SYMPOSIUM D

Polarization Dynamics in Ferroic Materials

November 27 – 29, 2001

Chairs

Ramamurthy Ramesh
Materials Engineering
Univ of Maryland
College Park, MD 20742
301-405-7364

Venkatraman Gopalan
Dept of MS&E
Pennsylvania State Univ
253 MRL
University Park, PA 16802
814-865-2910

Michael E. McHenry
Dept of MS&E
Carnegie Mellon Univ
2315 Wean Hall
Pittsburgh, PA 15213-3890
412-268-2703

Symposium Support
Army Research Office

* Invited paper

SESSION D1: SWITCHING DYNAMICS
Chair: Venkatraman Gopalan
Tuesday Morning, November 27, 2001
Room 205 (Hynes)

8:30 AM *D1.1
DYNAMICS OF FERROELECTRIC THIN-FILM SWITCHING.
J.F. Scott and M. Dawber, Centre for Ferroics, Earth Sciences Department, Cambridge University, Cambridge, UNITED KINGDOM.

First we point out that Poisson's Equation for charge density as a function of depth z from the electrode interface is given by $\rho(z) = \nabla \cdot D = (\varepsilon \nabla \cdot E) = \varepsilon E + E \cdot \nabla \varepsilon$, and the second term (often neglected) is typically 100x greater than the first. This is largely due to the oxygen vacancy concentration gradient. We show in $SrTiO_3$ that this gradient can be fitted to the classical $\exp(-z^{6/5})$ dependence for diffusion at the processing temperature and is quenched in at ambient temperatures. This effect is largely responsible for an n-type inversion layer at the surfaces of most (nominally p-type) ferroelectric oxides, as recently confirmed by the AFM data of Gruverman. Second, we find experimentally that the frequency dependence of the coercive field $E_c(f)$ follows the power law predicted by Ishibashi and Orihara above a characteristic frequency $f_0 = \mu(\text{ionic})\ E_{sc}/w = 10^{-9}\ cm^2/V \cdot s \times 10^5\ V/cm \times (10\ nm)^{-1} = 100$ Hz [here E_{sc} is the space charge field and w, the depletion width] but flattens out below 100 Hz, where the ions (and vacancies) follow the applied a.c. field. This division into two regimes may explain the sub-Hz switching data of Colla et al. in addition to our $E_c(f)$. In addition to PZT and SBT, we report $E_c(f)$ in PST films and find evidence for near-zero dimensionality (point-like nucleation rather than domain wall motion dominating switching). Third, we consider oxygen vacancy ordering; new results by Stremmer et al. show this effect strongly in LSCO, and we believe that it is typical of perovskite oxides. Finally, we point out that of the many recent models for 180-degree domain switching and for fatigue (Bratkowsky and Levanyuk; Molotskii, Rosenmann et al.; Shur et al.; Dawber and Scott; Tagantsev), only a small number (Shur et al.; Dawber and Scott) are compatible with the rejuvenation effect of "restore voltages" (Scott and Pouligny, 1988) and with the remarkable difference in fatigue for (100) and (111) PZT discovered by Bomand, Takemura, Randall, and Trolier-McKinstry.

9:00 AM *D1.2
INHERENT CONTRIBUTIONS TO THE FREQUENCY DEPENDENCE OF THE COERCIVE VOLTAGE OF FERROELECTRIC THIN FILMS. U. Boettger, O. Lohse, M. Grossmann and Rainer Waser, RWTH Aachen University, Institute of Materials in Electrical Engineering, Aachen, GERMANY.

The origin of the frequency dependence of the coercitive voltage of ferroelectric thin films reveals distinct differences to the behaviour of bulk crystals and ceramics. For high-quality films, the dependence can be quite low. The requirements on the measuring technique which allow to extract these dependencies and avoid the inference by artifacts will be discussed. Based on adequate experimental data, the impact of the Curie-von Schweidler currents and the shape of the hysteresis curve on the frequency dependence will be presented.

9:30 AM D1.3
FREQUENCY-DEPENDENCE OF COERCIVE FIELDS IN FERROELECTRIC FILMS. P. Chandra, M.M.J. Treacy, NEC Research Institute, Princeton NJ; P.B. Littlewood, Cavendish Laboratory, Cambridge University, Cambridge, UNITED KINGDOM; J.F. Scott, Earth Sciences Department, Cambridge University, Cambridge, UNITED KINGDOM.

Coercive fields in ferroelectric films are typically measured at line frequencies of 60 Hz. Because ferroelectric memory devices are expected to operate at 300 MHz, it is crucial to understand the frequency-dependence of the required switching fields. We present a phenomenological approach to this problem that incorporates both inhomogeneous nucleation and domain wall motion, with detailed comparison to experiment.

9:45 AM D1.4
EVOLUTION OF THE DOMAIN STRUCTURE IN CONGRUENT AND STOICHIOMETRIC LITHIUM NIOBATE. Vladimir Ya. Shur, Evgenii L. Rumyantsev, Ekaterina V. Nikolaeva, Eugene I. Shishkin, Ural State Univ., Inst. of Phys. & Appl. Math., Ekaterinburg, RUSSIA; Robert G. Batchko, Martin M. Fejer, Robert L. Byer, Stanford Univ., E.L. Ginzton Laboratory, Stanford, CA.

We present the detail investigation of domain kinetics in single crystals of congruent (CLN) and stoichiometric (SLN) lithium niobate during polarization reversal using continuous liquid electrolyte coating and lithographically defined metal periodical electrodes. The static nano-domain patterns have been visualized by scanning probe microscope (SPM). The characteristics of the domain evolution during switching and spontaneous backswitching were extracted from the set of in situ recorded instantaneous domain patterns and compared with the switching current data. It was shown that the coercive field in SLN is the order of magnitude lower than in CLN. We have measured the voltage dependence of the sideways wall motion velocity and found out considerable difference in the activation and bias fields between CLN and SLN. The statistical analysis of the static domain patterns at the polar surfaces and tilted cross-sections has been used for investigation of needle-like domain arising (nucleation) and forward growth (tip propagation). The domain merging and spreading out of electroded area have been attributed to domain-domain electrostatic interaction. The effects of self-organized field-induced nano-domain patterning during backswitching and formation of the charged domain walls (CDW) have been studied in CLN by in situ visualization. The domain expanded in electric field by growth of CDW area and propagation of neutral wall in sideways directions. The CDW relief essentially depends on the applied field and wall motion velocity. Sideways wall motion velocity appears to be spatially uniform ranged from 40 to 80 microns per second. The dielectric permittivity of the samples with CDWs demonstrates essential frequency dependence typical for relaxor ferroelectrics. The research was made possible in part by Programs "Basic Research in Russian Universities" (Grant No.5563) and "Priority Research in High School. Electronics" (Grant No. 03-03-29), by Grant No. 01-02-17443 of RFBR, by Award No. REC-005 of CRDF.

10:30 AM D1.5
THE NATURE OF SWITCHING DYNAMICS IN ULTRA-THIN FERROELECTRIC FILMS OF P(VDF-TRFE) COPOLYMERS. A.V. Sorokin, T.J. Reece, Stephen Ducharme, Department of Physics and Astronomy and Center for Materials Research and Analysis, University of Nebraska, Lincoln, NE; V.M. Fridkin, Institute of Crystallography, Russian Academy of Sciences, Moscow, RUSSIA.

Polarization switching in ferroelectric materials has invariably been controlled by extrinsic processes, nucleation and domain wall motion. Recent studies of ultra-thin ferroelectric films of the copolymer vinylidene fluoride (70%) with trifluoroethylene (30%), made by Langmuir-Blodgett deposition, have demonstrated that sufficiently thin films switch at the theoretical intrinsic coercive field [1]. Measurements of the switching dynamics revealed the remarkable result that films with high intrinsic coercive field switch much slower (about 10 s) than samples with lower extrinsic coercive fields (about 1 μs) [2]. Theoretical analysis of the dynamics of polarization reversal from the Lagrange equation shows that fluctuations lead to pronounced viscous drag on the evolution of polarization during intrinsic switching, dominating switching dynamics in ultra-thin films when extrinsic switching is suppressed. The distinction between extrinsic and intrinsic switching is apparent in the dependence of the switching time on temperature, field, and sample thickness.
This work is supported by National Science Foundation and by the Nebraska Research Initiative.
[1] S. Ducharme, V.M. Fridkin, A.V. Bune, S.P. Palto, L.M. Blinov, N.N. Petukhova, and S.G. Yudin, Phys. Rev. Lett. 84, 175-8 (2000).
[2] S. Ducharme, M. Bai, M. Poulsen, S. Adenwalla, S.P. Palto, L.M. Blinov, and V.M. Fridkin, Ferroelectrics 252, 191-9 (2001).

10:45 AM D1.6
OPTICAL MICROSCOPY STUDY OF SWITCHING IN SINGLE CRYSTAL $BaTiO_3$. K.-T. Park, M.E. Bisher, M.M.J. Treacy, NEC Research Institute Inc., Princeton, NJ.

