

“Ferroelectric-relaxors” can be distinguished from “classic ferroelectrics” by a diffuse ferroelectric-paraelectric phase transition where ϵ' reaches a broad high peak value with a strong frequency dispersion (the temperature at the maximum T_m increases with frequency). Additionally, the spontaneous polarization P_s is not suddenly lost at T_m , but decays more gradually to zero (at T_{Burns}) with increasing temperature. On cooling from high temperatures, $\text{PbZn}_{1/3}\text{Nb}_{2/3}\text{O}_3$ relaxors (PZN) transform from a paraelectric state to a state with ferroelectric nanodomain fluctuations in a paraelectric matrix and, to a disordered low-temperature polar phase. The existence of polar Nano-Regions in these materials, which can be associated with local lattice distortions with respect to the average structure can be linked to the presence of diffuse scattering in diffraction experiments. From a vibrational viewpoint, a ferroelectric polarization is found to exist based on the recovery of the soft optic mode without a rhomboedral distortion. From a technological viewpoint, PZN exhibits giant electromechanical coupling with a relatively modest applied electric field along the [001] direction. Many studies are related to the competition between short-range and long-range order in these materials, which is supposed to be responsible for the giant piezoelectric response. In this contribution, we present the first local “in-situ” investigation as a function of $\vec{E}\vec{E}$ to really understand what occurs at the atomic scale.

X-Ray Absorption Spectroscopy was performed at the 3 absorption-edges of interest in PZN (i.e., Pb(L_{III}), Zn(K), Nb(K)) and collected in the fluorescence mode as a function of the electric field up to 10 kV/cm along the [001] direction on BM29 at the ESRF. Careful analyses of our EXAFS data linked with an entire dielectric/piezoelectric characterization have clearly enabled us to determine the polarisation process, i.e. between the unpoled and poled states; above a critical value of $\vec{E}\vec{E}$ (the coercitive field here is about 3 kV/cm), one can observe the persistence of the polarization even when the $\vec{E}\vec{E}$ is removed. Additionally, our data permitted us to follow the local distortion of the Zn and Nb atoms in their oxygen octahedra cage, giving rise to the piezoelectric effect, to field reaching 10 kV/cm!!!

Keywords: ferroelectric-relaxor; X-ray absorption spectroscopy; electric field

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Domain Structure in $\text{PbZr}_{1-x}\text{Ti}_x\text{O}_3$, Roland Schierholz^a, Hartmut Fuess^b. ^a*Institute for Materials Science, Technische Universität Darmstadt, Germany.*

E-mail: schierholz@st.tu-darmstadt.de

The *Cm* phase of $\text{PbZr}_{1-x}\text{Ti}_x\text{O}_3$ (PZT) for compositions at the morphotropic phase boundary (MPB) has been proposed based on X-ray diffraction results [1]. Also nanodomains were observed in this compositional region [2]. We observed monoclinic symmetry within single domains of $\text{PbZr}_{0.54}\text{Ti}_{0.46}\text{O}_3$, by Convergent-Electron Beam Diffraction (CBED) [3].

The monoclinic phase allows 24 polarization directions. In

PZT the low temperature monoclinic phase is a subgroup of a tetragonal phase at higher temperatures. Due to group-subgroup relations four domains with $\langle uuv \rangle$ polarization can form within one tetragonal domain. In former 90° micro domain walls domains with $\langle uuv \rangle$ and $\langle uvu \rangle$ polarizations meet. This leads to restrictions in lattice parameters in the (110) plane for polarizations that cannot be transformed by a single twin operation.

On the rhombohedral site of the MPB within one rhombohedral micro domain at most three different domains can be formed. In experiments a finer nanodomain contrast is observed with orientations of the nanodomain walls, that differ from those on the tetragonal site. These nanodomain walls can be explained as domain walls of the *Cm* phase as a subgroup of *R3m* instead of *P4mm*.

An *in situ* study revealed a change from a tetragonal/monoclinic shape of the 110_{pc} reflection in X-ray diffraction to a more rhombohedral appearance with cycling [4]. For a sample cycled *ex situ* wide monoclinic domains and rhombohedral appearing domain configurations exist. Those rhombohedral domains show an inner contrast that is consistent with nanodomains. So we discuss the possibilities for a rearrangement of monoclinic nanodomains formed on the *P4mm* to *Cm* transition under an electrical field.

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Electron Diffuse Scattering in epitaxially grown SrTiO₃ thin film, Jérôme Pacaud^a, Frédéric Pailloux^a. ^a*Laboratoire PhyMat Université de Poitiers, France.* E-mail: jerome.pacaud@univ-poitiers.fr

The main requirement for many devices is the growth of a high purity and high structural quality thin film. The perovskite structure is extremely sensitive to the deposition condition and particularly to the temperature and the partial pressure of oxygen. Changes in deposition conditions may lead to a large deviation of the dielectric properties of thin films from those of bulk materials. The chemistry of defects is often proposed as an explanation of this deviation.

