng., University of Pennsylvania, Philadelphia, PA.

Electrostatic force microscopy (EFM) and scanning surface potential microscopy (SSPM) are applied to BaTiO₃ (100) surfaces. The combination of topographic and surface potential images is used to reconstruct surface domain structures. Distance and bias dependencies of the electrostatic force gradient data are used to determine the fraction of unscreened polarization charge. Experimental SSPM and EFM domain profiles are compared to calculated potential and field distributions for the unscreened and screened cases. Surface potential evolution during phase transitions and the potential distribution near moving domain walls allows the relationship between domain potential polarity and polarization orientation to be determined. Cycling through the phase transition results in new domain structure at each cycle. Our results indicate that polarization bound charge is completely screened on this surface when in air. This conclusion is corroborated by piezoresponse imaging technique. Surface potential is attributed to the formation of double layer due to the complete screening of polarization charge. The absolute value of the potential difference between domains of opposite polarity suggests that surface adsorbates play a governing role in potential formation mechanism, though intrinsic screening by free charge carriers is not completely excluded.

SESSION BB3: DOMAIN-RELATED PHENOMENA
IN FERROIC MATERIALS III

Chairs: Angus I. Kingon and Haydn Chen
Tuesday Morning, November 28, 2000
Room 201 (Hynes)

8:30 AM *BB3.1

FERROELECTRIC DOMAIN STRUCTURE IN KNbO₃/KTaO₃ HETEROSTRUCTURES BY MOLECULAR-DYNAMICS SIMULATION. M. Sepliarsky, Materials Science Division, Argonne National Laboratory, Argonne IL and Instituto de Fisica Rosario,

CONICET-UNR, Rosario, ARGENTINA; S.R. Phillpot, D. Wolf, Materials Science Division, Argonne National Laboratory, Argonne IL; M.G. Stachiotti and R.L. Migoni, Instituto de Fisica Rosario, CONICET-UNR, Rosario, ARGENTINA.

We have developed an atomic-level approach to the simulation of the ferroelectric perovskite KNbO_3 (KNO) based on the traditional Buckingham potential with shell model which correctly reproduces the ferroelectric phase behavior and dielectric and piezoelectric properties. Using this and a compatible interatomic potential for the incipient ferroelectric KTaO_3 (KTO), we have determined the structure and ferroelectric properties of coherent KNO/KTO heterostructures of varying layer thicknesses; such heterostructures have important opto-electronic applications. We find that there is a strong coupling of the polarizations parallel to the modulation direction of the KNO and KTO layers, but that the in-plane polarizations are only rather weakly coupled between layers. Moreover, we find that the differing strain states produced respectively by growth on a KNO substrate and on a KTO substrate result in qualitatively different calculated ferroelectric behavior in the heterostructures.

9:00 AM *BB3.2

APPLICATION OF TRANSMISSION ELECTRON MICROSCOPY FOR THE CHARACTERIZATION OF DOMAIN MICROSTRUCTURES IN MATERIALS. X.Q. Pan, Department of MS&E, University of Michigan, Ann Arbor, MI.

Domains and domain walls appear in all materials which undergo a structural phase transition. Domain structures, which are determined by the change of crystallographic symmetry through the phase transition, can influence many materials properties such as electrical, magnetic and dielectric properties. Therefore, the characterization of domain microstructures and domain wall motion during the structural phase transition is needed. Transmission electron microscopy (TEM) is a unique, powerful technique which provides the direct observation of domain structures and domain wall movement in materials during a phase transition. In this paper we will demonstrate (1) the in-situ TEM observation of domain formation and domain wall movement in ferroelectric materials ($\text{Ba}_2\text{NaNb}_5\text{O}_{15}$ and K_2ZnCl_4), (2) the characterization of complex ferroelastic domain structures in epitaxial oxide thin films (SrRuO_3 and BaRuO_3), and (3) the determination of the size and distribution of nano domains in a relaxor material ($\text{PbMg}_{1/3}\text{Nb}_{2/3}\text{O}_3$).

9:30 AM BB3.3

THEORY OF RELAXORS. Rinat Mamin, Kazan Physical-Technical Institute of Russian Academy of Sciences, Kazan, RUSSIA.

The diffuse phase transition theory for the relaxors is developing. The localization of charge carriers on the local centers play an important role in this theory. Localized charges create local electric fields which stimulate occurrence of induced polarization and as result domain microstructure appear. The direction of this polarization in microdomain is determined by space distribution of levels occupied by charge carriers. After phase transition spontaneous polarization is directed along this fields. Temperature behavior of dielectric permittivity is determined by dynamics of charge carriers localization on the local centers. The diffuse permittivity maximum means that the system is effectively very close to the phase transition, but it is in a paraelectric phase everywhere above "freezing" temperature. Dispersion of dielectric permittivity is determined by vibrational properties of the local states. As charge carriers are localized on local centers, deformation occurs around them and a local polarization arises. In these sense this formations are localized polarons. It should be noted that a characteristic vibrational frequency of this formation is determined by characteristic times both of the lattice and electron subsystems. The delay time of phase transition at field after cooling at zero field is also described. It is shown that the behavior of relaxors in low-temperature phase may be connected with a processes activation of charge local states. Passage of the phase transition to the phase with long-range ferroelectric order after application of electric field will be determined by local center ionization and consequent re-distribution of the direction of polarization in microregions. Good agreement between theory and experimental data have been obtained. We obtain a quantitative agreement for the temperature behavior of permittivity and a quantitative agreement for the dispersion behavior of permittivity. The value of parameters we used are in good agreement with experimental data on conductivity, and the temperature range of charge carrier localization is about phase transition temperature. All of these questions is clarified basing on rather simple, preferably thermodynamic, considerations.

9:45 AM BB3.4

INFLUENCE OF SURFACE INDUCED NUCLEATION ON SWITCHING BEHAVIOR IN FERROELECTRICS. Rajeev Ahluwalia, Wenwu Cao, Materials Research Laboratory, Pennsylvania State University, University Park, PA.

We simulate the domain switching in a finite sized ferroelectric system using the time dependent Ginzburg-Landau approach. The model also includes elastic and electrostrictive effects in the form of a long-range interaction obtained by eliminating the strain fields, subject to the elastic compatibility constraint. The surface effects are simulated by enforcing zero polarization constraint out side the system. We find that the switching process is strongly influenced by the presence of the surface. The surface acts as a nucleation source of orthogonally polarized domains. Interestingly, the actual switching occurs through ninety degree domain reorientations. Dependence of the hysteresis loops on the system size is also studied.

10:30 AM BB3.5

DOMAIN NUCLEATION AND RELAXATION KINETICS IN FERROELECTRIC THIN FILMS. C.S. Ganpule, A.L. Roytburd, V. Nagarajan, E.D. Williams and R. Ramesh, Materials Science and Engineering Center, Univ. of Maryland, College Park, MD; J.F. Scott, Department of Earth Sciences, Univ. of Cambridge, Cambridge, UNITED KINGDOM.

The time dependent relaxation of the remnant polarization in epitaxial lead zirconate titanate ($\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$, PZT) ferroelectric thin films, containing a uniform 2-dimensional grid of 90° domains (c-axis in the plane of the film), is examined using piezoresponse microscopy. The 90° domain walls preferentially nucleate the 180° reverse domains during relaxation, with a significant directional anisotropy. Relaxation occurs through the nucleation and growth of reverse domains, which subsequently coalesce and consume the entire region as a function of time. The rate of growth of reversed domains is closely related to the radius of curvature of the growing phase. A thermodynamic analysis has been carried out to model and explain this time dynamics. We will present the results of these experimental and theoretical studies.

This work is supported by National Science Foundation - Materials Research Science and Engineering Center (NSF-MRSEC).

10:45 AM BB3.6

THE INFLUENCE OF DOMAIN STRUCTURE ON THE DYNAMIC ELECTRO-OPTIC RESPONSE OF EPITAXIAL FERROELECTRIC THIN FILM. Brent H. Hoeraman, Barbara M. Nichols, Andrew R. Teren, David Towner, Bruce W. Wessels. Northwestern Univ, Dept of MS&E, Evanston, IL.

Ferroelectric epitaxial films provide several potential advantages over the single crystals used in current high-speed electro-optic (EO) modulators. The benefits include lower driving voltages, higher modulation frequencies, and the possibility of direct integration with silicon. Consequently, investigation of the dynamic EO response of thin film ferroelectric materials has been undertaken. The dynamic EO response of epitaxial KNbO_3 and BaTiO_3 thin films was examined using a transverse measurement technique. Initial measurements of the EO coefficient in these films resulted in values ranging from 1 to 20 pm/V. The EO coefficients are more than an order of magnitude lower than those observed in single crystals as a result of the presence of multiple ferroelectric domain variants. Through consideration of the thin film domain structure, the EO coefficients can be effectively increased by a factor of 5 to 10. For BaTiO_3 thin films EO coefficients as high as 210 pm/V have been observed. The dynamic response of the EO coefficient and the polarizability of the films was also measured between 6 ns and 1 s. For both thin film ferroelectrics a power law transient response of the EO effect and the polarization was observed after the removal of an applied electric field. This power law relaxation has been attributed to domain reorientation under the influence of an applied electric field.

11:00 AM BB3.7

DOMAIN STATE PROPERTIES IN $\text{SrTi}(\text{}^{16}\text{O}_{1-x}\text{}^{18}\text{O}_x)_3$ SINGLE CRYSTALS. Ruiping Wang, Mitsuru Itoh, Mat and Structures Lab, Tokyo Inst. Tech, Yokohama JAPAN; Katsunori Iio, Kiyoko Morishita, Dept Phys, Tokyo Inst Tech, Tokyo, JAPAN; Yuji Tsujimi, Toshiro Yagi, Res Inst Electronic Sci, Hokkaido Univ, Sapporo, JAPAN.

