I Lithuanian-Ukrainian-Polish Meeting on Ferroelectrics Physics

PROGRAMME & ABSTRACTS

12–16 September, 2010
Vilnius, Lithuania

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Programme

Quick Timetable

	Sunday	Monday	Tuesday	Wednesday	Thursday
9:00-9:15		Opening	O-3 B.Konieczna	O-14 A.Molak	O-20 B.Andriyevsky
9:15-9:30		I-1 Z.Czapla			
9:30-9:45			O-4 L.Kozielski	O-15 S.Miga	O-21 T.Marciniszyn
9:45-10:00					I-14 J. A.Eiras
10:00-10:15		I-2 M.D.Glinchuk	O-5 A.Džiaugys	O-16 E.E.Tornau	
10:15-10:30					
10:30-10:45			Coffee Break	Coffee Break	Award of young scientists
10:45-11:00		Coffee Break	I-11 Yu.Vysochanskii	O-17 S.Waplak	Closing
11:00-11:15		I-3 S.Wada			
11:15-11:30				O-18 B.Fugiel	
11:30-11:45			O-6 M.Dunce		
11:45-12:00		I-4 M.Polomska			
12:00-12:15			O-7 O.V.Malyshkina		
12:15-12:30		I-5 R.Vlokh		Excursion & Lunch	
12:30-12:45			I-12 B.Garbarz-Glos		
12:45-13:00		I-6 J.Petzelt			
13:00-13:15					
13:15-14:30		Lunch	Lunch		
14:30-14:45		I-7 W.Kleemann	O-8 A.Orliukas		
14:45-15:00					
15:00-15:15			I-13 S.Sorokov		
15:15-15:30		I-8 B.Hilczer			
15:30-15:45			O-9 M.P.Trubitsyn		
15:45-16:00		O-1 N.A.Korynevskii			
16:00-16:15	Registration		Coffee Break		
16:15-16:30		Coffee Break	O-10 J.Macutkevič		
16:30-16:45		I-9 A.Grabar			
16:45-17:00			O-11 W.Zapart		
17:00-17:15		O-2 A.Kania			
17:15-17:30			O-12 G.Völkel		
17:30-17:45		I- 10 J.Dec			
17:45-18:00			O-13 V.Samulionis		
18:00-20:00	Get – together party	Poster Session I	Poster Session II	Conference Dinner	
20:00-21:00					

Sunday, September 12, 2010

18:00 – 21:00 ***Get – together party***

Monday, September 13, 2010

Morning Sessions 9:00 – 11:00

8:00 – 9:00 ***Breakfast***

9:00 – 9:15 ***Meeting opening:***
J.Grigas, Z.Czapla, M.D.Glinchuk

SESSION I

Chaired by J.Grigas

9:15 – 10:00 Z.Czapla[1,2], A.Rokosa[1], S.Dacko, B.Kosturek[1].
Successive phase transitions in $(Gua)_4SO_4Cl_2$ crystal: dielectric, pyroelectric, optical and dilatometric studies
[1]Institute of Experimental Physics, University of Wroclaw, Poland
[2]Dept. of Physics, Opole University of Technology, Poland

10:00 – 10:45 M.D.Glinchuk[1], A.N.Morozovska[2], E.A.Eliseev[1], R.Blinc[3]
Prediction of size-induced ferroelectricity in BaO nanowires
[1]Inst. for Problems of Materials Science, Kiev, Ukraine
[2]V.Lashkarev Inst. of Semiconductor Physics, Kiev, Ukraine
[3]Jožef Stefan Inst., Ljubliana, Slovenia

10:45 – 11:00 ***Coffee Break***

Monday, September 13, 2010

Morning Sessions 11:00 – 14:00

SESSION II

Chaired by Z.Czapla

11:00 – 11:45 S.Wada[1], R.Mitsui, K.Yamato[1], P.Pulpan[1], I.Fuji[1], C.Moriyoshi[2], Y.Kuroiwa[2]
Piezoelectric properties of Barium titanate – Bismuth perovskite-type oxide system ceramics with microstructure control
[1]Material Science and Technology, Univ. of Yamanashi, Japan
[2]Dept. of Physics Science, Hiroshima University, Japan

11:45 – 12:15 M.Polomska[1], B.Hilczer[1], J.Wolak[1], A.Pawlowski[1], L.F.Kirpichnikova[2]
The role of ferroelasticity in the mechanism of superprotonic phase transitions
[1]Inst. of Molecular Physics, Polish Academy of Sciences, Poznan, Poland
[2]Inst. of Crystallography, Russian Academy of Sciences, Moscow, Russia

12:15– 12: 45 O.Mys[1], B.Zapeka[1], A.Say[1], I.Martynyuk-Lototska[1], R.Vlokh[1], A.A.Grabar[2], Yu.M.Vysochanskii[2]
Optical properties and new peculiarities of phase transition in $Sn_2P_2(Se_xS_{1-x})_6$ crystals
[1]Inst. of Physical Optics, Lviv, Ukraine
[2]Inst. for Solid State Physics and Chemistry, Uzhgorod Univ., Ukraine

12:45 – 13:15 T.Ostapchuk, J.Petzelt, J.Hlinka, V.Bovtun, P.Kuzel, M.Savinov
Ferroelectric phase transition dynamics in the $BaTiO_3$ – $SrTiO_3$ system by dielectric spectroscopy
Inst. of Physics, Acad. Sci. Czech Rep., Praha, Czech Republic

13:15 – 14:30 ***Lunch***

Monday, September 13, 2010

Afternoon Sessions 14:30 – 16:30

SESSION III

Chaired by Yu.M.Vysochanskii

14:30 – 15:15 W.Kleemann
Magnetoelectricity in disordered multiferroics
Angewandte Physik, Universität Duisburg-Essen, Germany

15:15 – 15:45 B.Hilczer[1], E.Markiewicz[1], K.Pogorzelec-Glaser[1], M.Polomska[1], A.Pietraszko[2]
Dielectric relaxation in confined ferroelectric polymer
[1]Inst. of Molecular Physics, Polish Acad. Sci., Poznan, Poland
[2]Inst. of Low Temperatures and Structure Research, Polish Acad. Sci., Wroclaw, Poland

15:45 – 16:15 N.A.Korynevskii[1,2], V.B.Solovyan[1]
Thermodynamic functions of hydrogen bonded ferro-antiferroelectric mixed compounds
[1]Inst. for Condensed Matter Physics, National Acad. Sci. Ukraine, Lviv
[2]Inst. of Physics, Univ. of Szczecin, Poland

16:15 – 16:30 ***Coffee Break***

Monday, September 13, 2010

Afternoon Sessions 16:30 – 20:00

SESSION IV

Chaired by B.Hilczer

16:30 – 17:00 A.Grabar[1], P.Mathey[1], G.Gadret[1], I.Stoika[1], Yu.Vysochanskii[1]
Photochromic effects in photorefractive $Sn_2P_2S_6$ crystals
[1]Uzhgorod National Univ., Uzhgorod, Ukraine
[2]Inst. Carnot de Bourgogne, Univ. de Bourgogne, Dijon, France

17:00 – 17:30 A.Kania[1], S.Miga[1], J.Dec[2]
Nonlinear dielectric response of $AgNb_{1-x}Ta_xO_3$ solid solutions
[1] Inst. of Physics, Univ. of Silesia, Katowice, Poland
[2] Inst. of Materials Science, Univ. of Silesia, Katowice, Poland

17:30 – 18:00 J.Dec[1], S.Miga[2], W.Kleemann[3]
Ferroelectric phase transitions viewed via nonlinear dielectric response
[1] Inst. of Materials Science, Univ. of Silesia, Katowice, Poland
[21] Inst. of Physics, Univ. of Silesia, Katowice, Poland
[3] Angewandte Physik, Universität Duisburg-Essen, Germany

18:00 – 20:00 **Poster Session** I *Chaired by* J.Banys
A.Molak
N.A.Korynevskii

Tuesday, September 14, 2010

Morning Sessions 9:00 – 10:45

8:00 – 9:00 ***Breakfast***

SESSION V

Chaired by A.Kania

9:00 – 9:30 B.Konieczna[1], K.Biedrzycki[2], H.W.Janus[2], L.Markowski[2]
Low density plasma-assisted electron and ion emission at the phase transition of LATGS single crystal
[1] Institute of Experimental Physics, University of Wroclaw, Poland
[2] Inst. of Physics, Opole Univ. of Technology, Opole, Poland

