# Tsukuba Workshop on Ferroelectrics 2005

Abstract Book

Jan. 17(Mon), 2005

University Hall, University of Tsukuba Tsukuba, Ibaraki 305-8573, Japan

### Maps of the University Hall



Lectures will be in the 6<sup>th</sup> conference room of the University Hall (Third Floor).

Traffic Routes to University of Tsukuba (Tsukuba area) If you need detailed train information or timetable, please refer to the following URL. http://www.tsukuba.ac.jp/eng/navi/access.html

## **Conference Program**

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13:00-13:10	Opening address	S. Kojima (IMS, Univ. Tsukuba)
13:10-14:10	"Polar nanoclusters in re	elaxors" R. Blinc (J. Stefan Institute)3
14:10-15:10	"Ferroelectric microregi	on in quantum paraelectrics" H. Uwe (IMS, Univ. Tsukuba)4
15:10-15:20	Coffee break	
15:20-15:50	"Phase diagram and MP	B of (Na,K)NbO <sub>3</sub> -ATiO <sub>3</sub> solid solution" R. Wang (AIST)5
15:50-16:20	"Electron-soft phonon ir	nteraction in SrTiO <sub>3</sub> " H. Minami (IMS, Univ. Tsukuba)6
16:20-16:50	"Relaxation mode and correlaxor ferroelectrics"	entral peak in the vibration spectra of the
		S. Lushnikov (Ioffe. Inst.)7
16:50-17:10	"Low temperature elastic	c properties of PLZT relaxors" G. Shabbir (IMS. Univ. Tsukuba, PINSTECH. Islamabad. Pakistan)9
17:10	Closing	

18:00- Banquet (Ichinokura)

### **Polar nanoclusters in relaxors**

#### R. BLINC

#### J. Stefan Institute, Ljubljana, Slovenia

The central problem in the physics of relaxors is the nature of the polar nanoclusters which are believed to be responsible for the multi-scale dynamics, spatial inhomogeneity and many other physical properties of these materials such as giant piezo-electricity and electrostriction. Whereas relaxors are homogenous at high enough temperatures, polar nanoregions immersed in a neutral matrix are formed below a certain temperature  $T_{\rm B}$ according to Burns and Dacol. This leads to a two component system. In spite of many investigations, direct physical evidence for the existence of polar nanoregions is still lacking. Here we present direct microscopic evidence for the two component nature of relaxors. We show that the <sup>207</sup>Pb NMR spectra of these systems consist of an isotropic component corresponding to a spherical glassy matrix which does not respond to an applied electric field, and an anisotropic component, corresponding to frozen out polar nanoclusters which order in a strong enough electric field, forming a ferroelectric phase. This is as well reflected in the dynamic properties where the relaxation time distribution function  $f(\tau)$  starts to become asymmetric with decreasing temperature and a second maximum – which is never seen in dipolar glasses and is obviously due to polar clusters - appears on further cooling both in the dielectric dispersion and in the NMR  $T_2$  data. Cross and Viehland pointed out the similarity between relaxors and dipolar glasses and suggested that nanoclusters are dynamic entities with thermally fluctuating dipole moments which freeze out at low enough temperatures. The present results demonstrate that the basic difference between relaxors and dipolar glasses is their response to applied electric fields: Polar nanoclusters - corresponding to the anisotropic component in the NMR spectra – can be oriented in a strong enough applied electric field and a ferroelectric phase can be induced. This is not the case in dipolar glasses where the response is due to single dipoles which cannot be ordered by applied electric fields.

### Ferroelectric microregion in quantum paraelectrics

#### H. UWE

#### Institute of Materials Science. Univ. of Tsukuba

Ferroelectric microregions (FMR) develop in nominally pure KTaO<sub>3</sub>, SrTiO<sub>3</sub>, and Nb-doped KTaO<sub>3</sub>. Lattice vibration of ferroelectric soft-phonon branch with finite wave number through FMR has Raman-active polarization with null wave number. The shape of FMR extracted from Raman scattering profile is found to be oval with longer axis length proportional to the square root of dielectric constant. System of interacting FMRs exhibits dielectric relaxation time obeying the Vogel-Fulcher rule. Experimental finding that BaTiO<sub>3</sub> with the displacive-type phase transition exhibits order-disorder behavior is discussed with consideration of FMR.

### Phase diagram and MPB of (Na,K)NbO<sub>3</sub>-ATiO<sub>3</sub> solid solution

#### R. WANG

### Nanoelectronics Research Institute National Institute of Advanced Industrial Science And Technology

Morphotropic phase boundary (MPB) plays an important role in lead-containing solid solutions such as PZT, PMN-PT, PZN-PT and so on, since excellent dielectric/piezoelectric properties are found around it. Therefore, we have tried to develop high-performance lead-free piezoelectric materials through forming MPB in solid solutions. For this purpose, solid solution of  $(1-x)(Na_{0.5}K_{0.5})NbO_3-xATiO_3$  (A = Pb, Ba, and Sr) has been prepared and their phase diagrams have been studied by dielectric and piezoelectric properties measurement.