Recent our discovery on the evolution of ferroelectricity in ^{18}O -exchanged SrTiO_3 (STO18)^{1,2} has attracted much attention mainly in the physics field because last 30 years quite many researches have been carried out to elucidate the mechanism of the change from quantum paraelectric to quantum ferroelectric in SrTiO_3 (STO16) and KTaO_3 (KTO) by means of atomic substitution. Atomic substitutions, e.g., Ca for Sr in STO and Li for K in KTO, induce fairly strong and long-range local field in the vicinity of substituted elements and in most cases the bulk properties of matrix change extensively. Contrary to the atomic substitution, oxygen-isotope exchange is a moderate way to observe net lattice effect of the phase transition phenomena. In this paper, we will report the domain state properties of $\text{SrTi}(\text{}^{16}\text{O}_{1-x}\text{}^{18}\text{O}_x)_3$. Both (100) and (110) oriented samples were subjected to the oxygen-isotope exchange. The exchanged and well-homogenized

samples were conducted to the various kinds of physical property measurements. STO18 and other samples with intermediate compositions showed a typical behavior of ferroelectric with $T_c < 24$ K. However, the facts that there is no heat anomaly at T_c and that the temperatures at which the DE loop and/or pyroelectricity become unobservable are higher than T_c are considered to come from the 'domain state', which is different from that observed in the typical ferroelectric with well-defined macroscopic size domain walls. Recent our experiments on the first Raman, birefringence, relaxation of polarization, and amplitude dependence of the dielectric constant clearly support the occurrence of the domain state with a quenched random-field disorder.

1. M. Itoh et al, Phys.Rev.Lett., 82, 3540(1999).
2. M. Itoh and R. Wang, Appl. Phys.Lett., 76, 221(2000).

11:15 AM BB3.8

MODELING OF BULK PPLN: DOMAIN STRUCTURE AND IMPURITY DISTRIBUTION. Inessa Naumova, Nina Evlanova, Tatyana Chaplina, Stanislav Blokhin, Dept of Physics, Moscow State Univ, Moscow, RUSSIA; Sergej Lavrishchev, General Physics Institute, Moscow, RUSSIA.

Doped LiNbO₃ single crystals with the bulk periodically poled structure (PPLN) with period 28 nm were grown by Czochralski technique along (X-axis). The goal of this work is to investigate of the correlation between the impurity distribution and the ferroelectric domain structure in the Y:LiNbO₃ single crystal. It has been established that in the periodically poled Czochralski-grown lithium niobate positive and negative domains can have unequal dimensions within the limits of one period. The reasons of this domain inequality should be revealed because this asymmetry decreases the efficiency of quasi-phase-matching nonlinear-optical conversions in the crystal. The yttrium distribution of across the domain boundaries was measured using X-ray wave dispersive spectrum microanalysis (WDS) by the scanning electron microscope (SEM) - JSM-840. Camebax SX-50 was used for impurity concentration measurements. The ferroelectric domain structure in the Y:LiNbO₃ crystal on the surface was studied by the selective chemical etching. The direct WDS study of the crystal surface with the domain boundaries revealed by chemical etching is not correct because of the changes in chemical composition of the surface. We elaborated the special method to study the correspondence between the impurity distribution and domain wall locations. The peculiarity of our method consists in the following: the domain wall patterns revealed by the selective chemical etching and the impurity concentration curves (WDS analysis) which were performed on the polished surface, were stored in the computer memory and then superposed with the help of the marks and the analysis traces on the studied crystal cut (with accuracy 1mm). The investigation of the upper and lower parts of the sample showed that the domain walls connected with the minima of the yttrium concentration move to a greater extent than the domain walls located near the maxima of Y concentration. We observed that in the annealing process the minima became broader what makes the movement of walls easier. We calculated the domain wall speed (5 mm/h) assuming that the main wall movement occurs under the high temperature below Curie point (1150°C). This study is supported by INTAS (Grant Number 97-31275).

11:30 AM BB3.9

DOMAIN STRUCTURE IN LaFeO₃ THIN FILMS AND ITS ROLE ON EXCHANGE COUPLING. Jin Won Seo^{1,2}, Jean Fompeyrine², Heinz Siegart², Jean-Pierre Locquet². ¹Institute de Physique, Université de Neuchâtel, Neuchâtel, SWITZERLAND; ²IBM Research Division, Zurich Research Laboratory, Rüschlikon, SWITZERLAND; Andreas Scholl, Frithjof Nolting, Simone Anders, Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA; Jan Lüning, Joachim Stöhr, Stanford Synchrotron Radiation Laboratory, Stanford, CA; Mike F. Toney, Eric E. Fullerton IBM Research Division, Almaden Research Center, San Jose, CA.

Antiferromagnetic (AF) materials are essential elements in the read-heads of current hard-disk drives, where they serve to pin the nearest ferromagnetic (FM) layer into one preferred orientation. This coupling between the spins in the AF and FM layers also called the exchange bias determines to a large extent the efficiency and the long-term stability of the giant magnetoresistance ratio, but is still a poorly understood phenomenon. However, a number of experiments and theoretical models have suggested that the exchange bias is correlated with the presence and morphology of AF domains. The AF orthoferrite LaFeO₃ is an interesting AF model system to explore this correlation, as the easy axis A is uniquely defined along the a-axis of the orthorhombic crystal (a = 0.5557 nm, b = 0.55652 nm, and c = 0.78542 nm). The orthorhombicity of this crystal is large enough that its structural recognition on a local scale allows a direct determination of the AF axis. By means of photo-emission electron microscopy AF domains have been found in LaFeO₃ thin films grown on SrTiO₃ substrates [1]. As observed by transmission electron

microscopy the domains are correlated to 90° twin structure in these films. Firstly we study the parameters such as growth condition, epitaxial relationship and strain which control the domain size and shape as well as the Neel temperature. Secondly, we investigate how the AF domains pin the FM domains leading to exchange coupling [2]. The results imply that the alignment of FM spins is determined, domain by domain, by the spin directions in the underlying AF layer. [1] A. Scholl et al., Science 287 (2000) 1014-1016. [2] F. Nolting et al., Nature 405 (2000) 767-769.

11:45 AM BB3.10

MICRO- AND NANOSCALE DOMAIN ENGINEERING: PERIODIC PATTERNING AND SELF-ORGANIZED STRUCTURES IN LITHIUM NIOBATE AND LITHIUM TANTALATE.

Vladimir Ya. Shur, Evgenii L. Romyantsev, Ekaterina V. Nikolaeva, Eugene I. Shishkin, Ural State Univ., Inst. of Phys. & Appl. Math., Ekaterinburg, RUSSIA; Robert Batchko, Martin Fejer, Robert Byer, Stanford Univ., E.L. Ginzton Laboratory, Stanford, CA.

We present the survey of our recent study of the field-induced domain kinetics in single-crystalline congruent lithium niobate (LN) and lithium tantalate. The proposed backswitched poling by field application to the lithographically defined metal strip electrodes allows to engineer the short-pitch periodical micro-scale structures for nonlinear optical application. First-order single-pass continuous-wave second harmonic generation of 60 mW at 460 nm is achieved at 6.1%/W efficiency in 0.5-mm-thick 4-micron-period LN. We demonstrate the formation of self-organized nano-scale domain patterns as the first achievement in domain nano-technology. It was shown how to obtain the giant dielectric response at low frequencies in LN by self-maintained formation of charged domain walls. Domain evolution has been investigated by in situ optical observation of the instantaneous domain patterns. SEM and SFM have been used for high-resolution visualization of the static domain patterns revealed by etching on polar surfaces and cross-sections. We have proposed several variants of domain manipulation during poling: the frequency multiplication of the domain patterns as compare with electrode ones, domain "erasing" and "splitting", formation of oriented arrays of nanoscale domains. We have demonstrated the production of domain patterns with period down to 2.6 microns in 0.5-mm-thick wafers and strictly oriented quasi-periodic domain arrays consisting of the individual nanodomains with diameter down to 30 nm and density up to 100 per square micron. The mechanism of self-maintaining correlated nucleation effect was revealed. It was shown that nanodomains arise due to the field anomaly, which is determined by the thickness of intrinsic dielectric gap or artificial insulating layer. The research was made possible in part by Program "Basic Research in Russian Universities" (Grant No.5563), by Grant No.97-0-7.1-236 of the Ministry of Education of the Russian Federation and by Grant No.98-02-17562 of the Russian Foundation of Basic Research.

SESSION BB4: DOMAIN-RELATED PHENOMENA IN FERROIC MATERIALS IV

Chairs: Alexander K. Tagantsev and Simon R. Phillpot
Tuesday Afternoon, November 28, 2000
Room 201 (Hynes)

1:30 PM *BB4.1

UNDERSTANDING AND MANIPULATING THE PROPERTIES OF MIXED A-SITE CATION RELAXOR PEROVSKITES.

Y.-M. Chiang, S.A. Sheets, A.N. Soukhojak, N. Ohashi, H. Wang, J. Assal, Massachusetts Institute of Technology, Dept of MS&E, Cambridge, MA.