9:30 – 10:00 L.Kozielski[1], M.Adamczyk[2], J.Erhart[3]
PLZT-based light driven piezoelectric transformer
[1] Inst. of Materials Science, Univ. of Silesia, Katowice, Poland
[2] Inst of Physics, Univ. of Silesia, Katowice, Poland
[3] Technical Univ. of Liberec, Czech Republik

10:00 – 10:30 A.Dziaugys[1], J.Banys[1], Yu.Vysochanskii[2], W.Kleemann[3], V.Shvartsman[3]
Dielectric spectroscopy of the mixed $CuIn_xCr_{1-x}P_2S_6$ crystals
[1] Faculty of Physics, Vilnius University, Lithuania
[2] Institute of Solid State Physics and Chemistry, Uzhgorod Univ., Ukraine
[3] Faculty of physics, Duisburg-Essen University, Germany

10:30 – 10:45 ***Coffee Break***

Tuesday, September 14, 2010

Morning Sessions 10:45– 14:30

SESSION VI

Chaired by W.Kleemann

10:45 – 11:30 Yu.Vysochanskii, K.Glukhov, M.Maior. K.Fedyo, A.Kohutych, V.Betsa, I.Prits, M.Gurzan
Ferroelectric and semiconductive properties of $Sn_2P_2S_6$ crystals with intrinsic vacancies
Uzhgorod University, Uzhgorod, Ukraine

11:30 – 12:00 M.Dunce, E.Birks, M.Antonova, K.Kalninsh, M.Kundzinsh
Structure and physical properties of $Na_{1/2}Bi_{1/2}TiO_3$-$CdTiO_3$solid solutions
Inst. of Solid State Physics, Univ. of Latvia, Riga, Latvia

12:00 – 12:30 O.V.Malyshkina[1], A.A.Movchikova[1], K.Penzov[1], M.Shashkov[1], R.Steinhausen[2], H.T.Langhammer[2], H.Beige[2]
The polarization distribution in graded BTS ceramics
[1]Tver State University, Tver, Russia
[2]Martin-Luther-University Halle-Wittenberg, Halle, Germany

12:30 – 13:15 B.Garbarz-Glos[1], K.Bormanis[2], D.Sitko[1]
Effect of Zr^{+4} doping on the electrical properties of $BaTiO_3$ ceramics
[1]Inst. of Physics, Pedagogical Univ., Krakow, Poland
[2]Inst. of Solid State Physics, Univ. of Latvia, Riga, Latvia

13:15- 14:30 ***Lunch***

Tuesday, September 14, 2010

Afternoon Sessions 14:30– 16:15

SESSION VII

Chaired by S.Waplak

14:30 – 15:00 A.F.Orliukas[1], T.Šalkus[1], A.Kežionis[1], A.Dindune[2], Z.Kanepe[2], J.Ronis[2], O.Bohnke[3], V.Venckutė[1], M.Lelis[4]
Structure and electrical properties of $Li_{3-x}Sc_{2-x}Zr_x(PO_4)_3$ (x=0; 0.1; 0.2) ceramics
[1]Faculty of Physics, Vilnius Univ., Lithuania
[2]Inst. of Inorganic Chemistry, Riga Technical Univ., Latvia
[3]Laboratoire des Fluorines, Univ. du maine, France
[4]Lithuanian Energy Inst., Kaunas, Lithuania

15:00 – 15:30 S.Sorokov, R.Levitskii, A.Vdovych
Cluster Theory Of $Rb_{1-x}(NH_4)_xH_2PO_4$ Type Mixed Crystals. Problems and Outlook
Institute for Condensed Matter Physics, National Academy of Sciences of Ukraine

15:30 – 16:00 M.D.Volnianskii, M.P.Trubitsyn, Y.A.H.Obaidat
Relaxation phenomena in $Li_2Ge_7O_{15}$ crystal
Dept. of Solid State Physics and Optoelectronics, Oles Gonchar Nat. Univ. of Dnepropetrovsk, Ukraine

16:00 – 16:15 ***Coffee Break***

Tuesday, September 14, 2010

Afternoon Sessions 16:15– 20:00

SESSION VIII

Chaired by B.Garbarz-Glos

16:15 – 16:45 J.Macutkevič[1], J.Banys[2], R.Grigalaitis[2], A.Mikonis[2], R.Adomavičius[1], A.Krotkus[1]
Soft mode investigations in ferroelectrics and relaxors by time domain THz spectroscopy
[1]Semiconductor Physics Inst., Center for Physical Sciences and Technology, Vilnius, Lithuania
[2]Dept. of Radiophysics, Vilnius Univ., Vilnius, Lithuania

16:45 – 17:15 W.Zapart[1], M.B.Zapart[1], R.Kowalczyk[1], K.Maternicki[1], M.Maczka[2]
Ferroelastic phase transitions in mixed $KSc(MoO_4)_2$ based trigonal double molybdates
[1]Inst. of Physics, Technical Univ. of Czestochowa, Poland
[2]Inst. of Low Temperatures and structure Research, Polish Acad. Sci., Wroclaw, Poland

17:15 – 17:45 G.Völkel
Order-disorder phenomena in the low-temperature phase of $BaTiO_3$
Zickra 1, D-07955 Auma, Germany

17:45 – 18:00 V.Samulionis, J.Banys and Yu.Vysochanskii*
Ultrasonic investigation of piezoelectric sensitivity in $CuInP_2S_6$ family layered crystals
Physics Faculty, Vilnius University, Lithuania,
**Institute of Solid State Physics and Chemistry, Uzhgorod Univ., Ukraine*

18:00 – 20:00 ***Poster Session** II* *Chaired by* E.Tornau
J.Dec
M.P.Trubitsyn

Wednesday, September 15, 2010

Morning Sessions 9:00 – 10:45

8:00 – 9:00 **Breakfast**

SESSION IX

Chaired by A.Grabar

9:00 – 9:30 A.Molak[1], K.Lawniczak-Jablonska[2], P.Nachimuthu[3], R.C.C.Perera[3]
The estimation of the Mn atoms chemical bonding in $(Na_{(1-x)}Bi_xNb_{(1-y)}Mn_y)O_3$ ceramics and changeover in the electrical properties
[1]Inst. of Physics, Univ. of Silesia, Katowice, Poland
[2]Inst. of Physics, Polish Acad. Sci., Warszawa, Poland
[3]Center for X-ray Optics, Lawrence Berkley Nat. Lab., Berkley, USA

9:30 – 10:00 S.Miga[1], Z.Czapla[2,3], W.Kleemann[4], J.Dec[5]
Nonlinear dielectric response of Rochelle salt
[1]Inst. of Physics, Univ. of Silesia, Katowice, Poland
[2]Dept. of Physics, Opole Univ. of Technology, Opole, Poland
[3]Institute of Experimental Physics, University of Wroclaw, Poland
[4]Angewandte Physik, Universität Duisburg-Essen, Germany
[5]Inst. of Materials Science, Univ. of Silesia, Katowice, Poland

10:00 – 10:30 E.E.Tornau
Formation of stripe structures with and without longer-range interactions
Semiconductor Physics Inst., Center for Physical Sciences and Technology, Vilnius, Lithuania

10:30 – 10:45 ***Coffee Break***

Wednesday, September 15, 2010

Morning Sessions 10:45 – 12:45

SESSION X

Chaired by S.Wada

10:45 – 11:15 S.Waplak, W.Jurga
Size-effect in ferroelectric TGS, TGSe crystals under high hydrostatic pressure
Inst. of Molecular Physics, Polish Acad. Sci., Poznan, Poland

11:15 – 11:45 B.Fugiel
Some less known aspects of the para-ferroelectric phase transition in ferroelectrics with hydrogen bonds
Inst. of Physics, Univ. of Silesia, Katowice, Poland

Wednesday, September 15, 2010

12:15 – 17:30 ***Excursion & Lunch***

18:00 – 21:00 ***Conference Dinner***

Thursday, September 16, 2010

Morning Sessions 9:00 – 11:00

8:00 – 9:00 ***Breakfast***

SESSION XI

Chaired by J.Banys

09:00 – 09:30 B.Andriyevsky[1], A.Patryn[1], C.Cobet[2], J.Przeslawski[3],
B.Kosturek[3], N.Esser[2], D.Dorywalski[1]
Electronic properties of KDP and DKDP crystals: *ab-initio* calculations and spectral ellipsometry experiment
[1]Faculty of Electronics and Computer Sciences, Koszalin Univ. of Technology, Poland
[2]Inst. for Analytical Sciences, Dept. Berlin, Germany
[3]Institute of Experimental Physics, University of Wroclaw, Poland