We use optical microscopy to observe domain wall motion and fatigue under electric fields in $BaTiO_3$ single crystals. Samples were prepared by mechanical methods to about 50-100 μm thickness. Electrodes were prepared by attaching metal wires using silver paint. Voltages up to 200V were sufficient for switching and fatigue in these experiments. We have observed the evolution of domain structure with field cycling, and several interesting observations emerge. In particular, our results confirm earlier transmission electron microscopy observations that show an increase in density of interlocked 90° domains with fatigue.

11:00 AM D1.7
EVOLUTION OF NONLINEAR POLARIZATION WITH FIELD AND TIME IN PZT THICK FILMS. Dragan Damjanovic, Alexander K. Tagantsev, Jonas Dorn, Juliette Mueller, Nava Setter, Ceramics Laboratory, Materials Department, Swiss Federal Institute of Technology - EPFL, Lausanne, SWITZERLAND.

The field and time dependence of the nonlinear dielectric response was investigated in PZT thick films with "hard" characteristics. Evolution of the polarization hysteresis and of the first, second and third harmonic amplitude and the phase angle was examined. The polarization nonlinearity is interpreted in terms of domain wall contributions. A model is proposed which describes qualitatively well the transition of the film from an ordered state (well-aged films at weak fields) to a disordered state that is achieved by driving the film

with large filed amplitudes for sufficiently long times. The phase angle of the third harmonic was found to be particularly sensitive to the film state. The order-disorder transition is characterized by switching of the third harmonic phase angle from 0° to 90°. Microscopically, the ordered state is characterized by domain walls locked by dipolar defects. In this case, the energy profile of a domain wall is "V" shaped, the polarization response is nonlinear and nonhysteretic and the phase angle of the third harmonic is 0°. Gradual increase in the field amplitude leads to local disorder in the orientation of defect dipoles and creation of local minima in the energy potential. The polarization response consequently becomes hysteretic and the third harmonic phase angle deviates from 0°. When a certain critical field is reached, the third harmonic phase angle abruptly switches from roughly 0° to 90°. This phase angle is characteristic of the systems that exhibit quasi-Rayleigh-type behavior, which is due to displacement of domain walls in a medium with random distribution of pinning centers. If the film, which defect structure is randomized by the field, is allowed enough time to relax under zero field, it will slowly evolve back to the ordered state and the phase angle of the third harmonic will switch back to zero.

11:15 AM D1.8
DIELECTRIC DISPERSION IN THE MIDDLE OF POLARIZATION REVERSAL IN TGS CRYSTALS. Toshio Kikuta, Toshinari Yamazaki and Noriyuki Nakatani, Faculty of Engineering, Toyama University, Toyama, JAPAN.

Dielectric dispersion of triglycine sulfate (TGS) has been measured in the ferroelectric phase. Although it shows three different dispersions in a multi domain single crystal, a DC bias field seems to weaken these dispersions. The fact suggests that domain walls cause the dispersions as a crystal becomes a single domain by a DC bias field. We have examined the dielectric dispersion in the middle of polarization reversal to find out the cause of the dispersions. To control the polarization and to measure the dielectric constant, first, a negative DC bias field is applied to a sample for several minutes to set it a single domain state. Subsequently, a positive DC bias field is applied only for a few milli-seconds to make the polarization partially reversed. Then the dielectric constant is measured by a LCR meter immediately. When the applying time of the positive DC bias field is shorter than the time required for the polarization reversal of the sample, the dielectric constant on the process of polarization reversal can be measured successfully. Consequently, it turns out that the dispersion intensity becomes large compared with the value measured just after annealing, and we obtain the result that the dielectric constant varies with time. Furthermore, the dielectric constant of a sample that becomes a single domain by the polarization control is different from that observed under a DC bias field after annealing. This may imply that some other relaxation processes exist besides domain wall motions. Since the reasons of this large dispersion have not been clarified yet, the results of further experiments will be presented in the meeting.

11:30 AM D1.9
FIELD-INDUCED EVOLUTION OF THE DOMAIN STRUCTURE IN CONGRUENT AND STOICHIOMETRIC LITHIUM TANTALATE. Vladimir Ya. Shur, Ekaterina V. Nikolaeva, Eugene I. Shishkin, Alexander P. Chernykh, Ural State Univ., Inst. of Phys. & Appl. Math., Ekaterinburg, RUSSIA; Kayuza Terabe, Kenji Kitamura, National Inst. for Materials Science, Tsukuba, JAPAN.

Switching kinetics has been investigated in congruent (CLT) and stoichiometric (SLT) lithium tantalate. The ingenious investigation allows us to reveal the new mechanism of fast domain kinetics in CLT driven by domain merging. The model of jerky domain wall motion in SLT has been proposed. The measurements were performed in commercial single crystalline CLT and SLT grown by double crucible Czochralski technique. Liquid electrolyte 1-mm-diameter electrodes allow us to TV record the domain evolution in the whole switching area visualized by polarizing microscope with subsequent image processing. The unique possibility to establish the correspondence between switching current data and domain kinetics was realized. Analysis of the instantaneous patterns allows us to distinguish mechanisms of domain evolution in CLT. Switching from the single domain state starts with appearance of small domains with the density up to 1000-per-mm-square and subsequent growth with velocity about one micron per second. The prevailing switching process is a fast anisotropic wall motion defined by step generation by merging of moving walls with isolated triangular domains and rapid growth of arisen steps along the wall. A super-mobile domain walls with the highest step concentration form after large domains merging. The hexagonal domains are obtained in SLT in contrast with triangular ones in CLT. The jerky motion of the strictly oriented domain walls prevails in SLT. We have proposed that such wall motion is due to interaction with pinning centers taking into account reduction of the local field at the wall by residual depolarization field produced by the partially compensated bound charges behind the moving wall. The

research was made possible in part by Programs "Basic Research in Russian Universities" (Grant No.5563) and "Priority Research in High School. Electronics" (Grant No. 03-03-29), by Grant No. 01-02-17443 of RFBR, by Award No. REC-005 of CRDF.

11:45 AM D1.10
TEMPERATURE DEPENDENCE OF POLARIZATION REVERSAL IN $SrBi_2Ta_2O_9$ FERROELECTRIC THIN FILMS. Pingxiong Yang, David L. Carroll, Dept. of Physics, Clemson Univ., Clemson, SC.

The temperature dependence of polarization reversal for $SrBi_2Ta_2O_9$ thin films was studied in a range from 10 to 300 K. Below temperatures of 200 K, no hysteresis loop or nonlinear dielectric peak was exhibited. The polarization fatigue at these temperatures was significant due to the temperature dependence of polarization reversal in the films. We propose pinning of the domain walls as a mechanism of this effect at low temperatures.

SESSION D2: COMPUTATIONAL APPROACHES
Chair: Simon R. Phillpot
Tuesday Afternoon, November 27, 2001
Room 205 (Hynes)

2:00 PM *D2.1
EFFECT OF LONG-RANGE POLARIZATION INTERACTION AND STRAIN ON MICROSTRUCTURE EVOLUTION. A. Saxena, T. Lookman, R. Ahluwalia, Los Alamos National Laboratory, Los Alamos, NM.

Considering ferroelectrics as improper ferroelastic materials we treat symmetry allowed couplings between polarization vector and strain tensor. Elastic compatibility constraint between strain components leads to an anisotropic long-range interaction between the square of the polarization amplitude at different spatial locations in the crystal. Within the time-dependent Ginzburg-Landau framework we study the microstructure evolution during the phase transition and as a function of applied stress or electric field for representative materials. We then generalize these concepts to multiferroic materials where other functionalities such as atomic shuffle and magnetization may couple to polarization through strain.

2:30 PM D2.2
FERROELECTRIC DOMAIN WALLS IN $PbTiO_3$. Bernd Meyer, David Vanderbilt, Rutgers University, Department of Physics and Astronomy, Piscataway, NJ.

A thorough understanding of ferroelectric switching requires a microscopic description of the underlying domain walls and their dynamics. We have carried out ab-initio calculations of 90° and 180° ferroelectric domain walls in $PbTiO_3$. We find that it is possible to construct metastable supercell structures representing both kinds of domain walls using supercells of only modest size. The relaxed structure, energy, and polarization profile are calculated for each case. Both kinds of domain wall are found to be atomically sharp. Two features of the 90° domain wall are surprising: the barrier for its motion is found to be miniscule, and a small offset of the electrostatic potential across the domain wall is found.

2:45 PM D2.3
SOFTENING THE POLARIZATION ROTATION: PRESSURE EFFECTS. Marco Fornari, Center for Computational Materials Science, Naval Research Laboratory, Washington, DC; David J. Singh, Center for Computational Materials Science, Naval Research Laboratory, Washington, DC.

We analyze whether the pressure effects can soften the polarization rotation by exploring the total energy landscape of various perovskite oxides. We performed general potential LAPW calculations to relax the internal degrees of freedom compatible with [111], [110], and [100] polarizations and considered the dependence of the energy barriers upon volume variation and chemical substitutions. Uniaxial stress is also imposed in order to interpret recent experimental results and to suggest directions to optimize the electromechanical properties. Work supported by ONR.