Lead perovskites with mixed B-site cations such as PZT, PMN, and PLZT have been developed over the course of nearly four decades, and have been the mainstay of the ferroelectrics industry. However, environmental concerns have recently renewed the interest in lead-free ferroelectrics and piezoelectrics. In this work, systematic exploration of composition-structure-property relationships in doped Na_{1/2}Bi_{1/2}TiO₃ perovskites has shown that many parallels to the B-site relaxors exist. These include: 1) the existence of structurally- and compositionally-modulated nanodomains over extremely broad ranges of composition; 2) broad and frequency-dependent dielectric relaxation following the Vogel-Fulcher relationship; 3) high electrostriction strain in compositions doped to isolate the paraelectric phase. By manipulating phase and domain stability with the guidance of high resolution electron microscopy, dielectric measurements, and electromechanical tests, compositions have been identified which have actuation comparable to that of many lead perovskites. Two particularly interesting fields of behavior identified in the Ba Zr codoped system are electrostrictive single crystals and polycrystals with actuation strains (0.45% and 0.2% respectively) exceeding that of optimized PMNs, and relaxor-ferroelectric compositions in which polycrystals show actuation nearly identical to that of PZT-8.

2:00 PM *BB4.2

THERMODYNAMICS OF POLYDOMAIN STRUCTURES ARISING AT FIELD-INDUCED TRANSFORMATION. Alexander L. Roytburd, Dept of Materials and Nuclear Engr, Univ of Maryland, MD.

Thermodynamic analysis of polydomain structures arising during phase transformation induced by mechanical, electrical, and magnetic fields shows that the equilibrium product phase is incompatible with the parent phase as a rule. Therefore, internal stresses, depolarising, and demagnetizing fields are created in the two-phase state. Corresponding increase of the free energy leads to the thermodynamic hysteresis of the phase transformation. This thermodynamic phenomenon is considered for superelastic stress induced transformation of shape memory alloys as well as for ferroelectric and ferromagnetic transformations with significant striction effects.

2:30 PM BB4.3

DOMAIN WALL MIGRATION IN PURE AND IMPURE SYSTEMS: SIMULATIONS AND ANALYTICAL MODELS. Mikhail I. Mendeleev, David J. Srolovitz, Princeton University, Princeton Materials Institute and Dept. of Mechanical & Aerospace Engineering, NJ; Weinan E, Princeton University, Program in Applied and Computational Mathematics & Mathematics Dept., NJ.

The properties of many active materials are determined by the relative ease with which domain walls can be moved. The mobility of domain walls can be quite different, depending on whether the system is pure or impure, whether the impurities or point defects are mobile or stationary, the nature of impurity/domain wall interactions, etc. We will present the results of a series of simulations of domain wall migration in systems without impurities, with static impurities and with impurities that are free to diffuse. We perform the simulations within a kinetic Monte Carlo formalism based on a simple spin model. While such domain wall migration has been analyzed theoretically, the present simulations demonstrate that the assumptions commonly made lead to significant errors (both qualitative and quantitative). Even in a pure system, the driving force-velocity relationship is non-linear (in disagreement with commonly used models). An analytical model for this relationship is derived and shown to provide good agreement with the simulations. The simulations of impure systems focussed on interstitial impurities and considered the effects of bulk impurity concentration, impurity diffusivity, interaction strength, and temperature. Two regimes of motion were distinguished (at low and high velocity) with a smooth transition between them under all conditions. Contrary to the classical continuum model of impurity drag, the simulation results demonstrate that attractive and repulsive impurity-domain wall interactions yield very different domain wall mobilities and the domain wall velocity never exhibits sharp jumps. The deficiencies in the classical continuum model are associated with dimensionality and the stochastic nature of the impurity distribution. A discrete model is developed that properly describes the transition between regimes and the differences between attractive and repulsive impurities.

2:45 PM BB4.4

RECENT PROGRESS ON SCANNING NONLINEAR DIELECTRIC MICROSCOPY WITH SUB-NANOMETER RESOLUTION. Yasuo Cho, Hroyuki Odagawa, Kaori Matsuura, Satoshi Kazuta, Koya Ohara, Research Institute of Electrical Communication, Tohoku University, Sendai, JAPAN.

We have recently proposed and developed a new purely electrical method for imaging the state of the remanent polarization of ferroelectric materials, which involves the measurement of point-to-point variations of the nonlinear dielectric constant of a specimen, and is termed the "scanning nonlinear dielectric microscopy" (SNDM). This is the first successful purely electrical method for observing the ferroelectric polarization distribution without the influence of the shielding effect from free charge. Now, the stronger demands for the observation of very small domains with nano- and subnano- meter sizes has arisen amongst researchers of ferroelectric materials, for example, to investigate domain wall structures, to clarify the minimum domain sizes etc. Therefore, we developed a new SNDM with nano or subnano meter resolution. We also report the results of the imaging of the ferroelectric domains in a BaTiO₃ single crystal and in PZT thin films using the SNDM with nano-meter resolution. Especially in the PZT thin film measurement, we succeeded to obtain a domain image with a sub-nanometer resolution. Next theoretically we clarify the reason why a very high (nano-sized) resolution can be easily obtained, even if a relatively thick needle is used for the probe. Using the results of the theoretical studies, quantitative measurements of the distribution of linear and nonlinear dielectric constants of ferroelectric materials are successfully

performed. Finally, we also demonstrate that SNDM technology is very useful for determining local crystal anisotropy such as a polarity of thin film deposited on polar substrates.

3:30 PM *BB4.5

DYNAMICS OF SHAPE-MEMORY STRINGS USED AS FLAGELLA AND CILIA. Kaushik Bhattacharya and Prashant Purohit, Division of Engineering and Applied Science, California Institute of Technology, Pasadena, CA.

Shape-memory alloys are very attractive for microactuators because they can produce the largest work per unit volume amongst all actuator materials. In particular, they have recently been proposed for use in micropulsion in the form of flagella and cilia. This talk will describe theoretical and experimental studies motivated by these applications. Of particular interest is the nucleation and propagation of phase and twin boundaries, their mobility, and their ability to create a beating motion in three dimensions. Parallel atomistic and continuum theory will describe the unique features of phase/twin boundaries in strings and contrast them with interfaces in bulk materials. These results will be compared with experimental observations.

4:00 PM BB4.6

MAGNETIC FORCE MICROSCOPY STUDY OF MAGNETIC DOMAIN MICROSTRUCTURES IN CoAg GRANULAR FILMS. M.F. Chiah, S.P. Wong, W.Y. Cheung, Department of Electronic Engineering, The Chinese University of Hong Kong, Hong Kong, CHINA.

Granular thin films of Co_xAg_{1-x} of various compositions were prepared using a pulsed filtered vacuum arc co-deposition system. The composition of the films was determined by Rutherford backscattering spectrometry. The magnetic domain microstructures were studied using magnetic force microscopy. Two distinguish types of domain structure patterns were observed for films of different Co composition. For films with a cobalt composition x of 0.7 and smaller, the magnetic domain structures show beautiful patterns consisting of long labyrinthine stripes of bright and dark regions, which represent domains with antiparallel perpendicular magnetization. The typical width of the stripes is slightly smaller than 0.1 μm and the typical length can be as long as a few μm. On the contrary, for films with a cobalt composition x of 0.8 and larger, the domain structure pattern changed dramatically to a much more irregular shape, resembling that of a pure Co film. The variation of the magnetic domain microstructures of the films with annealing temperature and their evolution under the application of an external magnetic field were also studied. These results will be presented and discussed in conjunction with the magnetic properties of these films.

This work is partially supported by the Research Grants Council of Hong Kong SAR (Ref. No.: CUHK 4152/98E).

4:15 PM BB4.7

STUDY OF THE THERMAL BROADENING OF FERROELECTRIC DOMAIN WALLS IN LEAD TITANATE BY QUANTITATIVE TRANSMISSION ELECTRON MICROSCOPY. Alain Sfera, Michael Foeth, Philippe A. Buffat, Pierre Stadelmann, Swiss Federal Institute of Technology, Interdepartmental Center of Electron Microscopy, Lausanne, SWITZERLAND.

The ferroelectric domain wall thickness is an important parameter and its behavior as a function of the temperature is directly related to the order of the phase transition. Several models exist to describe the thermal broadening of domain walls; however up to now there was no quantitative experimental observation of this phenomenon to check their validity. We have developed two methods for the measurement of 90° ferroelectric domain wall thickness. The first method uses high resolution transmission electron microscopy (HRTEM) and allows a direct observation of the lattice distortion across the boundary. The domain wall thickness is obtained from the measurement of the crystal lattice distortion near the interface using the geometrical phase method. This method consists in selecting a circular region of the power spectrum around a reflection peak, centering the Fourier space on this reflection and performing the inverse Fourier transform. The information about the local displacement of atomic planes corresponding to the selected reflection is extracted from the phase component of the obtained complex image. The second method consists in a quantitative analysis of the thickness fringes that appear on weak beam images of inclined domain walls. By fitting simulated fringe profiles to the experimental ones, the domain wall thickness can be extracted in a quantitative way. Both methods have been successfully applied to lead titanate single-crystals (PbTiO₃). This perovskite presents a first order phase transition at T_c = 492.2°C which corresponds to the transformation of the crystal from a high-temperature paraelectric cubic phase to a low-temperature ferroelectric tetragonal phase. The thermal broadening of 90° domain

walls has been measured and the results compared to the prediction obtained by using different Ginzburg-Landau models.

4:30 PM *BB4.8

COMPARISON OF DOMAIN STRUCTURE AND DOMAIN WALL MOBILITY IN $\text{Pb}(\text{Zr,Ti})\text{O}_3$ AND $\text{SrBi}_2\text{Ta}_2\text{O}_9$. Yong Ding, Yening Wang, National Laboratory of Solid State Microstructures, Nanjing University, Nanjing, PR CHINA; Jianshe Liu, Center for Advanced Study, Tsinghua University, Beijing, PR CHINA; Kehsin Kuo, Beijing Laboratory of Electron Microscopy, Institute of Physics and Center for Condensed Matter Physics, Chinese Academy of Sciences, Beijing, PR CHINA.