9:30 – 09:45 T.Marciniszyn, R.Poprawski
Physical properties and phase transitions in ADP embedded into the porous glasses
Institute of Physics, Wrocław University of Technology, Wrocław, Poland

9:45 – 10:30 J.A.Eiras
Ferroelectric and ferroelastic contributions to the dielectric response of ferroelectrics
Universidade Federal de São Carlos – Depto. Física – Grupo de Cerâmicas Ferroelétricas, BRASIL

10:30 – 10:45 ***Award of young scientists***

10:45 – 11:00 ***Closing of the Meeting***

POSTER SESSION I

Monday 18:00–20:00

P-1 J.Pozingis, J.Banys, J.Macutkevic, R.Adomavicius, A.Krotkus, D.C. Lupascu
BROADBAND DIELECTRIC SPECTROSCOPY OF FERROELECTRIC $(1\text{-}x)Ag_{0.9}Li_{0.1}NbO_3 - xBi_{0,5}K_{0,5}TiO_3$ CERAMICS

P-2 B.Wodecka-Duś, D.Czekaj
ELECTRIC PROPERTIES OF La^{3+} DOPED BARIUM TITANATE CERAMICS

P-3 K.Osińska, J.Dzik, H.Bernard, B.Wodecka-Duś, D.Czekaj
SYNTHESIS AND FABRICATION OF CERAMIC 0-3 COMPOSITE MATERIAL FOR THERMO-RESISTIVITY APPLICATION

P-4 H.Bernard, J.Dzik, K.Osińska, A.Lisińska-Czekaj, D.Czekaj
La-DOPING EFFECTS ON DIELECTRIC PROPERTIES OF $Bi_4Ti_3O_{12}$ CERAMICS

P-5 J.Dzik, H.Bernard, K.Osińska, A.Lisińska-Czekaj, D.Czekaj
IMPEDANCE SPECTROSCOPIC STUDIES OF BISMUTH FERRITE

P-6 B.Konieczna, K.Biedrzycki, H.W.Janus, L.Markowski
PECULIARITY OF LOW DENSITY PLASMA-ASSISTED CHARGE EMISSION PRODUCED AT LATGS CRYSTAL SURFACE

P-7 Z.Czapla, Yu.Eliyashevskyy, S.Dacko
RELAXATION OF PERMITTIVITY IN $[(CH_3)_2NH_2]_3CuCl_5$ CRYSTAL IN EXTERNAL ELECTRIC FIELD

P-8 I.E.Lipiński, N.A.Korynevskii, M.Soboń
PRESURE EFECT ON THE ANOMAL BEHAVIOUR OF PHYSICAL PROPERTIES OF SASD TYPE CRYSTALS

P-9 B.Andriyevsky, K.Dorywalski, I.Kityk, M.Piasecki, T.Łukasiewicz, A.Majchrowski, A.Patryn, J.Dec, C.Cobet, N.Esser
SPECTRAL ELLIPSOMETRY STUDY OF SBN SINGLE CRYSTALS IN VISIBLE AND ULTRAVIOLET REGION

P-10 B.Andriyevsky, M.Jaskólski
BAND STRUCTURE AND OPTICAL CHARACTERISTICS OF TDA CRYSTALS

P-11 J.Suchanicz, K.Pytel, K.Konieczny, M.Sokolowski, A.Finder, M.Antonova, A.Sternberg
INFLUENCE OF ELECTROMECHANICAL FIELDS ON DIELECTRIC PROPERTIES OF PLZT−x/65/35 CERAMICS (x=6 AND 7)

P-12 M. Jankowski
DIELECTRIC NONLINEARITY IN $(NH_4)_3H(SeO_4)_2$ CRYSTAL

P-13 M.Gabryś, C.Kajtoch, W.Bąk, B.Handke, F.Starzyk
INFLUENCE OF Sr-SUBSTITUTION ON DIFFUSENESS OF $(Ba_{1\text{-}x}Sr_x)TiO_3$ PHASE TRANSITIONS

P-14 B.Staskiewicz, S.Dacko, Z.Czapla
DILATOMETRIC AND DIELECTRIC INVESTIGATIONS OF PHASE TRANSITIONS IN $[(CH_3)_2CNH_3]_4Cd_3Cl_{10}$ CRYSTAL

P-15 M.B.Zapart, W.Zapart, P. Czaja, T.Mila and J.Solecki
EPR SPECTROSCOPY AND FERROELASTIC DOMAIN STRUCTURE IN THE $LiRbSO_4$-$LiCsSO_4$ SYSTEM

P-16 R. Skulski, D. Bochenek, P. Wawrzała
TECHNOLOGY AND MAIN PROPERTIES OF $(Pb_{0.75}Ba_{0.25})(Zr_{0.65}Ti_{0.35})_{1-z}Sn_z)O_3$ CERAMICS

P-17 D.Bochenek, R.Skulski
FERROELECTRIC PROPERTIES OF PBZTN CERAMICS

P-18 M.Palatnikov, O.Shcherbina, N.Sidorov, K.Bormanis
EFFECTS OF VTE TREATMENT ON COMPOSITION OF LITHIUM TANTALATE SINGLE CRYSTALS

P-19 K.Bormanis, A.I.Vaingolts, A.I.Burkhanov, and A.Kalvane
DIELECTRIC RESPONSE IN LEAD FERROTANTALATE WITH RESPECT TO BIAS FIELD EFFECT

P-20 Śmiga, B.Garbarz-Glos, M.Livinsh, A.Kalvane
STRUCTURAL AND ELECTRIC PROPERTIES OF SODIUM LITHIUM NIOBATE CERAMIC SOLID SOLUTION $Li_{0.08}Na_{0.92}NbO_3$

P-21 J.Suchanicz, G.Stopa, K.Pytel, K.Konieczny, A.Finder, M.Antonova, A.Sternberg
ELECTRICAL TRANSPORT PROPERTIES OF LEAD−FREE $(Na_{0.5}Bi_{0.5})_{(1-x)}Ba_xZr_{0.04}Ti_{0.96}O_3$ CERAMICS (x=0, 0.06, 0.085, 0.1)

P-22 K.Konieczny, W.Śmiga
STRUCTURAL, PYROELECTRIC AND ELECTRIC PROPERTIES OF $Na_{1-x}Li_xNbO_3$ CERAMICS

P-23 P.Konsin, B.Sorkin
MAGNETOVIBRONIC THEORY OF $BiFeO_3$-TYPE MULTIFERROICS

P-24 R.Bujakiewicz-Korońska, Ł.Hetmańczyk, B.Garbarz-Glos, A.Budziak, J.Koroński, J.Hetmańczyk, M.Antonova, A.Kalvane
INVESTIGATIONS OF LOW TEMPERATURE PHASE TRANSITIONS IN $BiFeO_3$ CERAMIC BY INFRARED SPECTROSCOPY

P-25 R.Kowalczyk, M.B.Zapart, W.Zapart, I.Mroz
AFM AND OPTICAL MICROSCOPY STUDIES OF $KFe(MoO_4)_2$

P-26 M.Zdanowska-Frączek, Z.J.Frączek
THE PRESSURE INFLUENCE ON THE PROPERTIES OF FERROELECTRIC $(C_3N_5H_5)_5Bi_2Cl_{11}$

P-27 A.N.Morozovska, E.A. Eliseev, G.S.Svechnikov
NANODOMAIN FORMATION IN THE HETEROSTRUCTURE “SPM TIP ELECTRODE − THIN FERROELECTRIC FILM − SEMICONDUCTOR SUBSTRATE”

POSTER SESSION II

Tuesday 18:00–20:00

P-1 K.Wolska, K.Matyjasek, J.Subocz
THE TEMPERATURE DEPENDENCES OF THE FERROELECTRIC AND DIELECTRIC PROPERTIES OF PURE AND NICKEL DOPED SBN SINGLE CRYSTALS

P-2 A. Osak
HOPPING CONDUCTIVITY IN FERROELECTRIC $[Pb(Fe_{1/3}Sb_{2/3})Ti_yZr_z]O_3$

P-3 P.P.Guranich, R.R.Rosul, A.G.Slivka, O.O.Gomonnai, I.Yu.Roman
PRESSURE BEHAVIOUR OF THE BIREFRINGENCE OF IN $TlInS_2$ LAYERED CRYSTALS WITH INCOMMENSURATE STRUCTURE