3:30 PM *D2.4
DYNAMICS OF POLARIZATION REVERSAL AND HOMOGENEOUS DOMAIN FORMATION IN PEROVSKITE FERROELECTRICS 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, Materials Science Division, Argonne National Laboratory, Argonne IL; M.G. Stachiotti and R.L. Migoni, Instituto de Fisica Rosario, CONICET-UNR, Rosario, ARGENTINA.

We use molecular-dynamics simulations to explore the atomic-level details of the dynamics of polarization reversal in the tetragonal phase of a monodomain order-disorder perovskite ferroelectric. We find that the transition from an [001] orientation to an [00$\bar{1}$] orientation involves polarization rotation through intermediate states with polarization parallel to [011] and [01$\bar{1}$]. We elucidate the strong interplay of polarization and lattice strain during polarization reversal. Finally, we examine in some detail the nucleation and growth of domains of reversed polarization and estimate the critical nucleus size required for domain reversal. This work was supported by the U.S. Department of Energy, Office of Science under Contact W-31-109-Eng-38, by the DOE S&P Center Project on Nanoscale Phenomena in Perovskite Thin Films, and by CONICET-Argentina.

4:00 PM D2.5
POLARIZATION DYNAMICS IN PHENOMENOLOGICAL THEORY OF RELAXORS. Rinat F. Mamin, Kazan Physical-Technical Institute of RAS, Kazan, RUSSIA.

The dynamic behaviour of relaxor ferroelectrics is described theoretically in the framework of the phenomenological model with "soft localized polarons". It is shown that the process of charge carrier localization on impurity centers is of importance in the relaxor theory. It is known that a relaxor behaviour is connected with disorder in the position of atoms. We believe, that only disorder connected with charge distribution is important, and that the phase transition must be in the temperature range where the concentration of localized charges has strong temperature dependence. Then the charge carriers localize on the defects in the vicinity of the phase transition, the deformation occurs around them, and in this sense the polarons arise. And because the probability of delocalization of this polarons is considerable these formations can be called the soft localized polarons. The temperature behavior of dielectric permittivity is determined by the dynamics of charge carrier localization. A characteristic vibrational frequency of these formations and a dispersion of dielectric permittivity are determined by characteristic times both of the lattice and electron subsystems. The probability distribution of relaxation times, $g(\ln(\tau))d\ln(\tau)$, is obtained analytically. It is found that 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. We obtaine a quantitative agreement for the temperature and frequency behavior of the dielectric permittivity. The polarization dynamics of relaxor ferroelectrics in the low-temperature phase are also discussed. The delay time of phase transition at field after cooling at zero field is described. Passage of the transition to the phase with long-range ferroelectric order after application of electric field is determined by local center ionization and consequent re-orientation of the polarization in microclusters. All of these questions are clarified basing on simple, preferably thermodynamic, considerations.

4:15 PM D2.6
RANDOM FIELD HYDRODYNAMIC MODEL OF A LARGE DIELECTRIC RESPONSE IN RELAXORS. Serguei Prosandeev, Rostov State Univ, Rostov on Don, RUSSIAN FEDERATION.

A Random Field Hydrodynamic (RFH) model is proposed in order to explain high values of the dielectric permittivity in relaxors. We assume the existence of polar regions but our model differs from the superparaelectric model appreciably. We consider the energy to be degenerated with respect to the direction of the polarization in the polar regions (the main assumption of the hydrodynamic model). As a result the dielectric susceptibility proves to be divergent while internal random fields (or/and stresses) stabilize the system. We show that the transversal hydrodynamic fluctuations of the local polarization lead to a large contribution to the dielectric susceptibility that can explain the large values of the dielectric susceptibility in relaxors.

4:30 PM D2.7
DIELECTRIC PROPERTIES IN THE $CaTiO_3$-$CaAl_{1/2}Nb_{1/2}O_3$ SYSTEM FROM FIRST PRINCIPLES. Eric Cockayne, Benjamin P. Burton, NIST, Ceramics Division, Gaithersburg, MD.

Fundamental studies of the microscopic origins of structure-property relations in dielectrics are essential for understanding the contrasting properties of materials with similar structures. $CaTiO_3$ (CT) and $CaAl_{1/2}Nb_{1/2}O_3$ (CAN) crystallize in very similar perovskite-related structures, but their dielectric permittivities are 170 and 27, respectively. CT-CAN solid solutions are of interest for microwave applications because a composition exists where $\tau_f = 0$. We use first-principles computations to determine interatomic forces as functions of cation configurations, using cluster expansion techniques. From the resulting model, we compute the phonon and dielectric properties in CT-CAN as functions of composition and cation ordering, and compare theoretical trends with experiment.

4:45 PM D2.8
POLARIZATION ROTATION-BASED SWITCHING MECHANISM OF A FERROELECTRIC LATTICE NEAR THE MORPHOTROPIC PHASE BOUNDARY. Dan Ricinschi, Yoshihiro Ishibashi[a], Makoto Iwata[b], Minoru Noda and Masanori Okuyama, Osaka Univ, Graduate School of Engineering Science. [a]Aichi Shukutoku Univ.; [b]Nagoya Inst of Technology, JAPAN.

The study of superior physical properties near the morphotropic phase boundary (MPB) of ferroelectric perovskites has been an interesting research topic for quite a long time. Recently, new information regarding the existence of a previously undetected monoclinic phase has been the focus of both experimental and theoretical work. The monoclinic distortion has been related to the unique piezoelectric properties close to the MPB. Additional investigations have been concerned with the phase symmetry changes induced by electric field, leading to engineered domain configurations. In this paper we have analyzed a novel ferroelectric switching mechanism based on a three-dimensional polarization vector kinetics, in a lattice model based on the Landau-Devonshire free energy functional. The Landau coefficients control the equilibrium phase symmetry and the MPB corresponds to the case when the free energy becomes isotropic in the space spanned by the polarization vector components. Starting from the up-polarization state of a lattice chosen in the tetragonal phase, an electric field oriented downwards will gradually induce switching to the opposite polarization state. Nucleation seeds with various densities have been randomly generated. The switching threshold has been found to decrease in the vicinity of the MPB, due to the contribution of polarization vector rotations. This leads to the formation of transient reduced-symmetry phases and a large switching current. Alternatively, in a lattice with an increased density of nucleation seeds, the polarization reversal can be induced by even lower electric fields. However, while the nuclei decrease the switching time, they inhibit the polarization rotations and cause direct switching by 180° domain walls, thus preserving the natural tetragonal phase. By drawing vector representations of the ferroelectric lattice at various switching stages, with the distance from the MPB, the electric field and the nucleation seed density as parameters, one can extract useful information of the polarization evolution at local level.

SESSION D3: NOVEL CHARACTERIZATION TECHNIQUES
Chair: Carol Thompson
Wednesday Morning, November 28, 2001
Room 205 (Hynes)

8:30 AM *D3.1
MAGNETODYNAMIC EXCITATIONS IN PATTERNED FERROMAGNETIC THIN FILM ENSEMBLES. Thomas Crawford.

ABSTRACT NOT AVAILABLE

9:00 AM *D3.2
CONFOCAL SCANNING OPTICAL MICROSCOPY OF FERROELECTRIC MATERIALS. Jeremy Levy, Dept of Physics and Astronomy, University of Pittsburgh, Pittsburgh, PA.

Ferroelectric materials have rich and interesting behavior that is derived from the complex relationship between polar structure and dielectric response. In disordered or "mesoscopic" ferroelectrics, the ferroelectric polarization can have large local variations in both magnitude and direction. The measured dielectric response of so-called "ferroelectric nanodomains" is a strong function of electric field, frequency and nanodomain size, and often exhibits resonances in the microwave regime. We have developed a number of optical probes to explore both the spatial and temporal structure of the ferroelectric response. Key results presented will include: (1) near-field optical probes ferroelectric domain structure in $BaSrTiO_3$ thin films, (2) diffraction-limited measurements (500 nm resolution) which probe mesoscopic microwave dispersion in ferroelectric thin films, and (3) temperature-dependent measurements which probe local phase transitions in these films. This work is supported by DARPA/ONR N00173-98-1-G011.

9:30 AM D3.3
TIME-RESOLVED X-RAY DIFFRACTION STUDIES OF POLARIZATION SWITCHING IN FERROELECTRIC AND RELAXOR EPITAXIAL THIN FILMS. Carol Thompson, Northern Illinois University, Dept. of Physics, DeKalb, IL and Materials Science Division, Argonne National Laboratory, Argonne, IL; M.E.M. Aanerud, Northern Illinois University, Dept. of Physics, DeKalb, IL; S.K. Streiffer, G.B. Stephenson, G.-R. Bai, Materials Science Division, Argonne National Laboratory, Argonne, IL; A. McPherson, W.-K. Lee, UPD, Argonne National Laboratory, Argonne, IL.

