

DF 3: Relaxor Ferroelectrics

Time: Monday 14:30–17:30

Location: H11

Invited Talk

DF 3.1 Mon 14:30 H11

Investigation of relaxor ferroelectrics by piezoresponse force microscopy — ●VLADIMIR SHVARTSMAN and WOLFGANG KLEEMANN — Angewandte Physik, Universität Duisburg-Essen, D-47048 Duisburg, Germany

Relaxor ferroelectrics (relaxors) belong to a special group of polar oxides. In contrast to normal ferroelectrics, dynamic polar nanometer-sized regions (PNRs) appear in these materials deep in the nominally paraelectric state. On cooling a transition either into a short-range ordered cluster-glass-like state or into a long-range ordered domain state occurs. In spite of numerous intense investigations the information about the evolution of the PNRs, in particular in the vicinity of the transition temperature, is still insufficient. New information about the polar structure of relaxors may be obtained by piezoresponse force microscopy (PFM). We present results of PFM studies done both on uniaxial relaxors $\text{Sr}_{1-x}\text{Ba}_x\text{Nb}_2\text{O}_6$ (SBN) and on cubic relaxors $\text{Pb}[\text{Mg}_{1/3}\text{Nb}_{2/3}]_{1-x}\text{Ti}_x\text{O}_3$ (PMN-PT). Peculiarities of the polar structures below the transition temperature are analyzed. Quasi-static polar regions are revealed in a certain temperature range above T_C . In both cases the sizes of the observed clusters are much larger than expected for PNRs. The temperature dependence of the observed structures was investigated. The origin of these mesoscale polar regions, their relationship with the PNRs and their role in the phase transition are discussed.

DF 3.2 Mon 15:10 H11

Electric-field induced phase transition in near-electrode region of $\text{PbMg}_{0.33}\text{Nb}_{0.67}\text{O}_3$ - 28% PbTiO_3 relaxor - ferroelectric (001) single-crystal wafer — ●ALEXANDER A. LEVIN, ANJA I. POMMICH, TORSTEN WEISSBACH, and DIRK C. MEYER — Institut für Strukturphysik, Technische Universität Dresden, 01062 Dresden, Germany

A (001) single-crystal $\text{PbMg}_{0.33}\text{Nb}_{0.67}\text{O}_3$ - 28% PbTiO_3 (PMN - 28% PT) wafer was investigated by means of Wide-Angle X-ray Scattering (WAXS) techniques in situ under the influence of an external static unipolar electric field (field strength E up to 13 kV/cm). Changes of profiles of X-ray 001 reflections of 1st and 3d orders and their rocking curves were recorded under applying the electric field along the [001] direction of the single-crystal plate. Evidences of a reversible phase transition between ferroelectric modifications of PMN-PT in a near-electrode region of about 1 micrometer depth are found. Under the influence of the static electric field, a part of the near-electrode layer (of about 5 vol. % and 15 vol. % at $E = 4.5$ kV/cm and 13 kV/cm, respectively) transforms from initially rhombohedral type of the structure (R, space group $R\bar{3}m$) to another one (most probably, monoclinic Mb (space group Cm)). The transition is manifested by change of the phase lattice constant ($a = 4.0227(1)$ Å and $c = 4.016(1)$ Å according to interpolation to $E = 0$ kV/cm for R and Mb phases, resp.), piezoelectric coefficient ($d_{33} = 1092(14)$ pC/N (R) and $1735(248)$ pC/N (Mb), resp.) and the profile shape of the rocking curves. Financial support by the DFG research unit FOR 520 is gratefully acknowledged.