P-4 K.Matyjasek, K.Wolska
MANIFESTATION OF STRUCTURAL DISORDER IN THE POLARIZATION KINETICS OF NON-UNIFORM FERROELECTRICS AND RELAXOR CRYSTALS

P-5 M.Adamczyk, M.Pawełczyk
INFLUENCE OF VANADIUM DOPANT ON DIELECTRIC PROPERTIES OF $BaBi_2Nb_2O_9$ CERAMICS

P-6 M.Adamczyk, L.Kozielski
IMPEDANCE SPECTROSCOPY IN $BaBi_2Nb_2O_9$ CERAMICS

P-7 E.Nogas-Ćwikiel
CERAMICS WITH TETRAGONAL TUNGSTEN BRONZE TYPE STRUCTURE FOR TEXTURED CERAMICS-POLYMER COMPOSITES

P-8 K.Ćwikiel
QUALITY OF TGS SAMPLES SURFACE INVESTIGATED BY AFM

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Abstracts

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Oral presentations

SUCCESIVE PHASE TRANSITIONS IN $(Gua)_4SO_4Cl_2$ CRYSTAL: DIELECTRIC, PYROELECTRIC, OPTICAL AND DILATOMETRIC STUDIES

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The crystal of $(Gua)_4SO_4Cl_2$ is room temperature ferroelectric crystal and undergoes successive phase transitions [1]. Our studies of dielectric and pyroelectric properties along the ferroelectric *a* - axes, optical observation along the *b*-axis and dilatometric measurements along the *a-, b-* and *c-* axes for of $(Gua)_4SO_4Cl_2$ crystal were performed in the temperature range 330-380 K covering the phase transitions region at various heating/cooling rates regime. At faster heating diffused dielectric anomaly in the range of phase transitions is observed. Slow heating (0.01 K/min) gives evidences for two successive phase transitions III-II at T_2 = 353 K and II-I at T_1 = 357,5 K. Pyroelectric studies evidenced ferroelectric properties of phase III and II. Disappearance of polarization is diffused at first heating run. The sample kept for l80 min in phase II undergoes direct sharp transition II-I during heating. Optical observations on heating run at the rate 0.01 K/min showed the successive phase transitions with the symmetry changes according to the scheme: orthorhombic(III) - orthorhombic(II) - tetragonal (I) and on cooling: tetragonal (I) - orthorhombic (II). Dilatometric studies at slow heating/cooling runs confirmed successive phase transitions. The dielectric, pyroelectric, dilatometric and optical studies allowed to conclude that the phase transition III-II at T_2 is characterized by slow kinetics. The studies of permittivity and dimensions changes of the samples which were kept at constant temperature inside of phase II during 180 min gave some detailed information about kinetics of III-II phase transition.

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PREDICTION OF SIZE-INDUCED OF FERROELECTRICITY IN BaO NANOWIRES

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We predict that a ferroelectric phase can be induced by a strong intrinsic surface stress $\delta \sim \mu/R$ inevitably present under the curved surface due to intrinsic stress μ in high aspect ratio cylindrical nanoparticles of nonferroelectric binary oxides (BaO, EuO, MgO, etc).

We calculated the sizes and temperature range of the ferroelectric phase in BaO nanowires. The analytical calculations were performed within Landau-Ginzburg-Devonshire theory with phenomenological parameters extracted from tabulated experimental data. In accordance with our calculations BaO nanowires of radius ~ (1−10) nm can be ferroelectric at room temperature (with spontaneous polarization values up to 0.5 C/m^2) for the typical surface stress coefficients ~ (10–50) N/m.

We hope that our prediction can stimulate both experimental studies of rocksalt binary oxides nanoparticles polar properties as well as the first principle calculations of their spontaneous dipole moment induced by the intrinsic stress under the curved surface. The same phenomena could lead to high temperature magnetoelectric effect in nanosystems of magnetic binary oxides.

PIEZOELRCTRIC PROPERTIES OF BARIUM TITANATE – BISMUTH PEROVSKITE-TYPE OXIDE SYSTEM CERAMICS WITH MICROSTRUCTURE CONTROL

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Barium titanate ($BaTiO_3$, BT), and bismuth titanate magnesium oxide ($Bi(Mg_{0.5}Ti_{0.5})O_3$, BMT) solid solution system ceramics were prepared by conventional sintering method in pursuit of the enhancement of the BT Curie temperature (T_C, 132 ˚C). Below BMT contents of 70 molar%, the BT-BMT ceramics with high density over 94 % were prepared in atmosphere despite BMT contents, and the formation of single-phase perovskite-type compounds was confirmed by X-ray diffraction (XRD) measurement. Using these ceramics, ferroelectric properties were measured. Below 10 molar% of BMT, the Tc of BT-BMT ceramics decreased from 132 ˚C to room temperature with increasing BMT content from 0 to 10 molar%, while above 10 molar% of BMT, the Tc of BT-BMT ceramics increased from room temperature to 360 ˚C with increasing BMT content from 10 to 50 molar%. On the other hand, above 50 molar% of BMT, the Tc of BT-BMT ceramics decreased from 360 to 290 ˚C with increasing BMT content from 50 to 70 molar%. Normal ferroelectric polarization vs. electric-field (P-E) hysteresis loops were observed for BT-BMT ceramics with BMT contents below 20 and above 60 molar%. On the other hand, broad P-E double hysteresis loops were observed for BMT contents from 30 to 50 molar%. The origin was investigated using synchrotron XRD measurement and Rietveld analysis. The crystal structure was assigned to ferroelectric phase with domain-pinning by certain defect structures. A modified phase diagram was proposed on the basis of the temperature dependence of the crystal structure.

THE ROLE OF FERROELASTICITY IN THE MECHANISM OF SUPERPROTONIC PHASE TRANSITIONS

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Solid acids such as those which belong to a family with general formula $M_xLi_yH_z(XO_4)_{(x+y+z)/2}$ where M=Rb, NH_4,Cs,K; X=S,Se are characterized by the fact that the XO_4 groups bound by hydrogen bonds form either chains, dimers, trimers or tetramers, specifically ordered in the structure depending on the crystal groups. Depending on x,y and z, the $M_xLi_yH_z(XO_4)_{(x+y+z)/2}$ family can be divided into the following groups of crystals: $MH(XO_4)$, where XO_4 groups linked by hydrogen bonds form chains, in the $M_3H(XO_4)_2$ group of crystals, we are dealing with isolated $H(XO_4)_2$ dimers. In the $M_4H_2(XO_4)_3$ group XO_4 tetrahedra are bound by hydrogen bonds to the trimers, and in the $M_4LiH_3(XO_4)_4$ the tetramers $H_3(XO_4)_4$ exist in the crystal structure. A variety of possible orderings of the tetrahedral groups XO_4 bound by hydrogen bonds lead to different long range orderings in the crystals which belong to the the $M_xLi_yH_z(XO_4)_{(x+y+z)/2}$ family like ferroelasticity or ferroelectricity and consequently to variety of phase transitions.

The common feature of all members of the family is that they undergo transitions to superionic phase, which is characterized by high proton conductivity. On passing through the transition, conductivity jumps by several orders of magnitude to a value of 10^{-2}-10^{-3} $\Omega^{-1}cm^{-1}$. The superprotonic phases are characterized by high structural disorder, which consists of translational disorder of protons, of orientational and translational disorder of XO_4 tetrahedra and in the case of ammonium containing crystals of orientational disorder of NH_4 groups.

The crystals have attracted the attention of researchers for a number of years. For many members of the family, the transition to superprotonic phase at $T=T_S$ is simultaneously a ferroelastic-paraelastic one. However, this does not apply to all crystals of the family. Therefore, a question arises about the role of the phenomenon of ferroelasticity in the superprotonic phase transition It was shown in our studies, that ferroelasticity plays a very important role in the mechanism of phase transition to the superprotonic state.

Present work reports results of our Raman scattering, ferroelastic domains and electrical conductivity studies of some crystals the representatives of $M_xLi_yH_z(XO_4)_{(x+y+z)/2}$ family which exhibit simultaneously superprotonic-ferroelastic phase transition as well as those for which ferroelasticity does not appear.

OPTICAL PROPERTIES AND NEW PECULIARITIES OF PHASE TRANSITION IN $Sn_2P_2(Se_XS_{1-X})_6$ CRYSTALS

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Present report is devoted to the consideration and analysing of the results of studying of optical (optical activity, birefringence, acoustooptic electrooptic, magnetooptic) and acoustical (ultrasonic wave velocities, elastic stiffness and compliances tensors, oblique of acoustic waves) and dilatometric properties of $Sn_2P_2(Se_xS_{1-x})_6$ (x=0; 0.28) crystals in the course of ferroelectric phase transition and at hydrostatic pressure.