$\text{Pb}(\text{Zr, Ti})\text{O}_3$ (PZT) and $\text{SrBi}_2\text{Ta}_2\text{O}_9$ (SBT) are good materials for use on non-volatile random access memory applications. But PZT with metal (platinum) electrodes suffers from serious fatigue. SBT, a layered perovskite oxide, has been proposed to make fatigue free ferroelectric capacitors with platinum electrodes. Generally fatigue dynamics is considered essentially as the competition between the pinning and unpinning of ferroelectric domain wall, the domain structure comparison between PZT and SBT is necessary. From space group analysis and TEM, HREM observation, 90° domain wall (DW) and 180° DW exist in PZT; whereas in SBT, in addition to 90° DW and 180° DW, new classes of DW, anti-phase boundary (APB) and 90° -APB also exist with high density. The configuration of 90° DW in SBT is irregular, while in PZT, it always straight and along (110) planes. The mobility of domain is also investigated by in situ TEM observation. In PZT, the new 90° domains nucleate at grain boundary and at the existed 90° DW. In SBT, because the existence of high density of APB which is not polarization domain wall but with distortion, the nucleation and growth of 90° domain along APB are much easier than that in PZT. According to the fatigue dynamics and the polarization switching mechanism, it is argued that APBs play a crucial role in the fatigue-free behavior in SBT.

SESSION BB5: POSTER SESSION DOMAIN-RELATED PHENOMENA IN FERROIC MATERIALS

Chairs: Antoni Planes and Venkatraman Gopalan
Tuesday Evening, November 28, 2000
8:00 PM
Exhibition Hall D (Hynes)

BB5.1

A STUDY OF OPTICAL PROPERTIES OF $\text{SrBi}_2\text{Ta}_2\text{O}_9$ THIN FILMS BY SPECTROSCOPIC ELLIPSOMETRY. D. Mo, Y. Liu, Department of Physics, Zhongshan University, Guangzhou, CHINA; J.B. Xu, G.D. Hu, Q.J. Li, K.Y. Wang and S.P. Wong, Department of Electronic Engineering and Material Science and Technology Research Center, The Chinese University of Hong Kong, Shatin, NT, Hong Kong, CHINA.

We have prepared the (200)-predominant and (0010)-predominant $\text{SrBi}_2\text{Ta}_2\text{O}_9$ films by metal organic decomposition (MOD). We have measured and analyzed ellipsometric spectra of these oriented films in the range of photon energy from 2 to 5.5eV. We find that the spectra of (200)-predominant SBT films show remarkable anisotropic effects. By analyzing the spectra, we have obtained the effective refractive index n and the extinction coefficient k of both SBT films. The n values between the (0010)-predominant and (200)-predominant SBT films show a large difference ($\Delta n \geq 0.2$). The band gap of the SBT thin films is found to be about 326 nm, i. e., 3.8eV. Also we have analyzed the correlation between the ellipsometric data and the results by the NSOM measurement.

BB5.2

ATOMIC SCALE ANALYSIS OF DOMAIN BOUNDARIES IN VACANCY-ORDERED BROWNMILLERITE-TYPE SrCoO_{3-x} . Yasuo Ito, Robert F. Klie, Nigel D. Browning, University of Illinois at Chicago, Dept of Physics, Chicago, IL; Terry J. Mazanec, BP Amoco Chemicals, Naperville, IL.

The high mobility of oxygen vacancies in oxygen-deficient perovskite-type oxides makes them suitable for applications as electrodes in solid oxide fuel cells and in oxygen separation membranes. In addition to the valence-state of the cations and clustering/ordering of oxygen vacancies, grain and domain boundaries also have a major influence to the performance of devices. As a model oxygen-deficient oxide, we have investigated oxygen-deficient SrCoO_{3-x} using the combination of atomic resolution electron energy-loss spectroscopy and the Z-contrast imaging in a scanning transmission electron microscope (STEM). The sample, which was stabilized by cyclic heat treatments in reducing and oxidizing atmospheres, consists of ordered microdomains where ordering of point defects occurs in every second cobalt-oxygen plane, and the

structure is brownmillerite-type. Domain boundaries in this structure are not sharp but interwoven across a few unit cells. Atomic resolution Z-contrast images along the $[010]_p$ axis shows that oxygen deficient planes of two domains meet each other at the boundary and perovskite-type units, defined by four Sr atoms, at the center of the boundary are distorted. These distorted units are periodically repeated along the boundary. Coupled with the distortion is an intermediate valence state of Co between that of the ordered planes in the bulk of the domains. This may indicate that the domain boundaries act as a branching point from one high ionic conductivity channel to another with various orientations intersect at the domain boundaries.

BB5.3

A MONTE-CARLO SIMULATION OF DYNAMIC HYSTERESIS AND DOMAIN SWITCHING IN MULTI-DOMAIN FERROELECTRIC LATTICE. J.-M. Liu, Z.-G. Liu, Nanjing University, Laboratory of Solid State Microstructures, Nanjing, CHINA.

The domain switching and dynamic hysteresis in multi-domain ferroelectric lattice are simulated using Monte-Carlo algorithm based on the DIFFOUR model. The possible scaling of the dynamic hysteresis against frequency and amplitude of the external field, domain size effect and remanence enhancement effect have been studied and discussed in details.

BB5.4

ROOM TEMPERATURE MAGNETIC IMAGING OF MICRO-METER-SIZED FERROMAGNETIC DOMAINS IN THE PRESENCE OF EXTERNAL MAGNETIC FIELDS BY SCANNING HALL PROBE MICROSCOPY. A. Sandhu, Tokai Univ, Dept of Electrical Engineering, JAPAN; H. Masuda, Toei Kogyo Ltd, Tokyo, JAPAN; A. Oral, Bilkent Univ, Dept of Physics, TURKEY; S.J. Bending, Univ. of Bath, Dept of Physics, UNITED KINGDOM.

The development of magnetic information storage and related technology requires a fundamental knowledge of the physical properties of magnetic domains. We will report on the room temperature imaging of surface magnetic field fluctuations of ferromagnetic structures using a scanning Hall probe microscope (SHPM) system that: (a) yields quantitative results (with measurements possible in the presence of very high external magnetic fields); (b) is highly sensitive (magnetic field fluctuation and planar/vertical spatial resolution); and (c) non-invasive. Samples studied included floppy disks (FD), ZIP media, MO disks, ferrite magnets and Bi substituted iron garnet epitaxial crystals. A micro-Hall probe (HP), fabricated using a GaAs/AlGaAs two-dimensional electron gas heterostructure grown by molecular beam epitaxy, was used as the magnetic field sensor. Photolithography was used to fabricate HPs having an active area width of 0.8 microns. The room temperature Hall coefficient and field sensitivity of the HP were 0.3 Ohm/G and 0.04 G/OHz, respectively. The corner of the HP chip was coated with a thin layer of gold to act as a scanning tunneling microscope (STM) tip. Measurements were made such that the HP was scanned over the sample surface while simultaneously monitoring the STM tunnel current thus enabling the analysis of magnetic field fluctuations in very close proximity to the surface of the samples. The room temperature SHPM images clearly showed: (1) magnetic transition regions of FDs to coalesce into small islands under external perpendicular fields exceeding 1500 Oe; (2) the garnet samples to exhibit configurational hysteresis where an initial magnetic bubble structure irreversibly transformed into a maze domain pattern under the action of a cyclic perpendicular magnetic field. We will describe the unique features of the newly developed SHPM system, elaborate on the aforementioned observations and attempt to correlate domain pattern changes observed by SHPM with related magnetization hysteresis (M-H) curves.

BB5.5

HIGH ELECTRIC FIELD CONDUCTIVITY IN MANGANITE PEROVSKITE THIN FILMS AND SINGLE CRYSTALS. Natalia Noginova, Norfolk State University, Norfolk, VA; Aleksandr Verevkin, Yale University, New Haven, CT; Edward Gillman, George B. Loutts, Empress Arthur, Norfolk State University, Norfolk, VA.

Simple high-electric field studies give the key to understanding the CMR materials microstructure. The high electric field phenomena are strictly affected by the material morphology. To verify this statement in CMR materials we studied the conductivity vs. lattice temperature dependences and the conductivity vs. electric field dependences in $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ ($x=0.3-0.4$) thin films, and $\text{LaGa}_{1-x}\text{Mn}_x\text{O}_3$ ($x=0.02-0.50$) single crystals. The high electric field conductivity features in the CMR films are in quantitative agreement with the multiphonon hopping model with the only adjustable parameter related to the average planar granular size d . The data for d obtained from the atomic force microscope analysis are very close to the

calculated values. This method can provide us a new simple microstructure characterization technique of the conductive oxide materials.

BB5.6

DYNAMICAL MODEL FOR HETEROGENEOUS NUCLEATION IN MARTENSITIC TRANSFORMATIONS. Rajeev Ahluwalia, Materials Research Laboratory, Pennsylvania State University, University Park, PA; G. Ananthkrishna, Materials Research Centre, Indian Institute of Science, Bangalore, INDIA.