Domain dynamics at high switching frequencies have been measured using time-resolved synchrotron x-ray diffraction to track structural signatures of the polarization reorientation in ferroelectric and relaxor epitaxial thin films. We present experimental results of the lattice response transients for a step in electric field, with 17 ns time resolution for the x-ray technique. In particular, the structural measurements can directly distinguish between non-switching and switching components of the polarization response. This complements high-speed electrical measurements which require comparisons across a pulse train (PUND) to determine the switched response. The time-dependence of polarization reorientation will be described for PMN-PT thin films.

9:45 AM D3.4
IN-SITU X-RAY SYNCHROTRON PROBING OF DOMAIN WALL STRAINS AND DYNAMICS IN FERROELECTRICS. Sungwon Kim, Venkatraman Gopalan, Pennsylvania State University, Dept. of Materials Science and Engineering, University Park, PA; Terence Jach, Bruce Steiner, National Institute for Standards and Technology, Gaithersburg, MD; Steven Durbin, Purdue University, W. Lafayette, IN.

We report in-situ imaging of strains and strain-evolution at ferroelectric domain walls in lithium niobate observed under highly a collimated X-ray synchrotron beam with a strain resolution of a part per million. The domain walls in lithium niobate have surprisingly wide regions of strains around them (under zero external field), extending over 50-100 microns. Though surprising from a theoretical viewpoint, which predicts domain wall widths on the order of many lattice units, these strains are shown to arise from defect-mediation through the presence of a small amount of lithium non-stoichiometry in the crystals. In addition, real-time movies of X-ray images under an external field reveal that the piezoelectric strain fields at domain walls extend and interact over hundreds of microns even at fields well below the coercive field for any domain motion. These strains also give rise to focusing and defocusing of X-rays due to lens-like distortion of domains. We present theoretical results based on finite element strain analysis coupled with dynamical theory of diffraction to explain these strain and optical effects.

10:30 AM D3.5
SCANNING NONLINEAR DIELECTRIC MICROSCOPY STUDY ON PERIODICALLY POLED $LiNbO_3$ FOR A HIGH PERFORMANCE QPM DEVICES. Yasuo Cho, Koya Ohara, Satoshi Kazuta and Hiromasa Ito, Tohoku Univ, Research Institute of Electrical Communication, Sendai, JAPAN.

Recently, periodically poled $LiNbO_3$ (PPLN) has been developed as quasi-phase matching (QPM) devices for optical parametric oscillation (OPO) for application to wavelength conversion. It is important to evaluate domain distribution of PPLN accurately for improvement of polling technique aiming for higher efficiency. By observation of the domain distribution of PPLN using scanning nonlinear dielectric microscopy (SNDM), we have succeeded to find very important facts for obtaining QPM devices with very high performance. First, we found that a single domain surface layer formed on PPLN. From this result, we found it is very effective to remove the single domain surface layer of PPLN to obtain high performance optical surface wave device. Next, SNDM studies also reveals that the very strong residual stress remains in PPLN fabricated by the high voltage applying method due to pinning effect in $LiNbO_3$ single crystal. This stress heavily reduces the nonlinear dielectric constant of $LiNbO_3$. By releasing this stress using annealing method, we confirmed the drastic recovery of the reduced nonlinear dielectric constant. And in optical region, this constant is equivalent to SHG constant, therefore, we propose that annealing will be very effective to obtain a high performance QPM device with higher efficiency.

10:45 AM D3.6
DYNAMIC HETERODYNED POLARIZATION IMAGING: A SCANNING PROBE TECHNIQUE FOR STUDYING POLARIZATION DYNAMICS IN FERROIC MATERIALS. D.R. Oliver, A. Pu, D.J. Thomson and G.E. Bridges, Electrical and Computer Engineering, University of Manitoba, Winnipeg, Manitoba, CANADA.

Electric fields in dielectric materials produce dipoles related to the polarizability of the material. In this paper we will present a technique that measures the polarization near a surface. This polarization may or may not be induced by a modulated signal applied to the conducting probe used for detection. The polarization dipoles in the surface layers of the material generate an electrostatic attraction between the between the probe and the dielectric material. Using techniques common in non-contact force microscopy these forces can easily be sensed. Remarkably, this measurement technique can be extended to frequencies well above the mechanical resonant frequency of the probe cantilever by utilizing amplitude modulation heterodyning. By rastering the probe over the surface an image of the dielectric properties of the surface can be produced. We expect this technique to be useable up to frequencies of at least 20 GHz and time resolution of less than 100 ps. We present calculations of the forces generated assuming simple probe geometries and also thermal noise that compare favourably with experimental results. The static detection of well-characterized mechanical and electrical resonances at a frequency significantly greater than the probe cantilever resonance is described, as well as a "frozen" image of an operating 434 MHz surface acoustic wave device. The experiments already completed demonstrate that this technique may be employed to produce images that display the local polarizability of materials at a given frequency. In more detailed studies, regions of interest can be imaged repeatedly, with different frequencies used to produce each image. Such investigations have the potential to make an important contribution to the studies of dynamic processes in ferroic systems.

11:00 AM D3.7
DOMAIN ELECTROLUMINESCENCE IN FERROELECTRIC CRYSTALS. Nataliya N. Krainik, Ioffe A.F. Physico Technical Institute RAS, St. Petersburg, RUSSIA; Svetlana A. Flerova, Dniepropetrovsk State Univ, Dept of Physics, Dniepropetrovsk, UKRAINE; Svetlana A. Sushko, Vadim V. Fomichov, Dniepropetrovsk, National Mining University of Ukraine, Dniepropetrovsk, UKRAINE.

Electroluminescence is a sensitive investigation method of the processes of the change of polarization in ferroelectric media. The time dependence of electroluminescence intensity characterizes the progress of the processes of the change of polarization inducing the electroluminescence. The connection of the processes of change of the domain structure and the luminescence appearing at switching of polarization is discussed in the paper for a number of broadband ferroelectric crystals: BaTiO3 (pure and with a large set of alloying additives), Bi4Ti3O12, KNO3, NaNO2, Gd2(MoO4)3 (pure and with additives Nd), three-glycerol sulfate, K2ZnCl4 (pure and with additives Rb, Cu, Mn, Co), Pb5Ge3O11, Sn2P2S6, Li2Gd4Mo7O28. The correlation of parameters of luminescence with the kinematics of switching is observed for all the crystals. At different stages of switching the excitation of the electronic subsystem and the creation of inverse population is conditioned by different reasons: the vacation of charges of screening with the displacement of the screened domain wall, the birth of non-equilibrium pairs by strong local electric fields, appearing with lateral moving of domains' boundaries as a result of their interaction with the defects of crystal lattice at the collapse of the domains boundaries at the closing stage of installation of homogeneous polarization in the whole crystal; at the dipole relaxation of polarization at uniform switching in definite volume of a crystal. A singularity of domain electroluminescence consists in a deference of emission spectrum on the switching regime. The definite conditions of depolarization a narrowing of the emission spectrum is observed and the transition to the regimes of generation. The study of polarization diagrams and directional diagrams has shown that the luminescence of a ferroelectric crystal can considered as that of an aggregate of switching domains which are directed by an external action. This work was supported by the Russian Fund for Fundamental Research, Grant N 01-02-17801.

11:15 AM D3.8
ELECTRICAL PROPERTIES OF FERROELECTRIC OXIDE NANOSTRUCTURES. M. Alexe, C. Harnagea, A. Visinoiu, and D. Hesse, Max Planck Institute of Microstructure Physics, Halle, GERMANY.

We have simultaneously addressed both issues of patterning ferroelectric crystalline oxide structures and characterizing the obtained structures. Firstly, we have create nanometer structures using both "top-down" approach based on electron-beam lithography, and a "bottom-up" approach based on self-assembly methods. The "top-down" approach is an electron beam direct writing method and allows fabrication of ferroelectric $Pb(Zr,Ti)O_3$ (PZT) and $SrBi_2Ta_2O_9$ (SBT) nanosize structures with lateral sizes under 100 nm. Alternatively, nanosize ferroelectric structures have been prepared using "bottom-up" approaches employing self-assembly fabrication methods based on chemical solution deposition (CSD) and pulsed laser deposition (PLD). In this way PZT and $BaTiO_3$ structures having lateral sizes from 20 nm to 60 nm were prepared on $SrTiO_3$:Nb substrates. Secondly, the ferroelectric, piezoelectric and switching properties were studied using scanning probe microscope (SPM) working in piezoresponse mode. Piezoresponse scanning force microscopy measurements performed on lead zirconate titanate (PZT) mesoscopic structures revealed a negative shift of the initial piezoelectric hysteresis loop. The shift is dependent on the size of the structure and is most probably due to the pinning of ferroelectric domains at the free lateral surface and the ferroelectric-electrode interface. Considering a simple model, the thickness of the pinned domain layers is found to be of about 15 nm and 70 nm at the

ferroelectric-electrode interface and the lateral free surface, respectively. The same SPM setup with conductive tips was used to measure leakage current in nanosize structures. Electronic transport mechanism in some nanosize structures was found to be of space-charge-limited (SCL) type, i.e. $J(V) = aV^2$, unlike in the case of large-area devices of the same material which the transport mechanism is Schottky-dominated.