It has been shown that $Sn_2P_2S_6$ crystals possess high figures of merit as electrooptic ($n_2^3r_{21} - n_3^3r_{31} = (1.23 \pm 0.12)\times 10^{-9}\,\mathrm{m/V}$) magnetooptic (the Verdet constant is equal to $512.7\ \mathrm{T^{-1}m^{-1}}$) and acoustooptic ($\mathrm{M_2} = (1.7 \pm 0.4)\times 10^{-12}\ \mathrm{s^3/kg}$) material. The acoustooptic figure of merit of the $Sn_2P_2(Se_{0.28}S_{0.72})_6$ is estimated to be also quite high $\approx 2\times 10^{-12}\ \mathrm{s^3/kg}$. The electrogyration coefficient in the $Sn_2P_2S_6$ crystals under test is equal to $\approx 10^{-10}\,\mathrm{m/V}$, whereas for the most of compounds studied previously it is of the order of $\approx 10^{-12}\,\mathrm{m/V}$.

The new peculiarities of the phase transitions and phase p,T and x,T diagrams of $Sn_2P_2(Se_xS_{1-x})_6$ crystals are discussed on the basis of obtained results.

FERROELECTRIC PHASE TRANSITION DYNAMICS IN THE $BaTiO_3$-$SrTiO_3$ SYSTEM BY DIELECTRIC SPECTROSCOPY

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Understanding the ferroelectric phase transition dynamics even in such a prototype ferroelectric solid solution as $Ba_xSr_{1-x}TiO_3$ (BST-x) still remains a challenging problem, which should be in the best way addressed by a careful study of the dielectric dispersion in the wide frequency and temperature range. Here we discuss our recent results on the study of high-density BST ceramics with $x = 0.1$, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, 1.0 and large average grain size (20-50 μm) performed by means of Fourier transform infrared (FTIR) reflectivity and time-domain terahertz (THz) transmission spectroscopy in the 20-900 K temperature range, complemented by high-frequency (HF) and microwave (MW) coaxial impedance measurements at 100÷500 K for x = 0.3, 0.6 and 1.0 and standard capacitance low-frequency (LF) dielectric spectroscopy. The results are compared and discussed using the published data on pure $SrTiO_3$ (STO) and $BaTiO_3$ (BTO) single crystals.

Our study demonstrates that far above the ferroelectric phase transition, the temperature change of the static permittivity in BST is completely accounted for by the soft-mode contribution, but in a certain temperature range $\Delta T_{CM}(x)$ above T_C an overdamped central-mode-type excitation appears below ~1 THz which takes over the softening on approaching T_C due to a coupling with the soft mode [1]. The $\Delta T_{CM}(x)$ temperature range is the largest (~300 K) for the pure BTO and narrows on decreasing x. The central mode exists also in the tetragonal and orthorhombic ferroelectric phases but completely disappears in the lowest-temperature (fully ordered) rhombohedral phase. Qualitatively the same behaviour was obtained from the Monte-Carlo dielectric function simulations, based on the first-principle effective Hamiltonian approach, which confirms the intrinsic lattice dynamic origin of the central mode in BST [2-5].

Our MW data also indicate existence of a relaxational dispersion close to T_C even in the paraelectric phase arising in the GHz range. This relaxation contributes of about half of the maximum value of the static permittivity at T_C and weakens on heating. In pure BTO, where we have studied it more carefully also in annealed samples, it is sample dependent but appears even in single crystals. Its nature in a paraelectric phase is probably due to some inevitable defects like dislocations. These results allow us to understand the presence of local polar regions in the paraelectric phase of BST-x, whose temperature range extends on increasing x and were observed in other recent experiments on BTO [6,7].

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MAGNETOELECTRICITY IN DISORDERED MULTIFERROICS

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The revival of the magnetoelectric (ME) effect [1] has vitally been boosted by recent intensified research on multiferroic materials [2], which promise to maximise the ME efficiency. While the primordial bilinear ME effect requires stringent symmetry properties, higher order ME effects are less demanding. In particular the biquadratic ME effect has recently attracted growing interest, e.g. in ferroelectromagnetic RE manganites, where it is related to the magnetocapacitance or magnetodielectric effect. In disordered systems with broken translational symmetry it is even dominating, while ME effects of lower order may be absent. In type I multiferroics, where magnetic and electric ordering have different origins, it controls the exchange interaction via quadratic spin-lattice interaction.

We have measured nonlinear ME_E effects in various ‘disordered multiferroics’ using SQUID susceptometry [3] and interpret the EH^2- and E^2H^2-type magnetoelectric (ME) effect in terms of electric field or polarization controlled exchange coupling. In the quantum paraelectric $EuTiO_3$ additional electric field-induced Dzialoshiskii-Moriya interaction has to be involved in order to explain the occurrence of large non-diagonal 3rd order magnetoelectric susceptibility [4]. The magnetic relaxor $Pb(Fe_{0.5}Nb_{0.5})O_3$ reveals unique features of a dilute antiferromagnet with a spin glass component emerging at low temperatures and giving rise even to linear magnetoelectric susceptibility [5]. Finally, both polar and spin disorder drive the solid solution ceramic $Sr_{0.98}Mn_{0.02}TiO_3$ into a magnetoelectric multiglass state [6], while $K_{0.97}Mn_{0.03}TaO_3$ was classified a magnetoelectric spin cluster glass [7].

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DIELECTRIC RELAXATION IN CONFINED FERROELECTRIC POLYMER

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Increasing tendency of miniaturization in microelectronics and informatics has driven practical and theoretical interest in nanostructures of functional materials [1]. Among all attempts especially important is nanostructuring of functional polymers since they are attractive as storage media for non-volatile memories, because of flexibility, low cost, ease of production, and integrability with silicon technology and all-organic electronics [2 – 4].

The work was aimed at studies of dielectric response of poly(vinylidene fluoride) confined in Al_2O_3 discs with pores of mean diameter of 200 nm (Anopore 200, Whatman). Temperature variation of the complex permittivity ε*(T) was studied in temperature range 150-425 K with Novocontrol Broadband Dielectric Spectrometer with Quatro Cryosystem in the frequency range 100 Hz ≤ f ≤ 1 MHz. We found that the main effect of Anopore confinement on the dielectric response ε*(f, T) appears as a suppressing of the local mode (wide angle oscillation of the dipoles attached to the main chain followed by the rotation with main chain cooperation). Moreover, the dynamics of segmental motion in the amorphous phase of the semi-crystalline polymer is faster in the confinement compared to that of the film. Fig. 1 shows dielectric absorption of PVDF hot-pressed film and PVDF confined in Anopore disc at various temperatures.

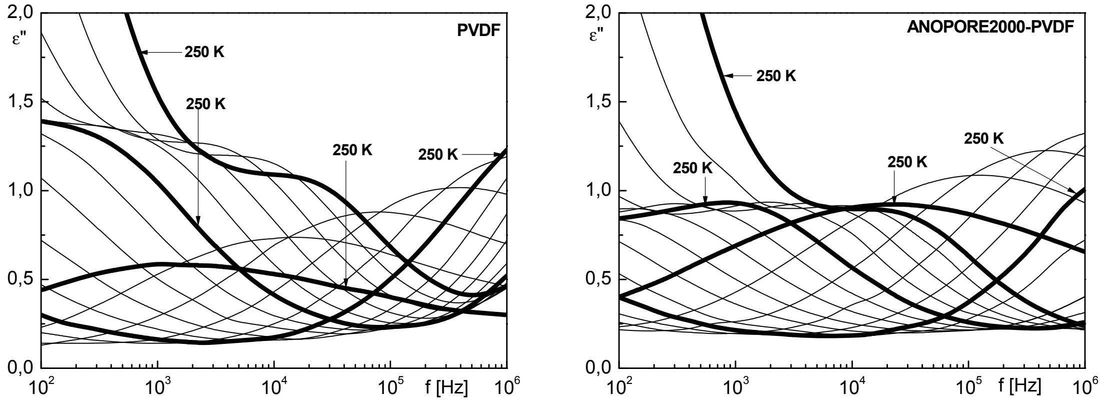

Fig. 1. Dielectric absorption of PVDF hot-pressed film and Anopore-PVDF composite

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THERMODYNAMIC FUNCTIONS OF HYDROGEN BONDED FERRO-ANTIFERROELECTRIC MIXED COMPOUNDS

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A new approach for a microscopic description of different phases including dipole glass phase in hydrogen bonded ferroelectric-antiferroelectric mixed compounds is suggested. It is based on the calculation and analysis of single and pair (nearest neighbours) correlation functions behaviour. Using the self-consistent cluster approximation, these functions were obtained in the second order of replica expansion. Due to chaotic distribution of different types of interparticle interactions the infinite number of solutions for pair correlation functions arises and the non-ergodic behaviour of frustrated ferroelectric-antiferroelectric mixed system is observed.