DF 3.3 Mon 15:30 H11

Relaxor ferroelectrics: ferroic clustering and phonon anomalies — ●BORIANA MIHAILOVA¹, ANNA-MARIA WELSCH¹, BERND GUETTLER², MARIN GOSPODINOV³, and ULRICH BISMAYER¹ — ¹Mineralogisch-Petrographisches Institut, Universität Hamburg, Grindelallee 48, D-20146 Hamburg — ²PTB, Bundesallee 10, D-38116 Braunschweig — ³ISSP-BAS, Blvd. Tzarigradsko Chausse 72, 1784 Sofia

Relaxor ferroelectrics are among the key materials in modern solid-state science due to their outstanding dielectric, electro-optic, and electro-elastic properties. Because of their complex local structure studies on the relaxor structure require the application of fine-scale structure-sensitive methods as inelastic light scattering, in addition to the conventional diffraction methods. Here we report on the relationship between the observed phonon anomalies and the local atomic arrangements in Pb-based perovskite-type relaxors studied on the basis of model representative lead scandium tantalates/niobates doped with various elements. It is shown that the ferroic clustering and the development of ferroelectric state can be followed by analysing quantitatively the intensity ratios of the Raman scattering arising from

the corresponding local structural distortions. The presence of point defects such as oxygen vacancies and incorporation of additional elements in the cation positions heavily influences the incipient ferroic species and suppresses the formation of proper ferroelectric state, thus favouring the non-ergodic state.

DF 3.4 Mon 15:50 H11

Crystal structure of the relaxor ferroelectric $\text{PbSc}_{0.5}\text{Ta}_{0.5}\text{O}_3$ from 300 K to 20 K — ●BERND MAIER¹, JÖRG IHRINGER¹, BORIANA MIHAILOVA², and ULRICH BISMAYER² — ¹Institut für Angewandte Physik, auf der Morgenstelle 10, 72076 Tübingen — ²Mineralogisch-Petrographisches Institut, Universität Hamburg, Grindelallee 48, 20146 Hamburg

High resolution X-ray powder data of $\text{PbSc}_{0.5}\text{Ta}_{0.5}\text{O}_3$ (PST) collected at $20 < T < 300$ K with a modified image plate Camera (Huber) in Guinier geometry exhibits diffuse phase transitions from the cubic ($Pm\bar{3}m$) paraelectric state to the rhombohedral ferroelectric state. Small domains of Sc/Ta -ordering lead to a doubled unit cell ($Fm\bar{3}m$) which gives weak and broad reflections. Also, the thermal dependence of lattice constants will be given.

Changes in behaviour due to Ba doping at the Pb sites and Ru doping at the Sc/Ta sites will be discussed.

DF 3.5 Mon 16:10 H11

Defect structure in acceptor- and donor-doped PZT compounds — ●RUEDIGER-A. EICHEL — Edurad-Zintl-Institut, TU Darmstadt

The macroscopic properties of functional PZT compounds may considerably be modified by doping with rare-earth or transition-metal ions. High-frequency and multi-pulse EPR provides a local probe to characterize functional centers and defect states in acceptor- and donor-doped PZT ceramics.

In hard-doped $\text{Fe}^{3+}:\text{PbTiO}_3$, high-frequency EPR proved the existence of an iron-oxygen vacancy defect associate - the orientation of this defect dipole being oriented persistently along the crystallographic c -axis and anti-parallel with respect to the orientation of spontaneous polarization. For the other end-member composition $\text{Fe}^{3+}:\text{PbZrO}_3$, again the creation of defect associates was observed. However, there is no persistent orientation of the corresponding defect dipole, but rather a 'multi-site*' situation that is characterized in a way that the compensating oxygen vacancy may be localized at any of the six possible neighbour oxygen sites.

In contrast, acceptor-type $\text{Cu}^{2+}:\text{PZT}$ modifications show no association of the Cu^{2+} functional centers to charge-compensating oxygen vacancies. This conclusion is based on the observation of an orientation-independent, quasi-isotropic ^{207}Pb -hyperfine coupling. As a consequence, considering the EPR results it could be predicted the Cu^{2+} -modifications induce an ionic conductivity in PZT compounds.