11:30 AM D3.9
SECOND-HARMONIC GENERATION FROM FERROELECTRIC DOMAINS IN $Sr_{0.75}Ba_{0.25}Nd_2O_6$ SINGLE CRYSTALS. Tatyana R. Volk, Institute of Crystallography RAS, Moscow, RUSSIA; Nataliya E. Sherstuk, Elena D. Mishina, Moscow Institute of Radioengineering, Electronics and Automation, Moscow, RUSSIA; Theo Rasing, Research Institute for Materials, University of Nijmegen, Nijmegen, THE NETHERLANDS.

Strontium-barium niobate Sr_xBa_{1-x} Nd_2O_6 (SBN-x) ferroelectric crystals are used for a frequency-conversion arising from random needle-like domains. Quasi-phase-matching can be obtained on both poled and unpoled crystals that leads to a sufficient self-frequency doubling. Multi-color diffused radiation can be obtained by self-sum-frequency mixing. The more the spread of domain sizes, the more the variety of colors is expected within radiation converted on random domains. The majority of present publication is devoted to the SBN-x crystals in a middle of a stable concentration range (x=0.6). In the same time, SBN-0.75 (that is on the edge of stability range) provides a more spread spectrum of domain sizes and higher domain mobility under applied external fields. This is due to a most diffuse phase transition and a lowest phase transition temperature. In this paper we studied an efficiency and spatial distribution of second-harmonic generation (SHG) intensity in poled SBN-75 crystal using an output of Ti:Sapphire laser at 700 nm. During the poling of the crystal up to 1 kV/cm the SHG efficiency increases 3 times. Scattering angle of SHG intensity is anisotropic in respect to a poling direction. Increasing the poling field leads to a decrease of scattering angle in a direction parallel to the poling field form 4° down to 2°, while in a direction perpendicular to the poling field a scattering angle is about 10° and does not depend on the poling field. Residual poled-enhanced SHG radiation was observed within several hours after poling. We compare domain structure of the crystal before and after poling and observed a time-dependent reorganization of needle-like domains. Studies in optical-frequency conversion on domains in SBN in addition to practical benefits may provide knowledge on polarization processes in relaxors, for example, on the formation of aligned polar clusters in the phase-transition temperature range.

11:45 AM D3.10
PREPARATION OF $(PLZT)_x(BiFeO_3)_{1-x}$ FERROELECTRIC-FERROMAGNET AND THEIR NON-LINEAR FARADAY EFFECT. Toshimitsu Kanai, Shin-ichi Ohkoshi, Kazuhito Hashimoto, Univ of Tokyo, RCAST, Tokyo, JAPAN.

We have prepared $(PLZT)_x(BiFeO_3)_{1-x}$ solid solutions by solid-state reaction. The sintered bodies for x = 0.10 - 0.45 showed both magnetic hysteresis loops and ferroelectric hysteresis loops at room temperature. The observed spontaneous magnetization is due to the weak ferromagnetism resulting from the distortion of the antipararell spins of Fe^{3+} ions through the structural distortion by the incorporation of PLZT. In order to study optical properties of the ferroelectric-ferromagnet, $(PLZT)_x(BiFeO_3)_{1-x}$ thin films were prepared by sol-gel method. The film for x = 0.1 showed Faraday effect and second harmonic generation (SHG). Moreover, non-linear Faraday effect was observed, that is, the optical rotation angle and intensity of SH light were controlled by the external magnetic field. The rotation angle was fairly larger than that of linear one. This material may allow us to develop novel types of optical devices based on the interaction between magnetic and electric properties.

SESSION D4: DOMAIN-RELATED PHENOMENA
Chair: William Jo
Wednesday Afternoon, November 28, 2001
Room 205 (Hynes)

2:00 PM D4.1
INTRINSIC CONTRIBUTIONS TO FATIGUE IN POLYAXIAL FERROELECTRICS. A. Krishnan, M.M.J. Treacy, M.E. Bisher, P. Chandra, NEC Research Institute, Princeton, NJ; P.B. Littlewood, Cavendish Laboratory, Cambridge, UNITED KINGDOM.

We present in-situ transmission electron microscopy observations of domain wall motion in freestanding $KNbO_3$ single-crystals. Tilted and curved 90° domain walls are common and are readily moved by electric fields, whereas untilted domain walls resist such motion. We explain these results by noting that a tilted domain wall has a polarization charge and an associated depolarization field reduces the local switching barrier, thereby enhancing the mobility of these wall regions under an applied electric field. The switching efficiency of a domain is then determined by the relative electrostatic energies of its dominant neighbors. Consequently, some domains are inhibited from switching due to their immediate environment, a phenomenon we call domain interlocking. Any increase in density of such field-resistant configurations with cycle time represents an inherent contribution to fatigue. Uniaxial ferroelectrics should not support such interlocked domains.

2:15 PM D4.2
POLARIZATION RELAXATION KINETICS AND 180° DOMAIN WALL DYNAMICS IN FERROELECTRIC THIN FILMS. C.S. Ganpule, V. Nagarajan, A. Roytburd, R. Ramesh, University of Maryland, Dept of Materials Engineering, College Park, MD; Ellen Williams, University of Maryland, MRSEC and Department of Physics, College Park, MD; J.F. Scott, Symetrix Center for Ferroics, Earth Sciences Department, Cambridge University, Cambridge, UNITED KINGDOM.

The time dependent relaxation of remanant polarization in epitaxial lead zirconate titanate ($Pb(Zr_{0.2}Ti_{0.8}O_3$, PZT) ferroelectric thin films, containing a uniform 2-dimensional grid of 90° domains (c-axis in the plane of the film), is examined using voltage modulated scanning force microscopy (SFM). 90° domain walls preferentially nucleate 180° reverse domains during relaxation, which grow and coalesce as a function of relaxation time. Relaxation is seen to saturate at different levels depending on the write-voltage. Late (saturation) stages of relaxation are accompanied by pinning and faceting of the domain walls (drastically reducing the wall mobility), which is a direct evidence of the role of defect sites and crystallographic features on polarization relaxation. The kinetics of relaxation is modeled through the nucleation and growth theory of Johnson-Mehl-Avrami-Kolmogorov with a decreasing driving force. This work is supported by NSF-MRSEC foundation.

2:30 PM D4.3
FERROELECTRIC POLARIZATION SWITCHING AS SEEN FROM PIEZORESPONSE-AFM MEASUREMENTS. C. Harnagea, A. Pignolet, M. Alexe, H.N. Lee, and D. Hesse, Max-Planck-Institut fuer Mikrostrukturphysik, Halle(Saale), GERMANY.

Voltage-modulated scanning force microscopy in contact mode, or piezoresponse scanning force microscopy, is now a widely recognized technique for imaging ferroelectric domains in ferroelectric thin films. The quantities measured, however, are the amplitude and phase of a locally induced periodic piezoelectric strain, and not the ferroelectric polarization itself. The domain structure visualized corresponds to those ferroelectric domains whose polarization has a component normal to the film surface, but the amplitude of the signal is actually not proportional to the magnitude of the normal component of the polarization. Extracting this information requires a more detailed consideration of the imaging process taking the tensorial nature of the piezoelectric response into account. Likewise, the shear mode of scanning force microscopy allows to image domains having a component of polarization in the plane of the film. By applying a suitable DC bias superimposed over the AC testing voltage, a reorientation of the ferroelectric polarization under the SFM tip is achieved. A local piezoelectric hysteresis loop can be recorded if the DC bias applied to the SFM tip placed above the region investigated is cycled. The shape of the piezoelectric hysteresis can be used to extract information about the ferroelectric switching induced. Analyzing the local hysteresis loops recorded from various grains in different ferroelectric films, several types of polarization reorientation were found to take place. While in epitaxial $SrBi_2Ta_2O_9$ films with different crystallographic orientations, only 180° switching was detected, both 180° - and 90° - switching were found on tetragonal lead zirconate titanate (PZT) films. An abnormal hysteresis shape was observed in $Bi_4Ti_3O_{12}$ films, implying that another type of switching takes place. This unusual behavior is explained by taking into consideration the particular geometry of the experiment and the complex domain states in bismuth titanate.

2:45 PM D4.4
DOMAIN BACKSWITCHING IN FERROELECTRICS: REAL-TIME STUDIES AND PHENOMENOLOGICAL MODELING. Sungwon Kim, Venkatraman Gopalan, Pennsylvania State University, Dept. of Materials Science and Engineering, University Park, PA.

We present direct real-time studies of domain backswitching phenomenon in ferroelectric lithium niobate and tantalate. A phenomenological framework is proposed within which the interrelations between dipolar point defects due to lithium deficiency in these crystals and many phenomena such as backswitching, domain stabilization, polarization cycling effects, large changes in coercive fields with composition, and "negative" coercive fields are elucidated.