It has been theoretically proved that the ordered ferroelectric- and antiferroelectric phases exist only separately. Between percolation barriers for both types of long-range ordering at sufficiently low temperature the dipole glass state takes place. The obtained phase diagram [1] represents both the existence of the regions of pure ferroelectric, antiferroelectric, dipole glass phases and of the mixed ferroelectric-dipole glass, antiferroelectric-dipole glass phases. For RADA ($Rb_{1-n}(NH_4)_nH_2AsO_4$) this diagram is in very good coincidence with the experimental one [2].

Thermodynamic functions (order parameters, static dielectric susceptibility, heat capacity) for investigated system of RADA type were calculated and tested in a wide temperature and concentration regions. For ordered phases (ferroelectric and antiferroelectric) they behave in a usual way demonstrating (susceptibility and capacity) an unlimited divergence in a ferroelectric phase transition point. In the dipole glass phase the behaviour of the susceptibility and the capacity is close to their behaviour in a non-ordered medium, but with some peculiarities (jumps) caused by the internal structure of dipole glass phase.

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PHOTOCHROMIC EFFECTS IN PHOTOREFRACTIVE $Sn_2P_2S_6$ CRYSTALS

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The $Sn_2P_2S_6$ crystal is an efficient photorefractive material sensitive in red and near IR spectral region. Doping of these crystal by Te, Sb, Bi and other elements permits to enhance substantially photorefractive parameters [1-3]. Meantime all details of the light-induced and charge transfer processes in this material are not clarified, that makes important the studies of various photoinduced effects. One of the related phenomena is photochromic effect. Photoinduced absorption in $Sn_2P_2S_6$ was first observed at temperature 10 K [4], but a possible contribution of the amplitude gratings in the photorefractive effect was not considered in previous studies. The present study of the light-induced optical absorption in the nominally pure and doped $Sn_2P_2S_6$ crystals at room and lower temperatures were carried out with an aim to get the complementary data on the defect states in the gap and to estimate the possible impact of the photochromic effect on the formation the photorefractive gratings.

The variation of the optical absorption induced by light irradiation was measured in $Sn_2P_2S_6$ samples including nominally pure, Sb- and Te-doped compounds. A weak photochromic effect was found out in all the studied compounds even at room temperature, but the light-induced absorption substantially increases at low temperatures where typical variation of the absorption coefficient $\Delta\alpha$ changes at wavelength ~600-700 nm from about ~0.2 cm^{-1} up to 2-3 cm^{-1} after intense illumination by a green light (532 nm). The induced absorption at low temperatures relaxes rather slowly at constant temperature, but is decreasing with heating. The curves of the thermo-induced relaxation of the absorption at relatively fast linear heating (~3 K/min) were measured in differently doped samples, and indicate on the presence of several deep-level defects in the gap connected with the type of impurity.

The holographic experiments were carried out using He-Ne laser irradiation (633 nm, 60 mW) in the $Sn_2P_2S_6$:Sb2% samples with highest two-wave mixing gain Γ at room temperature. The measurements were carried out in two regimes: a) measuring of the two-wave mixing gain, when weak signal beam was amplified by intense pump beam due to energy transfer, and b) measurements of the diffraction efficiency of light- induced grating in self-reading regime after writing by two coherent beams with equalized intensities after exposure time ~1 min. It was found that at low temperatures (120 -150 K) two-wave mixing measured in regime "a" is completely absent, but the hologram with the max diffraction efficiency ~1-2% and relaxation time about 10^2 sec is obtained in regime "b". On the contrary, at temperatures above 250 K the predominantly diffusion-type phase grating is formed. At intermediate temperatures (150-250 K) in the regime "a" the intensive regular oscillations of the signal beam are observed, that obviously occurs due to competition between the photorefractive (phase, 90-deg shifted) grating and the amplitude (local) grating. These oscillations are studied versus the light intensity and grating spacing at various temperatures.

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NONLINEAR DIELECTRIC RESPONSE OF $AgNb_{1-x}Ta_xO_3$ SOLID SOLUTIONS

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Silver niobate-tantalate $AgNb_{1-x}Ta_xO_3$ (ATN) ceramics ($0.46 \le x \le 0.6$) are very promising high permittivity microwave materials. Measurements performed in the 1 GHz region showed $\varepsilon = 375$, $Q \cdot f = 860$ GHz and $\Delta f/f = 0 - 0.02$ (-40 – 60 °C) [1]. Contribution of the sub-millimetre relaxational mode, connected with Nb/Ta ion dynamics, to dielectric susceptibility is responsible for these microwave properties as well as the appearance of diffuse $\varepsilon(T)$ maximum near the M_2-M_3 phase transition [2]. In ATN ($x < 0.94$) a sequence of phase transitions between the weak ferroelectric M_1, disordered antiferroelectric M_2 and M_3, and paraelectric O_1, O_2, T and C phases occurs. The diffuse M_1-M_2 and M_2-M_3 phase transitions are connected with the Nb/Ta displacement sublattice. In principle, the temperature dependence of dielectric nonlinearity coefficient a_3 may provide an important information on nature of appearing polar states.

Linear and nonlinear dielectric studies of ATN ceramics and crystals ($0 \le x \le 0.6$) were carried out in the temperature range: 80 – 680 K. The temperature dependences of χ_3 and a_3 exhibit two characteristic maxima at M_1-M_2 transition and at temperature of Nb/Ta dynamics freezing $T_{f.}$ [3]. T_f lies approximately 70 K below M_2-M_3 maximum. Both maxima shift towards low temperatures with the increase in Ta concentration (5 K/%Ta). Basing on the results of these studies the type of polar order of the particular M phases is proposed and the contribution of the Nb/Ta ion dynamics to dielectric susceptibility is also discussed.

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FERROELECTRIC PHASE THRANSITIONS VIEWED VIA NONLINEAR DIELECTRIC RESPONSE

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"*No phase transition can take place without nonlinearity, and therefore it is absolutely necessary to disclose the nonlinearity aspects in order to understand the mechanism of phase transitions.*" This statement of Ishibashi [1] is still very timely. Despite its obvious importance different aspects of phase transformations are still mainly investigated via some linear responses, which manifest their anomalies at the phase transition point. In the particular case of the dielectric response this unsatisfactory situation is due to the lack of commercially available devices for measuring the nonlinear electric *ac* susceptibility. Only a few methods have been developed to this end [2-5]. With materials featuring nonlinear dielectric properties many advanced devices can be constructed, where the electric susceptibility can be tuned by electric fields.

Using our susceptometer [4] we have obtained very reliable data on the nonlinear dielectric responses of barium titanate, BTO (discontinuous phase transition, PT), triglicine sulphate, TGS, and lead germanate, PGO (continuous PTs), as well as on the prototypical relaxor lead magnoniobate, PMN, the uniaxial relaxor strontium-barium niobate, SBN, and the doped quantum paralectrics $K_{0.995}Li_{0.005}TaO_3$, KLT, and $Sr_{0.98}Mn_{0.02}TiO_3$, SMnT. The classic ferroelectrics BTO, TGS and PGO display all features of the third order dielectric susceptibility, χ_3, where $P = \varepsilon_0(\chi_1 E + \chi_2 E^2 + \chi_3 E^3 + \ldots$, as predicted by mean-field theory (including the change in sign of χ_3 at the continuous PT). On the other hand, the positive sign of χ_3 measured in relaxors is due to the presence of polar nanoregions. The 3rd order susceptibility of diluted KLT discloses features of a weakly interacting dipolar system, while it hints at a divergence at the dipolar glass temperature in SMnT.

Thanks are due to the Polish Ministry of Science for financial support under grant No. N N507 455034. WK thanks the Foundation for Polish Science for an Alexander-von-Humboldt Honorary Fellowship.