DF 3.6 Mon 16:30 H11

Surface barrier layer contacts and colossal dielectric constants in calcium-copper-titanate — ●STEPHAN KROHNS¹, PETER LUNKENHEIMER¹, STEFAN EBBINGHAUS², and ALOIS LOIDL¹ — ¹Experimental Physics V, Center for Electronic Correlations and Magnetism, University of Augsburg, 86135 Augsburg, Germany — ²Solid State Chemistry, University of Augsburg, 86135 Augsburg, Germany

For tomorrow's energy-storage technologies and wireless electronics, materials with high dielectric constants are in great demand. A prominent example for this kind of materials is the perovskite-related $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$ (CCTO) [1]. It shows dielectric constants up to 10^5 being constant over a relatively broad temperature range. The origin of the colossal dielectric constant in CCTO is controversially debated and ascribed to internal barriers (e.g. grain boundaries) or external surface effects (e.g. electrode polarisation) [2,3]. A variety of single and polycrystalline samples, subjected to different treatments, like annealing, surface polishing and surface metallization, are investigated by broadband dielectric spectroscopy. There are at least two relaxations in CCTO, the second one being connected to even higher absolute values than the first one. From our experiments we obtain strong indications for a surface related origin of both relaxation modes and the colossal dielectric constants in CCTO.

[1] C.C. Homes, T.Vogt, S.M. Shapiro, S. Wakimoto, and A.P.

Ramirez, Science 293, 673 (2001). [2] P. Lunkenheimer, et al., Phys. Rev. B 66, 052105 (2002). [3] P. Lunkenheimer, et al., Phys. Rev. B 70, 172102 (2004).

DF 3.7 Mon 16:50 H11

Characterization of piezo ceramics on the nanoscale by Conducting AFM — YUE HOU, ●ANDREI ANDREEV, and CHRISTIAN TEICHERT — Institute for Physics, University of Leoben, Franz-Josef-Str., 18, A-8700 Leoben, Austria

The Conducting Atomic-Force Microscope (C-AFM) is a conventional AFM working in contact mode, where the usual AFM tip is replaced by a conductive tip. Between the tip and the sample a voltage is applied and the resulting current is measured using a special amplification circuit. The C-AFM is well known as a valuable tool for nanometer scale electric characterization of very thin oxide layers [1,2].

In this work we demonstrate the analytical capabilities of C-AFM technique for spatially resolved electrical investigations of cross sectional piezo ceramic samples (PZT). The applicability of C-AFM for the characterization (2D lateral current mapping and local I-V curves) of such complex nanostructures is presented and discussed in terms of electrical active grain boundaries in the ceramic matrix in the vicinity of the metal electrodes.

[1] S. Kremmer, S. Peissl, C. Teichert, F. Kuchar, H. Hofer, Mat. Sci. Eng. B102 (2003), 88.

[2] S. Kremmer, H. Wurmbauer, C. Teichert, G. Tallarida, S. Spiga,

C. Wiemer, M. Fanciulli, J. Appl. Phys. 97/7 (2005), 74315-1-7.

DF 3.8 Mon 17:10 H11

Stability of polypropylene ferroelectrets against ionizing alpha-radiation — ●MARIO DANSACHMÜLLER, IVAN MINEV, FRANCISCO CAMACHO-GONZALEZ, SIMONA BAUER-GOGONEA, REINHARD SCHWÖDIAUER, and SIEGFRIED BAUER — Soft Matter Physics, Johannes-Kepler University, Altenbergerstraße 69, 4040 Linz, Austria

Cellular polypropylene (cPP), a thin foil containing flat lens-like voids displays similarities to ferroelectric materials, after internally charging the surfaces of the voids, hence cPP is called a ferroelectret. The piezoelectric coefficient of charged cPP is proportional to the effective surface charge on the voids. By analyzing the nonlinearities in the current response of a charged cPP capacitor upon application of a sinusoidally varying ac-voltage, the piezoelectric response of cPP is directly determined from an electrical measurement alone. Samples of cPP with a density of 330 kg/m³ were irradiated by ionizing alpha-radiation of a Ra-226 (4.87 MeV) or an Am-241 (5.64 MeV) source for several hours corresponding to doses of up to 2000 Gy or 20 000 rad. The charge decay was measured in-situ during the irradiation by continuously recording the first and second harmonic contribution of the current response, by means of digital lock-in amplifiers. A decay of the relative surface charge of 5 % was observed for a dose of 50 Gy. The smallest resolvable dose in a temperature controlled measurement was found to be 1.4 Gy.