3:30 PM D4.5
DYNAMICS OF SPACE CHARGES AND POLARIZATION IN FERROELECTRIC THIN FILMS PROBED BY ELECTROSTATIC FORCE MICROSCOPY. William Jo, Theodore H. Geballe Laboratory for Advanced Materials, Stanford University, Stanford, CA.

We report results on charge dynamics in $Pb(Zr,Ti)O_3$ thin films by electrostatic force microscopy with metal-coated cantilevers. Effects of bottom electrodes and crystal orientation on polarization dynamics have been explored using Pt, RuO_2, and $LaNiO_3$ bottom electrodes. Charge retention can be controlled by a reverse-poling scheme via changing polarization of ferroelectric films with positive and negative bias sequentially. The surface charge density of the films was observed as a function of time in a selected area where a region is single-poled and another region is reverse-poled. Retention behaviors of the regions are very different; the single-poled region shows a declined response and the reverse-poled region reveals a retained characteristic. Logarithmic and extended-exponential decay mechanisms of polarization are explained by space charge redistribution and trapping of defects in the films. In addition, we observed a direct correlation between domain configurations and microstructural outgrown features in the as-deposited films. By local poling of the ferroelectric surfaces, it is found that grain growth with uneven polarization direction gives rise to asymmetric switching behavior (imprint) in thin-film ferroelectric capacitors.

3:45 PM D4.6
REDUCTION OF IMPRINT AND RECOVERY OF THE VIRGIN POLARIZATION STATE IN FERROELECTRIC CAPACITORS USING A SIMPLE DEPOLING TECHNIQUE. M.J. Higgins, NEC Research Institute, Princeton, NJ; A. Krishnan, Lucent Technologies, Breinigsville, PA; S. Bhattacharya, M.M.J. Treacy, NEC Research Institute, Princeton NJ.

A simple depoling technique is described which is applied to ferroelectric thin film PZT capacitors. We find that depoling the capacitors allows us to access the virgin (or as - grown) polarization state without heating the films above the transition temperature. In addition, we find that depoling can significantly reduce imprint effects when the imprint is caused by unipolar cycling or the application of a large dc bias.

4:00 PM D4.7
SMALL INVERTED DOMAIN ON FORMATION DOT IN STOICHIOMETRIC $LiTaO_3$ SINGLE CRYSTAL USING SCANNING NONLINEAR DIELECTRIC MICROSCOPY. Yasuo Cho, Yoshiomi Hiranaga, Satoshi Kazuta, Tohoku Univ, Research Institute of Electrical Communication, Seadai, JAPAN; Kazuya Terabe and Kenji Kitamura, National Institute for Materials Science, Nanomaterials Laboratory, Tsukuba, JAPAN.

Recently, we have developed and reported scanning nonlinear dielectric microscopy (SNDM) for imaging ferroelectric domain distribution. The resolution of SNDM is confirmed to achieve sub-nano meter order. We have studied artificial small domain inverted dots formation in the ferroelectric single crystal using SNDM to investigate domain dynamics in very small area quantitatively with good reproducibility. This time, we selected the stoichiometric $LiTaO_3$ (SLT) single crystal as a specimen, because coercive electric field of stoichiometric one is 1.7 kV/mm, which is about one thirteenth of that of congruent one. At first, as basic investigation of domain engineering, we revealed the relationship between applied voltages to SLT sample and inverted areas, and also clarified the inverted domain area as a function of voltage applying time. As a result, we succeeded to obtain nano-sized domain inverted dots in SLT single crystal. These results indicate that SLT is a promising material for a high performance electro-communication device such as a high-density memory in a next generation.

4:15 PM D4.8
LARGE STRAIN ACTUATION IN SINGLE-CRYSTAL BARIUM TITANATE UNDER COMBINED ELECTROMECHANICAL LOADING CONDITIONS. Eric Burcsu, G. Ravichandran, K. Bhattacharya, California Institute of Technology, Division of Engineering and Applied Science, Pasadena, CA.

The nonlinear electromechanical behavior of ferroelectric materials is governed by the motion of domains. Since many common ferroelectric materials, such as barium titanate and PZT, are also ferroelastic, the domain motion is highly affected by stress as well as electric field. Experiments are performed on (001) and (100) oriented single crystals of barium titanate under combined electromechanical loading conditions. The crystal is subjected to a constant compressive stress between 0 and 5 MPa, and an oscillating electric field along the [001] direction. Global deformation and polarization are measured as a function of electric field at different values of compressive stress and input frequency. The electrostrictive response is shown to be highly dependent on the level of applied stress with a maximum strain of 0.9% measured at a compressive stress of about 2 MPa. Comparisons of the experimental data with theoretical predictions are presented. In addition, the use of semi-transparent electrodes and transmitted illumination allows in situ, real-time microscopic observation using a long working-distance, polarizing microscope. The evolution of the domain pattern is observed simultaneously with the strain and polarization measurement.

4:30 PM D4.9
USE OF MECHANICAL PRE-LOADS TO ENHANCE THE PERFORMANCE OF STRESS-BIASED PIEZOELECTRIC ACTUATORS. Robert Schwartz, Manoj Narayanan, Clemson University, School of MS&E, Clemson, SC.

Stress-biased actuators are currently under investigation for a range of applications where high displacement and load-bearing capabilities are required. These devices are of interest since they offer performance characteristics (strain and stress generation) not attainable with unimorph or bimorph devices, or other flextensional actuators. Despite the exceptional performance of stress-biased actuators, devices with even greater performance are needed to pursue other applications. In this study, mechanical preloads were added to standard stress-biased devices, such as Thunder® actuators, to alter the domain configuration and to enhance the extrinsic piezoelectric response mechanism. The devices may be viewed as mechanical/piezoelectric actuators. We have investigated two different pre-load approaches for rectangular actuators: elongated springs and elastomers. Both approaches improve actuator performance characteristics and further dome the devices by pulling the ends of the device closer to each other. The free displacement response of the modified actuators was 2 to 6 times greater than the standard devices, depending upon drive frequency. The displacement response for modified devices that were subjected to a mass load was also improved. In this presentation, we discuss approaches to incorporate different types of mechanical preloads. We also review actuator mounting strategies that permit the above modifications and facilitate the use of these devices in different applications. We then discuss changes in displacement response as a function of applied voltage and frequency, which have been characterized by fiber optic and LVDT measurements for a range of mass loadings and mechanical pre-loads. Finally, results are presented for x-ray diffraction measurements of domain configuration and switching response as a function of mechanical preload. These results are compared to standard stress-biased actuators and changes in effective piezoelectric coefficients are reported.

SESSION D5: COMMON THEMES IN DYNAMICS
Chair: Ramamurthy Ramesh
Thursday Morning, November 29, 2001
Room 205 (Hynes)

8:30 AM *D5.1
DEFECT MIGRATION AND DYNAMICS OF DOMAIN SWITCHING IN FERROELASTIC MATERIALS. Ekhard Salje, University of Cambridge, Department of Earth Sciences, Cambridge, UNITED KINGDOM.

While the trajectories of ferroelastic twin patterns are generally determined by energy minimization we find that the dynamic response is often determined by defect pinning and defect migration. Twin Patterns may consist of individual twins, needle domains, combs and tweed patterns. Pinning is enhanced by fast diffusion of impurities along domain walls and a strong anchoring of the twin walls at the surface. Computer simulation using realistic interatomic potentials show wall energies and wall thicknesses to be close to experimental values. Defect migration in the same simulations are enhanced along twin walls in perovskite structures and simple cubic model systems. They are reduced in materials such as quartz where the diffusion is faster in the bulk than in the twin wall. Decorated walls can be collectively unpinned leaving the defect structure as a 'ghost' pattern behind the propagating front. Slow wall movement can carry the defects with the front. Very effective wall pinning happens at the tip of needle domains. Such needle tips then split when reaching the crystal surface.

9:15 AM *D5.2
DIVERSE BEHAVIORS IN RELAXOR FERROELECTRIC MATERIALS. L. Eric Cross, Pennsylvania State University, University Park, PA.

Relaxor ferroelectrics are dielectrics that have a high weak field dielectric permittivity maximum in some temperature region which is

massively dispersive at radio frequencies, but also at lower temperature exhibit the hysteretic behavior in high fields characteristic of a conventional ferroelectric i.e. they combine the properties of a relaxation dielectric and a ferroelectric, hence the name relaxor-ferroelectrics.
For these systems we have, over time, come to expect a range of common features such as: In zero field cooled samples the absence of the macro anisotropy associated with a ferroelectric domain structure. That the weak field dispersive character follow the empirical Vogel:Fulcher relation, suggesting a glass like freezing temperature T_F. Above T_F mean polarization $\overline{P}$ of zero, but large values of $\overline{P^2}$ persisting up to a much higher temperature T_d indicating strong polarization fluctuations. Usually large flexoelectric effects, and a remarkable number of useful morphotropic phase boundaries in relaxor based solid solutions.
In this talk, these and other features of behavior in perovskite, tungsten bronze structure and in polymer relaxor ferroelectrics will be examined to bring out the diversity of behavior within these families and to explore avenues to modify and improve the exciting properties which these systems afford.