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MONDAY, SEPTEMBER 13, 2010

Poster presentations

BROADBAND DIELECTRIC SPECTROSCOPY OF FERROELECTRIC (1-x)$Ag_{0.9}Li_{0.1}NbO_3$ - x$Bi_{0,5}K_{0,5}TiO_3$ CERAMICS

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Lead based relaxor ferroelectric single crystals and ceramics with the perovskite structure, like PMN, PZN, PLZT and PMN-PT have been widely studied and met promising applications e.g., as very efficient piezoelectric materials. Nowadays, is generally accepted that diffusive and frequency dependent dielectric anomaly in relaxors is caused of dynamics of polar nanoregions, with appears near the Burns temperature (T_B). Due a toxicity of lead one very important topic is searching of lead free relaxor ceramics and crystals, which exhibit piezoelectric properties similar to PMN-PT. On the other hand the investigations of broadband dynamics of polar nanoregions are rather, because it is very difficult to perform dielectric investigations of relaxors in frequency range 1 GHz – 1 THz near and below T_B. In this work we presented dielectric properties of (1-x)$Ag_{0,9}Li_{0,1}NbO_3$ - x$Bi_{0,5}K_{0,5}TiO_3$ (ALNBKT) ceramics in very wide frequency (20 Hz – 2 THz) and temperature ranges (500 K – 20 K). The dielectric dispersion of investigated ceramics appears in microwave and THz frequency range at high temperatures. On cooling the dielectric dispersion becomes broader and it splits into two parts below the dielectric permittivity maximum temperature at 1 kHz. The low frequency dielectric dispersion part anomalously slows down and at low temperatures its part appears without our frequency range. However, the high frequency part remains at microwave and THz frequencies. The distribution of relaxation times has been calculated from the dielectric spectra. The longest relaxation times diverge according to the Vogel-Fulcher law, while the most probable relaxation time diverges according to the Arhenius law. Such dielectric behaviour is typical for ferroelectric relaxors. At very low temperatures (below 100 K) the relaxation dielectric dispersion almost vanishes in THz range. Here the resonant soft mode was observed. The microscopic origin of polar nanoregions in lead free (1-x)$Ag_{0,9}Li_{0,1}NbO_3$ - x$Bi_{0,5}K_{0,5}TiO_3$ relaxors is discussed.

ELECTRIC PROPERTIES OF La^{3+} DOPED BARIUM TITANATE CERAMICS

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Barium titanate ceramic $BaTiO_3$ (BT) has been investigated extensively because of scientific interest as well as various applications to electronic devices. When pure $BaTiO_3$ is highly resistive at room temperature ($\rho > 10^{10}\Omega cm$). However its electrical resistivity can be dramatically lowered by some dopants, like rare earth elements [1-2]. In the present research lanthanum-doped BT ceramics described by the general formula $Ba_{1-x}La_xTi_{1-x/4}O_3$, for x=0.001 and x=0.003, was prepared by free sintering method. The electrical properties of (BLT) materials were studied by impedance spectroscopy with a Quadtech 1920-type precision LCR meter was utilized and parameters of the phase transition were determined. The equivalent circuit method was applied to fit the experimental data. Study the possibility of application of La-doped BT ceramics for fabrication piezoresistive transducers sensitive for small stress load.

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SYNTHESIS AND FABRICATION OF CERAMIC 0-3 COMPOSITE MATERIAL FOR THERMO-RESISTIVITY APPLICATION

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In this paper ceramic-ceramic composites with 0-3 connectivity were prepared from $Bi_4Ti_3O_{12}$ (BiT) and $BiFeO_3$ (BF) ceramic powders by free sintering and hot pressing method, for different concentration of the ceramic phases. $Bi_4Ti_3O_{12}$ and $BiFeO_3$ ceramic powders were synthesized by the conventional mixed oxide method (MOM). Nanosized BiT powder was dispersed in a BF solution and next such composite was pressing and sintering. The morphology of BiT//BF composites was observed by scanning microscopy, crystalline structure was studied by X-ray diffraction method. The dielectric permittivity and ferroelectric properties of the ceramic-ceramic composites were also investigated. It was found, that properties of the ceramic-ceramic composite are not a simple sum of properties of the phases constituting the composite but they depend on both the way of connectivity and mutual influence of the phases on each other. Composites that are constituted from electroceramics have gained widespread application in a number of thermo-resistive sensors [1, 2].

Keywords: $Bi_4Ti_3O_{12}$, $BiFeO_3$, composite

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La-DOPING EFFECTS ON DIELECTRIC PROPERTIES OF $Bi_4Ti_3O_{12}$ CERAMICS

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La-modified $Bi_4Ti_3O_{12}$ is a typical ferroelectric, piezoelectric and electro-optic material, having relatively low coercive field, low dielectric constant, high Curie temperature and high breakdown strength [1, 2]. The solid solutions $Bi_{4-x}La_xTi_3O_{12}$ BiTLax (x=0, 0.25, 0.5, 0.75) ceramic powders was prepared from mixture of oxide by a solid state reaction. Ceramic powders of BiTLax were next press and compacts sintered by the free sintering and hot pressing method at temperature T=1000^{0}C and T=1100^{0}C for time t=2h. $Bi_{4-x}La_xTi_3O_{12}$ ceramic powder was studied by simultaneous thermal analysis. A crystal structure with Bi-layered structured ferroelectrics was confirmed by a X-ray diffraction (XRD). Chemical composition and microstructure of the ceramic were performed by scanning electron microscope and EDS. Influence La-doping on dielectric properties of $Bi_4Ti_3O_{12}$ ceramics was studied by impedance spectroscopy at different temperature measurement.

Keywords: La-doping, $Bi_4Ti_3O_{12}$ ceramic, impedance spectroscopy

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IMPEDANCE SPECTROSCOPIC STUDIES OF BISMUTH FERRITE

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$BiFeO_3$ (BFO) belongs to perovskite class of complex oxides. In the present paper results of the studies devoted to fabrication and characterization of bismuth ferrite. Dielectric properties has been studied by impedance spectroscopy. The tool of impedance spectroscopy was applied to characterize relaxation processes present in the system at various, but kept constant temperature, as well as to separate the bulk, grain boundary, and electrode processes of the polycrystalline ceramic material. To analyze the impedance spectroscopy data the Nyquist (Z'' vs. Z') plots as well as the simultaneous representation of the imaginary part of impedance and electrical modulus (Z'', M'') vs. frequency were used. The Kramers-Kronig data validation test was employed in the present impedance data analysis.

Keywords: Bismuth ferrite; Perovskites; Impedance; Spectroscopy.

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PECULIARITY OF LOW DENSITY PLASMA-ASSISTED CHARGE EMISSION
PRODUCED AT LATGS CRYSTAL SURFACE

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Time and energy distribution measurements for a weak plasma-assisted electron and ion emission, initiated at the surface of L-α alanine doped TGS (LATGS) single crystals by a drive ac (500 Hz) electric field exceeding 5 x 10^3 V/cm, have been performed. The experimental procedure applied is described in papers by Biedrzycki *et all*[1,2]. Some of the obtained distributions are presented in Figures 1 and 2. As it is seen the time spectra of charges measured at temperatures below phase transition (Fig. 1a) are considerably different from that detected above phase transition (Fig. 2a). Moreover it was found that the total number of charges registered under switching conditions is about an order higher than that produced during mere charging of the sample.

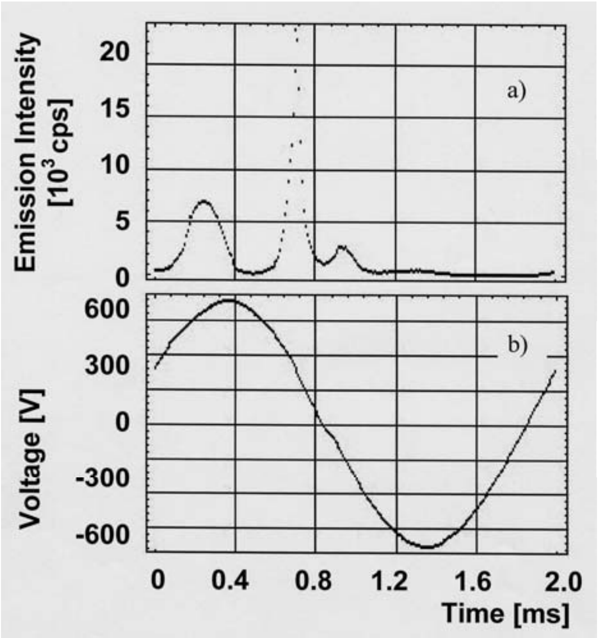

Fig. 1.