Another important parameter for thin perovskite films is the geometrical constraint imposed by the substrate. Most of the time, perovskite exhibit excellent epitaxy on each other and the films are tied to the substrate so the in-plane parameters are not free to reach their bulk equilibrium values. For materials as sensitive to phase transition as perovskite this effect and the associated relaxation processes can be extremely important for the fine tuning of the physical properties of the film.

For perfect crystal, diffuse scattering is mostly inelastic due to phonons, plasmons and other processes. Thermal

diffuse scattering or phonon scattering can lead to large effects in the electron diffraction pattern especially near second order phase transition where phonon softening occurs. Additionally, elastic diffuse scattering comes from structural deviations from a periodic lattice [1]. These structural deviations can be defects, partial ordering of otherwise disordered structure or structural fluctuations. By studying diffuse scattering we can obtain information about the crystal imperfections and dynamics which can not be obtained from other characterization methods.

The goal of this study is to characterize, through diffuse scattering in electron diffraction, the structure of defects in the epitaxial layers of perovskite structure and more specifically its influence on the dynamic of the lattice (lattice vibrations, structural fluctuations and continuous phase transition due to phonon softening) [3]. These properties have to be linked with the anomaly of the ferroelectric properties of such thin films. Different strain states can be probed by changing the substrate material or introducing different buffer layers.

The results of STO films deposited on different substrate show different behavior. On STO substrate, at 135K, the superstructure spots in the thin film, have already the same FWHM than the fundamental reflections, indicating a complete phase transition. This shift toward higher transition temperature in thin STO films has already been observed [4]. On LaAlO₃ (LAO) substrate, fine peaks appear near 140 K in the diffraction pattern of the film revealing a strain induced phase transition.

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Keywords: thin ferroelectric films; electron diffraction; phase transition

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Magnetic and Dielectric Properties of Fe based Langasites. Pierre Bordet^a, Karol Marty^a, Virginie Simonet^a, Mickael Loire^a, Eric Ressouche^b, Rafik Ballou^a, Celine Darie^a, Jakob Kljun^a, Olivier Isnard^a, Bartosz Zawilski^a, Pascal Lejay^a, Charles Simon^c. ^aInstitut NEEL, CNRS & UJF, Grenoble, France. ^bINAC, SPSMS/MDN, CEA, Grenoble, France. ^cCRISMAT, CNRS ENSICAEN, Caen, France.

E-mail: pierre.bordet@grenoble.cnrs.fr

We have investigated the magnetic and dielectric properties of the non-centrosymmetric Fe containing langasites of type A₃BFe₃D₂O₁₄ with A=Ba, Sr, Ca, B=Ta, Nb, Sb and D=Ge, Si. Using single crystal and powder neutron diffraction, a novel doubly chiral magnetic order was found in these structurally chiral compounds [1], in which the Fe cations form planar triangular lattices of triangle

units. The magnetic structure was found to be a helical spin arrangement propagating along the c axis of equal moments lying in the (a,b) plane at 120° from each other within each triangle. This 120° arrangement results from the usual compromise of frustrated Heisenberg spins on a triangle-based lattice. For all the investigated compounds, the magnetic ordering was found remarkably similar, with T_c ~ 25K and a propagation vector $\tau \sim 1/7$, except for those having the B site occupied by Sb⁵⁺, for which T_c ~ 35K and τ was closer to 1/5. Unpolarized neutron scattering on a single crystal associated with spherical neutron polarimetry proved that a single triangular chirality together with a single helicity was stabilized in a crystallographically enantiopure crystal. A mean-field analysis allowed us to show that the magnetic and structural chiralities are related through a twist in the plane to plane super-superexchange paths. The langasite structure is non-centrosymmetric P321 and a further lowering of symmetry is achieved in the magnetic phase leaving an overall polar structure for which the appearance of a spontaneous electric polarization is allowed. High resolution synchrotron powder diffraction suggests that the magnetic phase transition could be associated to a structural one. Dielectric measurements reveal the presence of magneto-electric coupling through an anomaly at the Néel temperature in the thermal variation of the dielectric permittivity and its quadratic dependence with the magnetic field. The Fe-langasite could then be a new example enlarging the class of magnetoelectric/multiferroic triangle-based antiferromagnets

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Ferroelectric Ordering of Cd₂Nb₂O₇. Thomas Malcherek. *Mineralogie, Universität Hamburg, Germany.*

E-mail: thomas.malcherek@uni-hamburg.de

The pyrochlore Cd₂Nb₂O₇ (CNO) exhibits unusual ferroelectric properties. Similarity to relaxor materials is indicated by a frequency dependence of the dielectric function over a limited temperature range below 200K. Contrary to standard relaxor material, the formation of polar nanoclusters associated with this behaviour has to be explained in the absence of chemical mixing. Heat capacity anomalies have been observed immediately below 204K, at 85K and at 46K [1]. The exact crystal structure of the corresponding ferroelectric low temperature phases has remained largely undetermined because of their strongly pseudo-cubic nature. Recently the low temperature structure of CNO has been explored by all electron ab-initio calculations in the framework of density functional theory [2]. According to this study, CNO distorts to monoclinic symmetry (space group Cc) at low temperatures via an intermediate orthorhombic phase (space group Ima2). The calculated monoclinic low temperature structure is