10:30 AM *D5.3
STRUCTURAL FLUCTUATIONS AND STRUCTURAL EVOLUTION. Ellen D. Williams, Materials Research Science and Engineering Center, University of Maryland, College Park, MD.

Structural transformations, whether they involve long-range mass transport or phase changes, are governed by the underlying stochastic behavior at the molecular scale. The cumulative effect of correlated molecular fluctuations results in fluctuations at the nanometer scale at structural boundaries. These fluctuations can be observed in real-time, real-space imaging using scanned probe techniques. Analysis of their temporal and spatial correlations allows the rate-limiting processes governing structural evolution to be determined, and the appropriate rate parameters quantified. Experimental observations of such fluctuations, for instance at crystalline boundaries (steps) on lead and aluminized silicon surfaces will be presented. The analysis of the observations in terms of structural persistence and rate limiting processes will be described. The application of the physical understanding obtained from the fluctuation analysis to predictions of structural evolution will be illustrated for the case of crystallite reshaping. The extensions of the formalism to understanding the evolution of ferro-electric phase boundaries will be discussed and briefly illustrated for scanned probe observations of polarization relaxation in ferroelectric thin films.
Supported by the NSF-MRSEC under grant #DMR-00-80008.

11:15 AM *D5.4
SPIN DYNAMICS IN MAGNETIC THIN FILM STRUCTURES. Jian-Gang Zhu, Carnegie Mellon University, Department of Electrical and Computer Engineering, Data Storage Systems Center, Pittsburgh, PA.

Over the past decade, micromagnetic, modeling has significantly enriched our understanding of magnetization behavior in thin film magnetic structures at length scales significantly below a micron, resulting development of various magnetic thin film devices and material microstructure with much improved properties. The talk will focus on the dynamic properties of microscopic magnetization structures in various small thin film elements, especially, the formation, evolution, and annihilation of magnetization vortex structures in patterned magnetic thin film elements. Studies on thermally excited ferromagnetic resonance in confined thin film structures and its impact in many applications will also be presented.

SESSION D6: DYNAMICS IN DIELECTRICS, RELAXORS AND ORGANIC FERROICS
Chair: Ichiro Takeuchi
Thursday Afternoon, November 29, 2001
Room 205 (Hynes)

1:30 PM *D6.1
CORRELATION BETWEEN LATTICE DYNAMICS AND LOW FREQUENCY DIELECTRIC PROPERTIES OF FERROELECTRIC THIN FILMS. X.X. Xi, A.A. Sirenko, D.A. Tenne, A.M. Clark, A. James, K. Chen, Department of Physics, The Pennsylvania State University, University Park, PA.

Lattice dynamics is of central importance for the mechanism of ferroelectricity. In particular, the soft mode behavior are directly related to many of their ferroelectric and dielectric properties. We report the experimental studies of the vibrational spectra of $SrTiO_3$ films grown by pulsed laser deposition. Raman scattering, with and without bias electric field, and Fourier-transform far-infrared ellipsometry were utilized. These results are compared with the low-frequency dielectric properties. We found that in the films the soft mode is harder compared to that in bulk crystals, in agreement with the Lyddane-Sachs-Teller formalism. The existence of local polar regions is proposed as an important factor determining the dielectric properties of ferroelectric thin films. The study was extended to $Ba_xSr_{1-x}TiO_3$ films with Ba contents x = 0.05, 0.1, 0.2 and 0.5. The temperature dependence of the soft mode frequency shows evidence of the ferroelectric phase transition in the films. Relative Raman intensity of hard phonon modes shows the ferroelectric phase transition occurs over a broad range of temperatures in thin films, which is different from bulk behavior. Comparison of temperature evolution of Raman spectra for films grown on $SrTiO_3$ and $LaAlO_3$ substrates shows the influence of strain on the temperature of ferroelectric phase transition.

2:00 PM *D6.2
OBSERVATION OF HIGH FREQUENCY DIELECTRIC RELAXATION IN BARIUM STRONTIUM TITANATE THIN FILMS. James C. Booth, Ronald H. Ono, NIST, Boulder, CO; Ichiro Takeuchi, Kao-Shuo Chang, Univ. of Maryland, College Park, MD.

Ferroelectric thin films such as $Ba_{0.3}Sr_{0.7}TiO_3$ hold promise for electronically tunable microwave devices. However, the losses in such films are unacceptably high, while the origins of the losses are poorly understood. We report evidence that a major contribution to the enhanced losses in these materials at microwave frequencies comes from dielectric relaxation effects. We directly observe dielectric relaxation in the frequency dependence of the measured complex permittivity in $Ba_{0.3}Sr_{0.7}TiO_3$ thin films over a wide temperature range. By fitting both the real and imaginary parts of the measured permittivity to the Cole-Cole function over the frequency range 0.05 to 40 GHz, we are able to directly obtain values for the relaxation time for this material over the temperature range from 55 to 295 K. The relaxation time obtains a minimum value of approximately 1 ps at the "Curie temperature" (the temperature at which the real part of the temperature-dependent permittivity obtains a maximum). Above the Curie temperature the relaxation time appears to be thermally activated, with a barrier height on the order of 0.05 eV. These results are consistent with relaxation of the polarization of a distribution of small polarized regions throughout the film. We will discuss the effects of defects and of film growth conditions on the value and distribution of the relaxation times, and discuss the implications of these results for reducing losses in ferroelectric thin films for device applications at GHz frequencies.

2:30 PM D6.3
COOPERATIVE LATTICE DYNAMICS OF Li DOPED $KTaO_3$ AT SMALL AND MODERATE Li CONCENTRATIONS.
S.A. Prosandeev, V.A. Trepakov, M.E. Savinov, L. Jastrabik, S.E. Kapphan, Physics Department, Rostov State University, Rostov on Don, RUSSIA; A.F. Ioffe Physical- Technical Institute, St. Petersburg, RUSSIA; Institute of Physics AS CR, Praha, CZECH REPUBLIC; FB Physik, University of Osnabrück, Osnabrück, GERMANY.

The frequency and temperature dependencies of the dielectric permittivity of $KTaO_3$:Li were measured for dilute and moderately concentrated samples. A coupling mode model was employed in order to describe the results obtained. It is shown that a deviation of the data obtained from the Arrhenius law for the dilute sample can be explained by the strong temperature dependence of the dielectric permittivity. The characteristics of the potential barrier distribution function for the moderately concentrated sample are found to depend on temperature. In particular the mean potential barrier is found to be strongly linearly decreasing with temperature that explains earlier found unphysical values of the preexponential factor obtained for π-relaxation from the Arrhenius plot.

2:45 PM D6.4
FIELD-INDUCED EVOLUTION OF NANODOMAIN STRUCTURES IN RELAXOR X/65/35 PLZT CERAMICS.
Vladimir Ya. Shur, Gennadii G. Lomakin, Stanislav S. Beloglazov, Stanislav V. Slovikovski, Oksana V. Yakutova, Ekaterina V. Nikolaeva, Eugene I. Shishkin, Ural State Univ., Inst. of Phys. & Appl. Math., Ekaterinburg, RUSSIA; Andris Sternberg and Andris Krumins, Inst. of Solid State Phys., Univ. of Latvia, Riga, LATVIA.

We report the study of field-induced kinetics of multi-domain polar micro-regions in nonpolar matrix by in situ recording of scattered light intensity and switching charge/current during application of field pulses and by visualization of the static nano-domain patterns using scanning probe microscopy (SPM) in x/65/35 PLZT ceramics. Fractal approach enables us to gain the information about the nano-domain kinetics in wide temperature range. We have investigated the 100-micron-thick-plates of coarse-grained hot-pressed (5-12)/65/35 PLZT ceramics covered by transparent electrodes. The sequence of instantaneous angular dependence of scattered light intensity measured in transmitted mode and switching current was recorded

during application of the rectangular and triangular field pulses. The reversible field-induced intensity variation is observed within the temperature range from the dielectric permittivity maximum T_m to complete transition from relaxor to ferroelectric state (freezing temperature) T_f. Supposing that the heterophase structure is a self-similar (fractal) object its field-induced evolution has been characterized by the fractal scaling range (correlation length/upper cutoff), which diminishes essentially during backswitching. The correlation length decreasing during cooling is due to diminishing of mean size of windows in infinite percolation cluster formed at T_m. The parameters characterizing the kinetics of field-induced irreversible enlargement of nano-scale domain structure at $T<T_f$ have been obtained by statistical treatment of SPM patterns. Analysis of the switching current data recorded in relaxor phase allows us to determine the activation field for switching of the nano-scale multi-domain structure. The research was made possible in part by Programs "Basic Research in Russian Universities" (Grant No.5563) and "Priority Research in High School. Electronics" (Grant No. 03-03-29), by Grant No. 01-02-17443 of RFBR, by Award No.REC-005 of CRDF.