We study the dynamics of martensitic transformations in the context of a two dimensional square to rectangle transition. Our model is based on an underlying free energy functional that incorporates the effects of long-range strain-strain interactions. The dynamical model is constructed by including the inertial effects associated with the displacement fields. The lagrangian formalism with appropriate dissipational functional yields an equation of motion for a coarse-grained strain order parameter. An inhomogeneous stress field is incorporated to describe the effect of lattice defects. These defects serve as nucleation centres for the transformation. Computer simulations of our model reveal morphological features similar to those observed in real systems. We have also investigated the thermal cycling of the transformation by continuous cooling and heating simulations. Thermal hysteresis is observed. We also calculate the rate of energy dissipation in both cooling and heating runs. We find that the energy dissipation occurs in jerks. Interesting, the distribution of amplitudes of the energy dissipation obeys a power-law, in accordance with acoustic emission experiments.

BB5.7

REAL-TIME OBSERVATION OF EVOLUTION OF MAGNETIC DOMAIN STRUCTURE AND ITS CORRELATION WITH ELASTIC DOMAINS IN CRYSTALS OF (La,Sr) MANGANITE PEROVSKITE AND SHAPE MEMORY ALLOY. Marina Turchinskaya, MS&E Lab, National Institute of Standards and Technology, Gaithersburg, MD; Alexander Roytburd, Dept of Materials and Nuclear Engineering, Univ of Maryland, College Park, MD; Sang-W. Cheong, Bell Labs, Lucent Technologies, Murray Hill, NJ; Vladimir Kokorin, Institute of Metal Physics, Kiev, UKRAINE; Mark Vaudin, Debra Kaiser, MS&E Lab, National Institute of Standards and Technology, Gaithersburg, MD.

The evolution of a magnetic domain structure and its relation to an elastic (twin) microstructure was observed using a magneto-optical imaging technique. The twin microstructure was observed using a polarizing microscope and was characterized by a backscattered electron diffraction in a scanning electron microscope. It was found that the magnetic domain structure during magnetization and demagnetization under an applied magnetic field of ± 130 mT at room temperature was developed within the framework of the steady-state twin microstructure. The domain fractions and the directions of the magnetic moments during remagnetization changed in accordance with hysteresis loops measured using a SQUID system. A correlation of the magnetic domain structure with the twin microstructure was also observed in a Ni_2MnGa shape memory alloy, demonstrating the similarity of the magnetic domain evolution under an applied magnetic field in these two crystalline materials. The interrelationship between the elastic and magnetic domains will be discussed.

BB5.8

IN-SITU X-RAY DIFFRACTION STUDIES OF PHASE TRANSFORMATION AND DOMAIN DEVELOPMENT DURING THE INITIAL COOLING OF EPITAXIAL PbTiO_3 THIN FILMS. J.A. Eastman, Carol Thompson^a, S.K. Streiffer, G.B. Stephenson, M.V. Ramana Murty, O. Auciello and G.-R. Bai, Materials Science Division, Argonne National Laboratory, Argonne, IL; A. Munkholm, Chemistry Division, Argonne National Laboratory, Argonne, IL. ^aPrimary affiliation: Department of Physics, Northern Illinois University, Dekalb, IL.

We are examining the growth and phase-transition behavior of epitaxial PbTiO_3 thin films grown on (001) SrTiO_3 substrates. A new, unique processing facility at the Advanced Photon Source allows us to grow PbTiO_3 films by metal-organic chemical vapor deposition (MOCVD) while simultaneously characterizing their growth behavior and structure by x-ray diffraction. While bulk PbTiO_3 undergoes a cubic-to-non-centro-symmetric-tetragonal phase transition when cooled below 490°C , the behavior of epitaxial thin films can be significantly different due to lattice matching of the film to the substrate. In the present study we were able, for the first time, to characterize approximately 30 nm-thick PbTiO_3 films grown at temperatures $>650^\circ\text{C}$ prior to the initial cooling through the phase transition to the ferroelectric phase. These films grow fully coherent with the substrate and thus are tetragonal at all temperatures. Investigations of the centro-symmetric-tetragonal to non-centro-symmetric-tetragonal phase transition and of strain-induced

ferroelectric domain formation during the initial cooling of samples will be presented.

This work is supported by the U.S. Department of Energy, Office of Science, under Contract W-31-109-Eng-38, and by the State of Illinois, under HECA.

BB5.9

DYNAMIC CHARACTERIZATION OF MAGNETIC DOMAIN STRUCTURES IN SOFT FERROMAGNETIC MATERIALS. K.L. Garcia and R. Valenzuela, Institute of Materials Research, National Univ of Mexico, MEXICO.

Ferromagnetic domain structures, formed essentially by domains separated by domain walls lead to several magnetization processes, such as domain wall bulging (elastic deformation of domain walls, pinned to material's defects), domain wall displacement (for fields large enough to unpin the walls; these irreversible displacements lead to magnetic hysteresis) and spin rotation (change of precession axis as a result of the applied field). Since each of these processes possess a different dynamics, or time constant, they can be separated by means of frequency measurements. Also, their dispersions (i.e., the way each of them becomes unable to follow the excitation field as frequency increases) show clear differences. In this paper, we present a systematic study of the frequency behavior of some amorphous ferromagnetic alloys, in the form of wires and ribbons, which can be known as "Inductance Spectroscopy". We use the complex inductance formalism, especially convenient for magnetic studies, since inductance is proportional to magnetic permeability. We make use also of equivalent circuits to model experimental results; in some cases, we show that a direct association between equivalent circuit elements and physical parameters of the sample can be established. The main experimental tool is a system including a HP 4192A Impedance Analyzer. The excitation field can be applied by two different geometries: longitudinal, by using a 220-turn solenoid, or the GMI geometry, where an ac current flows through the sample, and the excitation field, with a circumferential or transverse geometry is produced by the current itself. An evaluation of the size and geometry of domains, as well as the pinning strength of domain walls can be carried out.

BB5.10

TEMPERATURE AND FREQUENCY DEPENDENCE OF THE DIELECTRIC CONSTANT OF RELAXATION FERROELECTRICS. H.K. Guo, Y.N. Wang, F. Yan, National Laboratory of Solid State Microstructures and Department of Physics, Nanjing University, Nanjing, PR CHINA.

A relationship describing temperature and frequency dependence of dielectric constant in relaxor ferroelectrics is derived from a exponential distribution model of relaxation times, and found to be in good agreement with experimental results. We propose a new model of dipolar correlation, which is the physical origin of the broad distribution of relaxation times. We discuss the physical meaning of the activation energy E in the relaxor ferroelectrics, when the dipolar correlation of local polar regions is taken into account.

BB5.11

X-RAY POWDER DIFFRACTION INVESTIGATIONS OF Ga-Mn-Ni SHAPE MEMORY ALLOY BULK MATERIAL. B. Wedel, DOWA Mining Company Ltd., Central Research Laboratory, Hachioji, JAPAN; C. Wedel, K. Itagaki, Tohoku University, Institute for Advanced Materials Processing, Sendai, JAPAN.

The shape memory effect combined with the magnetic properties of Ni-Mn-Ga alloys opens an interesting field of future applications. It is known that the phase $\text{Ni}_{50}\text{Mn}_{25}\text{Ga}_{25}$ and the non-stoichiometric alloys $\text{Ni}_x\text{Mn}_y\text{Ga}_z$ (with $x = 47-55$, $y = 20-26$ and $z = 23-26$) are a ferromagnetic Heusler alloy with the L_{21} structure and undergo a thermoelastic martensitic transformation to a tetragonal low temperature phase. With increasing nickel content in the non-stoichiometric alloys the transformation temperature increases up to room temperature.

The martensitic phase transformation was observed with an X-ray diffraction study of the high and low temperature phase of bulk material. A powder pattern analysis and a whole profile pattern analysis were made to identify the space group atomic layer and to construct a structure model for the low temperature phases of these alloys and compare these models with previous results. The results of this analysis led to a new structure type for the non-stoichiometric alloys for the stoichiometric structure with similarity to the AuCu type and to a superlattice of the AuCu type.

The alloys show the expected cubic and tetragonal basic units. For the $\text{Ni}_{50}\text{Mn}_{25}\text{Ga}_{25}$ high temperature phase the known cubic symmetry, space group $Fm\bar{3}m$, with the cell parameters: $a = 0.5826(2)$ nm, $Z = 4$, was detected. For the low temperature phase at liquid nitrogen temperature, a tetragonal symmetry, space group I_4/mmm , with the cell parameters: $a = 0.4186(1)$ and $c = 0.5562(1)$

nm, $Z = 2$, was found. The $\text{Ni}_{55}\text{Mn}_{20}\text{Ga}_{25}$ alloy shows the same tetragonal symmetry, with the cell parameters: $a = 0.3841(1)$ and $c = 0.6563(2)$ nm, $Z = 2$ at room temperature. The c -axis varied with different Nickel contents between 0.55 and 0.66 nm. Further the martensitic twinning and impurities are observed. These results are compared with TEM observations. The compositions were checked with EPMA and chemical analysis methods.

BB5.12

STRUCTURE - PROPERTY RELATIONS ACROSS THE ANTI-FERROELECTRIC/FERROELECTRIC PHASE BOUNDARY IN LA DOPED PbZrO_3 - PbTiO_3 SOLID SOLUTIONS. Jesper Knudsen, Ian M. Reaney, Univ of Sheffield, Dept of Engr Matls, Sheffield, UNITED KINGDOM; Jonathan Fitzmaurice, Mechanical Science Sector, DERA, Farnborough, UNITED KINGDOM.