Phase diagram for the three solid solutions is nearly the same. Upon introducing a small amount of ATiO<sub>3</sub> (A = Pb, Ba, and Sr), both the cubic-tetragonal (C-T) and the tetragonal-orthorhombic (T-O) phase transition temperatures rapidly decrease. The transition temperature of C-T,  $t_{c1}$ , is different for different A (A = Pb, Ba, Sr). However, the transition temperature of T-O,  $t_{c2}$ , is almost independent of A (A = Pb, Ba, Sr). A tetragonal/orthorhombic MPB is found around  $x \sim 0.05$ , where  $dt_{c2}/dx$  is about -3400°C/mol. This value is much larger than  $dt_c/dx$  for the usual solid solution (e.g. -400°C/mol for (Ba,Sr)TiO<sub>3</sub>) and is the same order as the  $dt_c/dx$ ~-5800°C/mol for PZT ceramics near MPB. It is interesting to notice that with increasing x, the T-O phase transition becomes more and more diffused. The T-O phase transition is completely diffused out around x =0.05-0.06, and form a 'critical point' in the phase diagram. In the vicinity of the 'critical point', piezoelectric properties are improved to a great extent.

### **Electron-soft phonon interaction in SrTiO<sub>3</sub>**

#### H. MINAMI

#### IMS, University of Tsukuba

Electron mobility in pure  $SrTiO_3$  has been measured at low temperature by using a technique of transient photoconductivity measurement. A non-linear transport phenomenon, "streaming motion" is observed. The saturated drift velocity is much lower than expected from LO phonon, but close to that expected from soft TO phonon. The temperature dependence also suggests that the soft phonon is the predominant process of electron scattering. It is probably because ferroelectric, as well as rigid rotation, mode phonon can produce deformation potential, and because of the increase in the chances of phonon absorption and emission by the softening, and the extremely weak coupling of electrons to the LO phonons in  $SrTiO_3$ .

### Relaxation mode and central peak in the vibration spectra of

### the relaxor ferroelectrics.

S.G. LUSHNIKOV<sup>1</sup>, S.N. GVASALIYA<sup>1,2</sup> and S. KOJIMA<sup>3</sup>

<sup>1</sup>Ioffe Physical Technical Institute, 194021, 26 Politekhnicheskaya, St. Petersburg, Russia <sup>2</sup>Laboratory for Neutron Scattering ETHZ & PSI, CH-5232 Villigen PSI,

Switzerland

<sup>3</sup>Institute of Materials Science, University of Tsukuba, Tsukuba, Ibaraki 305-8573, Japan

Relaxor ferroelectrics have an abnormally broad in temperature and frequency-dependent dielectric anomaly which does not link to a structural phase transition. In the range of this dielectric anomaly many other properties of relaxors exhibit a peculiar behaviour. Temperature range where the properties of relaxors exhibit anomalous features is referred to as diffuse phase transition. Understanding of the appearance of these peculiarities has been a challenge for a long time. Despite achievements in this field there is no coherent picture of relaxor behaviour.

We have carried out investigations of the low-frequency part of vibration spectra of two model relaxor crystals –  $PbMg_{1/3}Nb_{2/3}O_3$  (PMN) and  $PbMg_{1/3}Ta_{2/3}O_3$  (PMT) by Brillouin light and neutron scattering. Dielectric anomaly in PMN appears around Tcm~270K, while PMT has Tcm~170K. Another difference between these two crystals is the presence of a structural phase transition in PMN in an applied electric field, while PMT crystal is cubic in a whole studied range of the temperatures and external fields.

Cold neutron scattering measurements were performed on TASP 3-axis spectrometer (SINQ) [1-3]. A strong quasielastic scattering and central peak in both crystals were observed in the vicinity of (001) and (110) Bragg peaks, but it was not possible to detect it in the vicinity of (002) Bragg reflection. The energy width of this quasielastic scattering was found to follow  $\Gamma_0+Dq^2$  dependence where  $\Gamma_0$  is temperature-dependent. It

was shows the q- and temperature dependence of the width of this quasielastic scattering obtained from the inelastic neutron scattering spectra of PMN in the vicinity of (001) Bragg peak. Presence of the quasielastic scattering with such properties (relaxation mode) shows that despite previous expectations order-disorder behaviour plays an important role in the dynamics of the diffuse phase transition.

In present report we would like to discuss: (1) Our new results of neutron and light scattering studies (2) Relation of our observation with previously published neutron and light scattering data.

- [1] S.N. Gvasaliya, B. Roessli, S.G. Lushnikov // Europhys. Lett. 63 (2003) 303.
- [2] S.N. Gvasaliya, S.G. Lushnikov, B. Roessli // Crystallography Reports, **49** (2004) 108.
- [3] S.N. Gvasaliya, S.G. Lushnikov, B. Roessli // Phys. Rev. B 69 (2004) 092105.

### Low temperature elastic properties of PLZT relaxors

<u>G. SHABBIR</u><sup>1,2</sup> and S. KOJIMA<sup>1</sup>

### <sup>1</sup>Institute of Materials Science, University of Tsukuba, Japan <sup>2</sup>PINSTECH. Islamabad. Pakistan

Relaxor ferroelectrics belong to the class of materials that show strong chemical disorder. This leads to a low temperature state with frozen-in polarization devoid of ferroelectric long-range order. Many experimental evidences show that relaxors resemble glassy materials as they follow Vogel-Fulcher like freezing and significant 'aging' effect.

To illustrate glassy nature of relaxors high-resolution Brillouin light scattering experiments have been performed to investigate the low temperature 'aging' in the lanthanum lead zirconate titanate (PLZT) relaxor ferroelectric ceramics. From the temperature dependence of the frequency shift of longitudinal acoustic mode, an acoustic anomaly was observed at  $T_{o}$ ~46 K in these ceramics with clear 'aging' effect below  $T_{o}$ . The results have been discussed in the light of stepwise freezing of the related degrees of freedom causing transition from proper to improper dipole glassy states at  $T_{o}$ .

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