3:30 PM D6.5
MATERIALS SIMULTANEOUSLY EXHIBITING SUPER-PARAELECTRIC AND SUPERPARAELASTIC BEHAVIOR. Andrey Soukhojak, Yet-Ming Chiang, Massachusetts Institute of Technology, Dept of Materials Science and Engineering, Cambridge, MA.

Recent time-resolved electromechanical measurements of relaxor ferroelectrics based on sodium bismuth titanate at different frequencies of the applied field, and their macroscopic modeling [Soukhojak & Chiang, J. Appl. Phys. 88 [11] 6902 (2000)], provide evidence for independent time-dependent responses of polarization (P) and strain (S) to the applied electric field (E). This decoupling in the time domain has not been considered in previous microscopic models of relaxor ferroelectrics, which implicitly assume that polarization and strain are coupled in a time-independent manner in addition to explicitly stated relaxation of field-induced polarization. We show that polarization and strain are in general independent order parameters and their relaxation as a result of changing applied electric field can occur independently. In this case, two kinetic equations are necessary to describe the time dependent responses: (1) E-P, electric field induced polarization and (2) P-S, polarization induced strain. We consider the simultaneous existence of ferroelectric and ferroelastic nanodomains, the latter interacting constructively to result in superparaelastic response, to be the most plausible explanation of the different time-dependent electromechanical responses. Experimentally this behavior has been seen in both poly- and single crystals that show no hysteresis at vanishing frequencies. The different E-P (superparaelectric) and P-S (superparaelastic) relaxations may be attributed to different sensitivities of ferroelectric and ferroelastic domain sizes to ionic disorder and other quenched defects. In addition to electromechanical and dielectric data, we present optical and HREM observations that support the existence of such nanodomains. The theoretical possibility of static P-S decoupling is shown by Landau-Ginzburg expansion of the free energy in terms of polarization and strain. Finally, a microscopic model of simultaneous superparaelectricity-superparaelasticity utilizing the concept of the energy landscape will also be presented. Research supported by ONR Grant No. N00014-97-0989 and AFOSR/DARPA Grant No. F49620-99-2-0332.

3:45 PM D6.6
INFLUENCE OF THE MICROSTRUCTURES ON THE DIELECTRIC PROPERTIES OF $ZrTiO_4$ THIN FILMS AT MICROWAVE-FREQUENCY RANGE. Yongjo Kim, Jeongmin Oh, Tae-Gon Kim, Byungwoo Park, Seoul National Univ, School of Materials Science and Engineering, Seoul, KOREA.

The dielectric properties of paraelectric $ZrTiO_4$ thin films were investigated in the microwave-frequency range. The dielectric losses (δ) and dielectric constants (ϵ) were successfully measured up to ~5 GHz using a circular-patch capacitor geometry[1]. The effects of the microstructures on the dielectric properties were also investigated. The $ZrTiO_4$ films were deposited at different temperatures and RF power densities to vary the film microstructures. As the deposition temperature and RF power density were increased, the film crystallinity was enhanced and dielectric losses decreased. The microwave dielectric losses correlated very well with the level of local strain and unit-cell dilation, while the dielectric constants did not alter significantly. [1]Y. Kim et al., Appl. Phys. Lett. 78, 2363 (2001).

4:00 PM D6.7
SOME INVESTIGATIONS OF THE DOMAIN-LIKE PROCESSES IN THE MODEL FERROELECTRIC RELAXOR LEAD MAGNESIUM NIOBATE BY THE ELECTROLUMINESCENCE. Nataliya N. Krainik, Ioffe A.F. Physicotechnical Institute RAS, St. Petersburg, RUSSIA; Svetlana A. Flerova, Dniepropetrovsk State Univ, Dept of Physics, Dniepropetrovsk, UKRAINE; Svetlana A. Sushko, Ludmila Ya. Fomichova, Dniepropetrovsk, National Mining University of Ukraine, Dniepropetrovsk, UKRAINE.

In the report is discussed a cycle of investigations on the study of the processes of switching of polarization by luminescence method in the crystals of the model ferroelectric (and ferroelastic) of lead magnesium niobate (PMN) with diffuse phase transition (DPT) in the transition range. The obtained temperature, field and frequency dependencies of the intensity of electroluminescence in different regimes of exciting fields testify to a relation of luminescence in the range of DPT with the passing of collective processes of a change of polarization. In the range of DPT in PMN there was discovered a luminescence with the decreasing and stopping of the action of the electric field, occurring as a result of destruction of polarized regions of the ferrophase at depolarization of a crystal. There was shown and studied the influence of the processes of depolarization in sinusoidal electric fields/The discovered bell-form maximum and small anomalies (at the temperature T=230K) in the temperature dependence of electroluminescence intensity in the PMN testify to a heterogeneous temperature dependence of the polar phase concentration. The discovery and study of tensoluminescence in PMN in the range of DPT confirmed that even in the absence of external electric field, in a crystal appears some spontaneously deformed regions of the polar phase which can be collectively reoriented by an external mechanical stress. With decreasing of mechanical stress a luminescence is seen as a result of destruction of the state appearing from the processes of increasing stresses. It is possible that electroluminescence at the depolarization and tensoluminescence at the falling mechanical stresses in PMN result from the processes of leaking type in the reverse phase transition in some volumes of ferroelectric phase to the original homogeneous state. This work was supported by the Russian Fund for Fundamental Research, Grant N01-02-17801.

4:15 PM D6.8
ELECTRIC AND MAGNETIC INVESTIGATIONS OF $BiFeO_3$ AND RELATED LAYERED-TYPE COMPOUNDS. Matthew R. Suchomel, Peter K. Davies.

The simultaneous ferroelectric and ferromagnetic ordering of the perovskite $BiFeO_3$ has generated much interest in its possible use for novel magnetoelectric device applications. Studies of several $BiFeO_3$ based solid solutions have been previously reported. However, due to the difficult nature of synthesis, true single phase $BiFeO_3$ has been inadequately examined. In this work the effectiveness of several synthesis routes to produce single-phase material is studied. A multi-step leaching and reheating solid-state synthesis route has been found to minimize secondary phases. The electric and magnetic properties of samples with varying impurity type and concentration were studied at both bulk and microstructural levels and related to composition. Additionally, solid solution with $Pb_2Fe_2O_5$ (<30 mol%) was found to decrease secondary phase growth and reduce sample conductivity. Magnetic and electric data from these samples will be reported and discussed. More complex $BiFeO_3$ related layered-type compounds, such as fluorine substituted Aurivillius and so called 'Bipox' phases, have also been synthesized. Their previously unreported magnetoelectric properties will be presented and discussed. This work is supported by the National Science Foundation through the MRSEC program.

SESSION D7: POSTER SESSION
Thursday Evening, November 29, 2001
8:00 PM
Exhibition Hall D (Hynes)

D7.1
PHASES AND POLARIZATION ANISOTROPY IN RELAXOR-BASED FERROELECTRIC CRYSTALS $(PbMg_{1/3}Nb_{2/3}O_3)_{0.68}$-$(PbTiO_3)_{0.32}$ (PMN-0.32PT). Chi-Shun Tu, Chi-Long Tsai, J.-S. Chen, Fu Jen Univ, Dept of Physics, Taipei, TAIWAN; V. Hugo Schmidt, Montana State Univ, Dept. of Physics, Bozeman, MT.

Polarization-electric field hysteresis loops and domain structures have been measured as functions of temperature and external E-field in relaxor-based ferroelectric PMN-xPT single crystals for x=0.32 with {110} and {211} orientations. The PMN-0.32PT crystal exhibits apparent crystalline anisotropy in polarization and domain structures. In order of increasing temperature, the crystal undergoes successive phase transitions: rhombohedral phase, coexistence of rhombohedral phases, tetragonal phase, coexistence of tetragonal and cubic phases, cubic phase. In addition, a field-induced tetragonal symmetry was evidenced and coexists with rhombohedral symmetry in the low-temperature region.

D7.2
Abstract Withdrawn.

D7.3
DIFFUSE DIELECTRIC ANOMALY IN PEROVSKITE OXIDE MATERIALS. Byung S. Kang, Si K. Choi, Korea Advanced Institute of Science and Technology, Dept. of MS&E, Taejon, SOUTH KOREA.

The diffuse dielectric anomaly observed in the temperature range of 400~700°C was investigated in various perovskite oxide materials. The detailed data on the diffuse dielectric anomaly was obtained in $Pb(Zr,Ti)O_3$, $(Pb,La)TiO_3$, $(Pb,La)(Ti,Zr)O_3$, $BaTiO_3$ single crystal and ceramic and etc. The frequency and temperature dependence of the diffuse dielectric anomaly was phenomenologically explained using the modified Debye equation. The diffuse dielectric anomaly in perovskite materials was successfully described by introducing the relaxation strength as a fitting parameter. The effect of the heat treatment on the diffuse dielectric anomaly was studied to discuss whether the diffuse dielectric anomaly is intrinsic or extrinsic effect effect of the materials. The role of donors and acceptors on the diffuse dielecltric anomaly was also investigated.