Antiferroelectric/ferroelectric (AFE/FE) switching can be induced in lead zirconate titanate (PZT) based compositions close to the lead zirconate (PZ) end of the PZT phase diagram. Most commercial compositions are doped with either La, Sn or Nb to decrease the free energy between the FE and AFE phases. It has been suggested that switching is optimised if compositions have an AFE tetragonal as opposed to the AFE orthorhombic structure, more commonly associated with PZ. Recent work has suggested that the AFE tetragonal phase in La doped PZT compositions is, in reality, an incommensurate AFE orthorhombic phase and not a unique structure. The mixed oxide route, utilising attrition milling, has been used to produce dense (>95%) ceramics with the general formula $\text{Pb}_{1-3x/2}\text{La}_x[\square]_{x/2}(\text{Zr}_{1-y}\text{Ti}_y)\text{O}_3$ ($[\square]$ = vacancy), to investigate the microstructure/structure - property relations across the AFE/FE phase boundary. X-ray diffraction has been used to investigate macroscopic changes in structure as a function of La concentration (x) and Zr:Ti ratio (y). Microscopic changes in structure and domain structure were monitored as a function of composition and temperature using conventional and in-situ transmission electron microscopy, respectively. Dielectric properties were also studied as a function of temperature. This work was carried out as a part of a CASE funded studentship of Technology Group 1 of the MoD corporate research programme. Crown copyright 2000. Published with the permission of the Defence Evaluation Research Agency on behalf of the Controller of HMSO.

BB5.13

DIFFRACTION ANOMALOUS FINE STRUCTURE AS UNIQUE STRUCTURAL TOOL TO CHARACTERIZE DOMAINS IN SUPER-ORDERED GaInP. Lucilla Alagna, Stefano Turchini, Tommaso Proserpi, ICMAT-CNR, Rome, ITALY; Angelo Mascarenhas, NREL, Golden, CO.

We performed successfully DAFS measurements at the Gallium K-edge, probing super-ordering in InGaP/GaAs epitaxial growth. The sample chosen was lattice matched InGaP MOCVD epitaxially grown on GaAs with a miscut angle of 6 degrees respect to the (001) direction and exhibited (111)-type alloy ordering of alternating InP and GaP-like planes and giving rise to (-5/2, 5/2, -5/2) Bragg peak reflection which becomes allowed. The data analysis allows to extract with an EXAFS-like study, the local structure of the Gallium in the epi-layer, comparing the information averaged on the whole epi-layer and the information arising from the super-ordered local domain. The domains are accessible to EXAFS-like study, using the collected DAFS data and some results will be presented showing that the Gallium environment within the domains. We will show a comparison, both experimental and calculated, between the Diffracted Anomalous Fine Structure of an allowed and of a forbidden reflection for the same sample, the latter arising from a local arrangement. The theoretical approach followed is capable of reproducing the experimental results for the forbidden reflection. We demonstrate how the spatial and site selectivity for the same element (here Ga) can be used to distinguish between a situation due to an arrangement in domains and one involving long range ordering. For the first time structural data are available, at the same time, for the surroundings of Gallium in the bulk and in the epi-layer from allowed reflections, while the forbidden reflection gives structural details around Gallium in the ordered domains.

BB5.14

INFLUENCE OF AXIAL PRESSURE ON DIELECTRIC PROPERTIES OF NaNbO_3 SINGLE CRYSTALS AND CERAMICS. Krzysztof Konieczny, Institute of Physics, Pedagogical Akademia, Krakow, POLAND.

The dielectric properties for sodium niobate NaNbO_3 single crystals and ceramics in the temperature range 300K - 750K and for frequencies of external measurement electric field in range 0.1 kHz - 20 kHz were investigated. The axial pressure in range 0 GPa - 1 GPa was applied to the sample parallel or perpendicular to the direction of measurement electric field. A strong dependence of value of dielectric

permittivity on axial pressure and in the case of single crystals a diffusing of the antiferroelectric - paraelectric phase transition was observed. Such behaviour of this material was connected with domain kinetics. Around 550K was observed the anomaly in the dependence of dielectric permittivity on temperature. This anomaly is dependent on axial pressure. The experimental data have shown the possibility of inducing the ferroelectric state in antiferroelectric material by axial pressure.

BB5.15

CHARGE ORDERING PHENOMENA IN THE COLOSSAL MAGNETORESISTIVE MANGANITES. D.J. Miller, Z.P. Luo, J.F. Mitchell, Matls Sci Division, Argonne Natl Lab, Argonne, IL.

Charge ordering (CO) in the colossal magnetoresistive manganites gives rise to an insulating, high-resistance state that can be melted into a low-resistance metallic-like state by the application of magnetic field. Thus, the potential to attain high values of magnetoresistance with the application of small magnetic fields may be aided by a better understanding of the charge ordering phenomenon. This study focused on microstructural characterization during charge ordering of both the perovskite and the layered manganites. In $\text{Nd}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$, a commensurate charge ordered supercell of $a = 2\sqrt{2}ap$, $b = 2ap$, and $c = \sqrt{2}ap$ forms via nucleation and growth upon cooling. Twins and other defects are mild barriers for charge ordering. In addition, some regions remain untransformed even to the lowest temperatures studied, and these regions may provide a link to compositional or structural variations. In the layered compound $(\text{La,Sr})_3\text{Mn}_2\text{O}_7$, charge ordering was studied as a function of stoichiometry for compositions near $\text{LaSr}_2\text{Mn}_3\text{O}_7$. CO behavior similar to that observed in single crystals was observed. However, the CO structure varied based on composition. Details of the CO structure and the compositional dependence will be discussed. This work was supported by the U.S. Department of Energy, Basic Energy Sciences-Materials Sciences, under contract #W-31-109-ENG-38.

BB5.16

EFFECT OF DOMAIN ORIENTATION AND APPLIED TEST VOLTAGE ON PTCR BEHAVIOR: PART I. Rodney Roseman, Niloy Mukherjee, University of Cincinnati, Department of Materials Engineering, Cincinnati, OH.

Semiconducting, PTCR barium titanate shows a directional alignment of the ferroelectric domain structure throughout the expanse of individual grains, the orientation, though, is random from grain to grain. A high temperature poling of these type materials results in alignment of the domain structure in a coherent nature in direction of the applied voltage, resulting in a highly suppressed resistivity transition in direction of the alignment and typical PTCR behavior in the anti-poled direction. The degree of poling is dependent on previous heat treatment. A high degree of domain orientation and corresponding resistivity suppression can also be obtained through changes in stoichiometry and processing routes. The semiconducting behavior and resistivity transitions can be directly associated with domain structure which is also a function of grain size, extent of grain-to-grain contact area and stress at the grain boundary.

BB5.17

EFFECT OF DOMAIN ORIENTATION AND APPLIED TEST VOLTAGE ON PTCR BEHAVIOR: PART II. Rodney Roseman, Niloy Mukherjee, University of Cincinnati, Cincinnati, OH.

Our previous studies have shown that domain structure orientation plays a dominant role in determining PTCR behavior in barium titanate. Although, increasing test voltage is well known to suppress the resistivity transition, this study has found that the voltage sensitivity of the PTCR effect is dependent on the domain orientation and grain-to-grain contact area. In samples that have a large degree of domain orientation across grain boundaries and a large amount of grain contact area, the initial PTCR effect is low and the voltage sensitivity of the base and transition resistivity is high. The resistivity transition can be practically suppressed with relatively low applied fields. In contrast with samples that demonstrate significant PTCR behavior and low degree of domain orientation, the voltage sensitivity is low. These studies have shown that the observed potential barrier can be directly related to the domain structure at the grain boundary which is related to the defect states and space charge in the near boundary regions. The observed voltage sensitivity of different samples must be a function of both the mechanical and space charge state of the grain boundary.

BB5.18

EFFECTS OF ANISOTROPIC EIGENSTRAIN TRANSFORMATIONS ON POINT-DEFECT INTERACTIONS. Igor Dobovsek, University of Maribor, Faculty of Mechanical Engineering, Maribor, SLOVENIA.

For stress and strain fields around point defects in an elastic crystal induced by anisotropic eigenstrain transformations we investigate a mutual dependence among defects and their interaction in resulting deformation fields. Such an interaction is a consequence of non-zero deviatoric strain, since for fields that are strongly anisotropic strains cannot be reduced to pure dilatation and consequently an interaction energy is not zero. Point defect is simulated using the Dirac-delta function and a general, constant, at the point of action spatially independent second order tensor describing the anisotropies of the defect. Possible implications of the derived strain and stress fields in the analysis of dipole strains induced by interstitial atoms in the b.c.c. lattice models are discussed as well.

BB5.19

PHASE TRANSITIONS IN DIPOLAR SYSTEMS WITH MODULATED SHORT-RANGED ATTRACTIONS AND LONG-RANGED REPULSIONS. Girija S. Dubey, Department of Natural Sciences, York College of City University of New York, Jamaica, NY; Ras B. Pandey, Department of Physics, University of Southern Mississippi, Hattiesburg, MS.

We report extensive Gibbs ensemble simulations of the dipolar system. The long-range interaction is summed up using the Ewald summation technique. By scaling the attractive $(\sigma/r)^6$ term in the Lennard-Jones pair potential by a factor λ ranging from 0 to 1. We try to find out for what values of λ including $\lambda = 0$ and dipolar strength, one can observe the vapor-liquid coexistence in the simulation. Our result suggests that the phase transition in dipolar fluids is related to the strength of the dipole moment relative to the depth of the Lennard-Jones potential well.

BB5.20

HYSTERESIS IN LaCoO₃ BASED FERROIC PEROVSKITES. Nina Orlovskaya, Yury Gogotsi, Drexel University, Dept of Materials Engineering, Philadelphia, PA.

The work characterizes nonelastic ferroelastic behavior of LaCoO₃ based perovskites. These perovskites are considered as a promising material for high temperature oxygen separation membrane application and therefore is an important industrial material. The unusual mechanical behavior of cobaltites was observed. Nonelastic behavior is observed during bending tests. Hysteresis loops are obtained during bending tests. Ferroelastic behavior was observed during contact loading (indentation) tests. The nano- (Berkovich diamond indenter) and microindentation (cone diamond indenter) hysteresis curves are analyzed as a function of perovskite composition and an applied pressure. Plasticity of LaCoO₃ based perovskite was evaluated by analyzing the loading-unloading curves during indentation. Fracture toughness of LaCoO₃ based materials was measured by SEVNB method. The kinks on the load-time curve during K_{Ic} measurements was observed that can lead to increased absorption of energy during fracture. The possible toughening mechanisms of LaCoO₃ based perovskites are discussed. The visualization of domains was done by atomic force and laser confocal microscopy, which allowed estimating the size of switched/transformed zone around impressions. The investigation of domain structure was done by TEM at RT and up to 500°C, which allowed to determine the ferroelastic to paraelastic transition temperature of La_{0.6}Ca_{0.4}CoO₃ perovskite.

BB5.21

NANODOMAIN DYNAMICS AND THEIR ROLE IN ELECTROMECHANICAL RESPONSE OF HIGH-STRAIN A-SITE PEROVSKITE RELAXORS. Andrey N. Soukhojak, Yet-Ming Chiang, Massachusetts Institute of Technology, Dept of Materials Science and Engineering, Cambridge, MA.

It is generally recognized that the presence of nanodomains is a characteristic feature of relaxors. Lead-based B-site relaxors with perovskite structure have been extensively studied due to their exceptionally high electric field-induced strain and electromechanical coupling coefficients. Our research is focused on alternative lead-free A-site relaxors in the alkaline bismuth titanate family, which recently have been shown to possess highly promising electromechanical properties in both single crystal and polycrystalline form. In addition to large piezoelectric and electrostrictive coefficients multiple samples exhibited pronounced electrorheological behavior (<40 ms time constant of electrostrictive response). We present optical, HREM and STEM observations of both macro- and nanodomains in samples of different doping levels, and show how the electromechanical response depends on the domain structure. A newly developed model of nanodomain dynamics is presented. It explains the origin of time-dependent electromechanical response of relaxors. Research supported by ONR Grant No. N00014-97-0989.

BB5.22

SPIN POLARIZED AUGER ELECTRON SPECTROSCOPY OF MAGNETIC MATERIALS. Onder S. Anilturk, Ali R. Koymen, University of Texas at Arlington, Physics Dept., Arlington, TX.

Surface sensitive experiments, in which the spin-polarized electrons are involved, play an important role for magnetic characterization, since the spin-polarized electrons are fingerprints for the local magnetization. Scanning Electron Microscope with Polarization Analysis (SEMPA) is one of the most powerful tool to investigate the surface magnetic domain microstructures of magnetic materials. On the other hand, at energies enough to generate two-hole final state arising from Auger transitions, it is possible to observe the spin polarization of Auger electrons which reveal element-specific local magnetic information, particularly valuable for surface magnetic studies with composite systems. By using the uniqueness of the UTA-SEMPA tool, one can obtain the magnetic domain picture and also perform Spin Polarized Auger Electron Spectroscopy (SPAES) studies, specific to the domain image of the material, by probing to single domain at the surface. In this study, precisely knowing the probed domain, spin polarization of electrons from super Koster-Kronig MMM Auger emission on Fe whisker, Gd(x)Ni(1-x) alloy and Ni samples have been investigated. With these transitions involving the valence bands, it is shown that there is a considerable polarization enhancement above 3p threshold.

BB5.23

SUPERFAST DOMAIN KINETICS IN FERROELECTRICS. Vladimir Ya. Shur, Evgenii L. Rumyantsev, Ekaterina V. Nikolaeva, Eugene I. Shishkin, Alexander P. Chernykh, Ural State Univ., Inst. of Phys. & Appl. Math., Ekaterinburg, RUSSIA.

The new mechanism of superfast switching was proposed on the basis of the investigation of domain kinetics by in situ visualization of instantaneous domain patterns in single crystals of congruent lithium tantalate (LT). The proposed model of domain evolution was verified by computer simulation of domain kinetics. We simulated the wall motion, which represents the sequence of the step generation acts through wall merging with individual isolated domains and rapid growth of arisen steps. We have observed that the switching from single-domain state in LT starts with arising of a great number of small domains. The following growth of isolated domains is very slow (about one micron per second), but their merging leads to formation of large rapid domain walls with motion velocity up to 130 microns per second. The rapid wall motion was investigated in details by computer simulation. We have shown the possibility of formation of super-mobile walls possessing the constant step density. The simulated wall motion velocity dependence on wall length and initial nucleation density is in accord with experimental data. Computer simulation allowed to reveal five different regimes of the domain kinetics during analysis of the dependence of the switching current parameters on nucleation density. The jump-like domain reconstruction as a result of merging with large stable domains has been demonstrated. Two inputs in switching current data have been considered. The initial decrease of switching current corresponds to merging of nuclei and can be fitted by the power law. The main part of the current corresponds to conventional growth of small number of large domains and can be fitted by modified Kolmogorov-Avrami formula. The research was made possible in part by Program "Basic Research in Russian Universities" (Grant No.5563) and by Grant No.97-0-7.1-236 of the Ministry of Education of the Russian Federation.

SESSION BB6/CC8: JOINT SESSION DOMAIN STUDY IN FERROELECTRIC THIN FILMS

Chairs: Wenwu Cao and Ramamoorthy Ramesh
Wednesday Morning, November 29, 2000
Room 312 (Hynes)

8:30 AM *BB6.1/CC8.1

DOMAIN STRUCTURE AND SWITCHING IN FERROELECTRIC FILMS OBSERVED BY AFM. Angus I. Kingon, NCSU, Department of MS&E, NC; B. Rodriguez and R. Nemanich, NCSU, Department of Physics, NC; C.B. Parker, D.J. Kim and J.-P. Maria, NCSU, Department of MS&E, NC.

Ferroelectric nonvolatile memories are currently entering production. Despite the very significant development of the films and devices over the past many years, there remains a great deal which is not known regarding the ferroelectric domain structure, and the role of the ferroelectric domain walls in determining device switching characteristics and device lifetimes. In the first part of the presentation we describe methods used to characterize the spatial variation of properties of the ferroelectric capacitors by characterizing the spatial distribution of the piezoelectric properties (by AFM). We

deduce that the scale of the distribution is far smaller than the grain size, for the case of PZT films. We discuss the significance of the results in terms of the statistical methods used, and the role of domain walls, and contrast the results to previously reported results for SBT. We show that the spatial distribution of properties is markedly increased after fatigue. However, the size of the fatigued regions is generally submicron. We discuss the implications for scaling of PZT-based nonvolatile memories to higher densities. Secondly, we describe experiments in which the extent of local switching is characterized as a function of a single voltage pulse, for samples which include the variables: PZT composition, film orientation, film microstructure, domain wall density, electrode type, capacitor dimensions, and prior fatigue cycling. Implications are drawn for high density devices.

9:00 AM *BB6.2/CC8.2

ELECTROMECHANICAL RESPONSE OF UNPOLED FERROELECTRIC STRUCTURES. Alexander Tagantsev, Olivier Steiner, Ceramics Laboratory, EPFL, Swiss Federal Institute of Technology Lausanne, SWITZERLAND.

The piezoelectric coupling, which basically controls the electromechanical response of poled ferroelectric materials, is averaged down to zero in unpoled structures. Under these conditions, the link between the strain and the electric field becomes quadratic and can be described in terms of an effective electrostrictive coupling. This coupling is controlled by various contributions which are related to variation of the domain structure (extrinsic contribution) and to the anharmonicity of the crystalline lattice of the material (intrinsic contribution). The goal of this talk is to discuss these contributions with an accent on the intrinsic one. Here two results, which do not match the routinely accepted general picture of phenomenon, are reported. First, there is no violation of the correspondence between the direct and converse electrostrictive effects when passing from the paraelectric to ferroelectric phase of the material as it has been proposed by Zang et al. [Q.M. Zang, W.Y. Pan, S.J. Jang, and L.E. Cross, *Ferroelectrics* v. 88, 147 (1988)]. Second, the effective intrinsic longitudinal electrostrictive coefficient of polydomain structure of a perovskite ferroelectric, Q_{33} , can be negative.

9:30 AM BB6.3/CC8.3

DOMAIN SWITCHING KINETICS OF PZT THIN FILMS AT HIGH FREQUENCIES. Takaaki Tsurumi, Song-Min Nam, Young-Bae Kil and Satoshi Wada, Tokyo Inst. Technology, Dept. of Metallurgy & Ceramics Science, Tokyo, JAPAN.

Domain switching behavior of sol-gel derived PZT thin films at high frequencies have been investigated for ferroelectric memory applications. A measuring apparatus of D-E hysteresis curves was developed using a voltage-current converter with virtual ground circuit. The coercive field of the PZT thin films strongly depended on the measuring frequency, nevertheless their remanent polarization was almost independent of it. The coercive field was also dependent on temperature and electrode area. The domain switching kinetics of PZT thin films could be explained using a nucleation-controlled model. A linear relation was obtained between $\ln \nu$ and $1/E_c^2$, where ν is a frequency and E_c is a coercive field. The intercept at $1/E_c^2=0$ is a limiting frequency of domain switching. The limiting frequency of domain switching ($\ln \nu_0$) increased with decreasing electrode area. The slope of the line was determined by a binding energy between domain wall and defects, domain wall energy and the change in the polarization with domain wall motion. From the results obtained in this study, a guideline to develop ferroelectric films was proposed for ferroelectric memories with high speed and low operating voltage.

9:45 AM BB6.4/CC8.4

ABRUPT APPEARANCE OF THE DOMAIN PATTERN AND FATIGUE OF THIN FERROELECTRIC FILMS. A.M. Bratkovsky, A.P. Levanyuk, Hewlett-Packard Laboratories, Palo Alto, CA.

We study the domain structure in ferroelectric thin films with a "passive" layer (material with damaged ferroelectric properties) at the interface between the film and electrodes within a continuous medium approximation. An abrupt transition from a monodomain to a polydomain state has been found with the increase of the "passive" layer thickness d . The domain width changes very quickly at the transition (exponentially with d^{-2}). We have estimated the dielectric response dP/dE (the slope of the hysteresis loop) in the "fatigued" multidomain state and found that it is in agreement with experiment, assuming realistic parameters of the layer. We derive a simple universal relation for the dielectric response, which scales as $1/d$, involving only the properties of the passive layer. This relation qualitatively reproduces the evolution of the hysteresis loop in fatigued samples and it could be tested with controlled experiments. It is expected that the coercive field should increase with decreasing lateral size of the film. We believe that specific properties of the

domain structure under bias voltage in ferroelectrics with a passive layer can resolve the long-standing "paradox of the coercive field".

10:30 AM *BB6.5/CC8.5

FERROELECTRIC THIN FILMS: NANOSCALE CHARACTERIZATION BY SCANNING FORCE MICROSCOPY. Alexei Gruverman, Sony Corporation, Yokohama, JAPAN.

In this paper, results of nanoscale characterization of ferroelectric thin films and capacitors by means of scanning force microscopy (SFM) will be presented. Polarization reversal dynamics and degradation effects, such as fatigue and retention loss, were studied in ferroelectric thin films via direct observation of their domain structures using the SFM piezoresponse method. SFM approach allowed direct nanoscale studies of correlation between crystallinity, domain structure and switching behavior of the ferroelectric films. Results of comparative nanoscale studies of SBT films grown by different techniques will be presented. It will be shown that the SFM data are consistent with the macroscopic parameters. SFM can be used for studying nanoscale variations of switching parameters in ferroelectric films and for investigating the scaling effect on switching performance of submicrometer ferroelectric capacitors. It will be demonstrated that for implementation of reliable high-density ferroelectric memories a certain capacitor/grain size ratio should be maintained. Particular attention will be given to the investigation of the mechanism of polarization retention loss in ferroelectric films. The retention behavior of ferroelectric films was studied as a function of switching conditions, electrode material, surrounding domain patterns and sample prehistory. SFM allowed direct mapping of leakage sites and nanoscale investigation of electrical conduction mechanism at these sites. Results of SFM measurements of leakage current characteristics of submicrometer ferroelectric capacitors will be presented.

11:00 AM BB6.6/CC8.6

GROWTH AND CONTROL OF DOMAIN STRUCTURE OF EPITAXIAL $PbZr_{0.2}Ti_{0.8}O_3$ FILMS GROWN ON VICINAL (001) $SrTiO_3$. V. Nagarajan, C.S. Ganpule, S.P. Alpay, A. Roytburd, R. Ramesh, Univ of Maryland, Dept of Materials Engineering, College Park, MD; D.G. Schlom, Pennsylvania State University, Dept of Materials Engineering, University Park, PA.

Highly tetragonal, epitaxial PZT films with a nominal composition of $PbZr_{0.2}Ti_{0.8}O_3$ exhibit a 2-dimensional grid of 90° domains (a-domains, i.e., c-axis in the plane of the film). Our previous studies have revealed that the 90° domains are preferential sites for the nucleation of 180° reverse domains during polarization switching and relaxation. Furthermore, we have observed that this array of 90° domains effectively isolate neighboring c-axis oriented regions. Therefore, we are studying approaches to control the spacing and periodicity of the 90° domains. Such self-assembled arrays of periodic domain structures can form the templates for novel memory arrays. In this paper, we report on the use of vicinally cut [along [100], [010], and [110] directions in the substrate plane] single crystal substrates to control the 90° domain formation. Epitaxial thin films have been deposited by pulsed laser deposition onto the vicinal substrates, with epitaxial conducting oxide bottom electrodes (LSCO and SRO). We have been able to control the nucleation of the 90° twins to occur preferentially at the steps on the substrate. We show that the orientation of these domains can be controlled such that they exhibit only 2 of the 4 possible variants. By using 4-circle x-ray diffraction, TEM and Electric Force Microscopy (EFM) we have investigated the structural and electrical properties of these artificially engineered structures. The control of such structures as a function of film thickness and substrate miscut orientation will be presented.

11:15 AM BB6.7/CC8.7

DOMAINS IN $SrBi_2Nb_2O_9$ AND $SrBi_2Ta_2O_9$ FERROELECTRIC FILMS. M.A. Zurbuchen, J. Lettieri, Y. Jia, G. Asayama and D.G. Schlom, Penn State Univ, Dept of Materials Science and Engineering, University Park, PA; S.K. Streiffer, Argonne National Laboratory, Materials Science Division, Argonne, IL; M.E. Hawley, Los Alamos National Laboratory, Materials Science and Technology Division, Los Alamos, NM.

We recently reported $SrBi_2Nb_2O_9$ films with the highest remanent polarization value attained to date in $SrBi_2Nb_2O_9$ or $SrBi_2Ta_2O_9$ films, $P_r = 15.7 \mu C/cm^2$ [1]. This was achieved by tilting the c-axis of $SrBi_2Nb_2O_9$ by 57° from the substrate surface normal in order to get a significant component of the polar axis, the a-axis, aligned with the direction of the applied electric field in these (103) $SrBi_2Nb_2O_9$ / (111) $SrRuO_3$ / (111) $SrTiO_3$ epitaxial films. In this talk, the microstructural features of these films, revealed by high-resolution and dark-field transmission electron microscopy (TEM), are reported, including domains, domain boundaries, domain populations, and out-of-phase boundaries. Portions of the same films used for electrical characterization were examined by TEM. Films grown in a 3-fold twin structure on the 3fold symmetric (111) $SrTiO_3$ surface. Dark-field

TEM imaging over a $12 \mu\text{m}^2$ area shows no evidence of second phases (crystalline or amorphous), which is important for high-density FRAM applications.

[1] J. Lettieri, M.A. Zurbuchen, Y. Jia, D.G. Schlom, S.K. Streiffer, and M.E. Hawley, *Appl. Phys. Lett.* v76, p2937 (2000).

11:30 AM **BB6.8/CC8.8**

CELLULAR DOMAIN ARCHITECTURE OF STRESS-FREE EPITAXIAL FERROELECTRIC FILMS. S.P. Alpay, A.L. Roytburd, V. Nagarajan, Univ of Maryland, Dept of Materials and Nuclear Engineering, College Park, MD; L.A. Bendersky, National Institute of Standards and Technology, Materials Science and Engineering Laboratory, Gaithersburg, MD; R. Ramesh, Univ of Maryland, Dept of Materials and Nuclear Engineering, College Park, MD.

Epitaxial ferroelectric films undergoing a cubic-tetragonal phase transformation relax internal stresses due to the structural phase transformation and the difference in the thermal expansion coefficients of the film and the substrate by forming polydomain structures. The most commonly observed polydomain structure is the $c/a/c/a$ polytwin which relieves the internal stresses only partially. Relatively thicker films may completely reduce internal stresses if all three variants of the ferroelectric phase are brought together such that the film has the same in-plane size as the substrate. We provide experimental evidence on the formation of the 3-domain structure based on transmission electron microscopy in 450 nm thick (001) PZT (20/80) films on (001) strontium titanate substrate grown by pulsed laser deposition. X-ray diffraction studies show that the film is fully relaxed. Experimental data is analyzed in terms of a domain stability map. It is shown that the observed structure in epitaxial ferroelectric films is due to the interplay between relaxation by misfit dislocations at the deposition temperature and relaxation by polydomain formation below the phase transformation temperature. The effect of the domain structure on the switching characteristics and physical properties is discussed.

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11:45 AM **BB6.9/CC8.9**

NEAR-FIELD OPTICAL SECOND HARMONIC IMAGING OF THE POLYDOMAIN STRUCTURE OF EPITAXIAL $\text{PbZr}_x\text{Ti}_{1-x}\text{O}_3$ THIN FILMS. I.I. Smolyaninov, H.Y. Liang, C.H. Lee, C.C. Davis, Univ of Maryland, Electrical and Computer Engineering Dept, College Park, MD; V. Nagarajan, C. Ganpule, R. Ramesh, Univ of Maryland, Dept of Materials and Nuclear Engineering, College Park, MD; E. Williams, Univ of Maryland, NSF Materials Research and Science Engineering Center, College Park, MD.

Near-field optical second harmonic microscopy¹ has been applied to imaging of the $c/a/c/a$ polydomain structure of epitaxial $\text{PbZr}_x\text{Ti}_{1-x}\text{O}_3$ thin films in the $0 \leq x \leq 0.4$ range. Comparison of the near-field optical images and the results of AFM and x-ray diffraction studies show that the optical resolution of the order of 80 nm has been achieved. Symmetry properties of the near-field second harmonic signal allow us to obtain good optical contrast between the local second harmonic generation in c - and a -domains. Experimentally measured near-field second harmonic images have been compared with the results of theoretical calculations. Good agreement between theory and experiment has been demonstrated. Thus, novel optical technique for nanometer scale ferroelectric domain imaging has been developed. Its main advantage with respect to the other scanning probe techniques is the possibility of fast time resolved measurements using optical pump and probe technique.

¹ I.I. Smolyaninov *et al.*, *Opt. Lett.* **25**, 835 (2000).