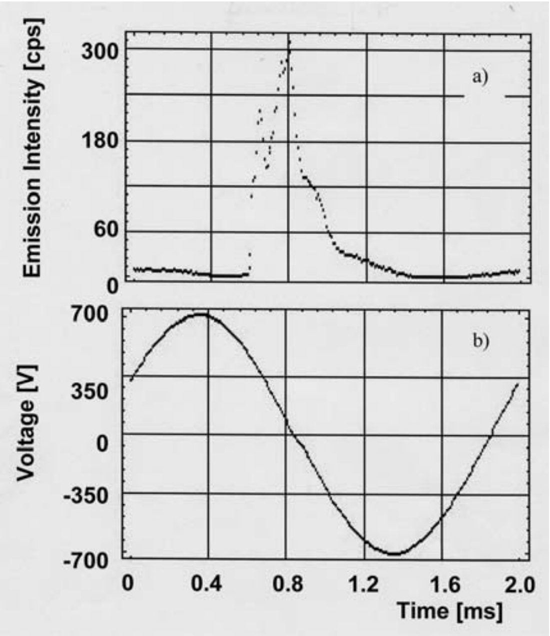

Fig. 2

Time distributions of the charge emission for LATGS crystal sample excited with a drive ac electric field with amplitude 7.75 x 10^3 V/cm integrated over 5 x 10^5 (Fig. 1a) and 10^6 (Fig. 2a) field cycles, corresponding to 321 K (Fig. 1) and 333 K (Fig. 2). The waveforms of applied ac electric field is displayed in the Fig. 1b and Fig. 2b. The time window (2 ms) is divided into 250 time channels.

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RELAXATION OF PERMITTIVITY IN $[(CH_3)_2NH_2]_3CuCl_5$ CRYSTAL IN EXTERNAL ELECTRIC FIELD

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The time influence of constant external electric field on the properties of $[NH_2(CH_3)_2]_3CuCl_5$ ferroelectric crystal are presented. We studied the time dependence of the permittivity under influence of electric field at few constant temperatures above T_c. The slow transition to ferroelectric phase through long-time metastable states and coexistence of incommensurate and ferroelectric phases was observed. The time dependence of permittivity under influence of electric field at constant temperatures above T_c shows its slow but significant decrease, which can be described with two main relaxation processes. The saturated value of permittivity and induce transition to ferroelectric phase can be reached after long time lasting application of electric field and similar behaviour was observed in measurements of induced polarization. The observed dielectric behaviour can be related to coexistence of both phases and slow transition to ferroelectric one through long-time metastable states connected with presence of incommensurate phase in studied crystal. The coexistence of phases and slow transition was confirmed in appearance of induced polarization under external electric field measured as depolarization current.

PRESURE EFECT ON THE ANOMAL BEHAVIOUR OF PHYSICAL PROPERTIES OF SASD TYPE CRYSTALS

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All observed experimentally anomalies in dielectric susceptibility, heat capacity, lattice dynamics behaviour for the isomorphic crystals $NaNH_4SO_4 \cdot 2H_2O$ and $NaNH_4SO_4 \cdot 2H_2O$ was consistently interpreted upon on modified-Mitsui model taken into account the EPR results of subjected systems. We found that the inclusion of short and long range correlations which takes place for two inequivalent active groups $\mathrm{SO}_4(\mathrm{I})\,(\mathrm{SeO}_4(\mathrm{I}))$ and $\mathrm{SO}_4(\mathrm{II})\,(\mathrm{SeO}_4(\mathrm{II}))$ modify temperature behaviour of the spin-Hamiltonian D parameter and gives much better explanation of experimental results. The experimental data subject to our analysis are partially taken from papers [1, 2, and 3]. Using the two-time Green functions method the spectrum of bonded quasispin-fonon vibration has been founded and analysed. The shift of both the phase transition temperature T_C and the points of observed experimentally anomalies at T_1 and T_2 under hydrostatic pressure were calculated. The problem of the phase transition order change caused by external pressure is discussed. The peculiarities of some thermodynamic functions (dielectric susceptibility, heat capacity) behaviour are explained.

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SPECTRAL ELLIPSOMETRY STUDY OF SBN SINGLE CRYSTALS IN VISIBLE AND ULTRAVIOLET REGION

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Strontium-barium niobate $Sr_xBa_{1-x}Nb_2O_6$ (SBN) is a well known ferroelectric crystal considered as one of the very attractive material for practical applications. In particular, much attention is paid to the optical properties of SBN: electro-optic, photorefractive, and acousto-optic. Properties of SBN are known to be substantially dependent on Sr/(Sr+Ba) ratio x revealing a gradual crossover from typical ferroelectric ($x = 0.40$) to extreme relaxor ($x = 0.75$). However, the photon energy, E, dependence of electric permittivity $\varepsilon(E)$ (dielectric function) of SBN in the wide range of transparency and fundamental absorption (visible and ultraviolet regions) is found to be rather unknown.

In the present study, the dielectric function $\varepsilon(E)$ of SBN crystals of the anisotropic crystal cut [100] for five values of the ratio x in the range of 0.40 – 0.75 has been measured by the spectral ellipsometry method using the synchrotron radiation of BESSY II (Berlin) in the photon energy range of 1.5 – 9.9 eV. The experimental results thus obtained for the real (ε_1) and imaginary (ε_2) parts of the permittivity are discussed in comparison with those obtained from the first principles calculations. Our experimental data $\varepsilon(E)$ agree satisfactorily with the reference near-normal incidence reflectance spectra $R(E)$ of SBN70 ($x = 0.70$) [1].

Two maxima of the imaginary part of dielectric function $\varepsilon_2(E)$ of SBN crystals are found. The first maximum is placed in the range of 4.6 – 5.3 eV and second one is in the range of 8.3 – 8.9 eV. However, exact positions depend on the direction of light polarization and x.

The real and imaginary parts of dielectric permittivity are established not to be monotonically dependent on x displaying an extremum near $x = 0.61$. Complementarily, anisotropy of the components of dielectric function, $\Delta\varepsilon_1$ and $\Delta\varepsilon_2$, in respect to the dependences on the photon energy E and x have been analyzed.

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BAND STRUCTURE AND OPTICAL CHARACTERISTICS OF TDA CRYSTALS

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Dielectric crystals Tris(hydroxymethyl)aminomethane(Dihydrogen)Arsenate (TDA), $[(CH_2OH)_3CNH_3]H_2AsO_4$, are known to be characterized by the monoclinic polar group of symmetry $P2_1$ [1]. They show temperature change of electrical polarization $\Delta P = (20\ldots120)\mu\mathrm{Cm}^{-2}$ in the range 100…380 K, dielectric permittivity, $\varepsilon = 6\ldots7$, in the low-frequency region near 1 kHz and can be of interest for the nonlinear optical applications, such as second harmonic generation (SHG) [1].

We present results of the first-principles calculations of electronic band structure, density of states (DOS), dielectric function $\varepsilon(\omega)$ in the photon range (ω) of electronic excitations 0…40 eV and phase matching conditions for SHG in the range of optical transparency $\omega < E_g$ (E_g is a band gap) of TDA obtained using CASTEP code (CAmbridge Serial Total Energy Package), which functions in a framework of the density functional theory (DFT).

The results obtained reveal that TDA is characterized by small electron wave vector dispersion of electronic bands $E(k)$ and relatively big valence-to-conduction energy gap $E_g = 5$ eV. Theoretical values of the refractive indices n_i ($i = 1,2,3$) were found to be in the range 1.62…1.64 for the photon energy $\omega = 1$ eV. However, taking into account a known decrease of the band gap E_g obtained by the DFT based *ab-initio* methods, the real magnitudes of refractive indices can be near 5% smaller than the calculated ones. On the basis of the real part of dielectric function $\varepsilon_{1i}(\omega)$ ($i = 1,2,3$) in the range of transparency of TDA (0.4…5 eV), the phase matching angles θ and ϕ for SHG have been calculated. The results obtained reveal possibility of SHG ($\omega \rightarrow 2\omega$) in TDA for the frequency range $\omega = 0.5\ldots0.8$ eV.

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INFLUENCE OF ELECTROMECHANICAL FIELDS ON DIELECTRIC PROPERTIES of PLZT–x/65/35 ceramics (x=6 and 7)

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High density and transparent PLZT–x/65/35 (x=6 and 7) ceramics with the average grain size of 5-7µm was obtained by two–stage hot–pressing technology [1]. The first stage was performed at 1150-1180°C for 1h in vacuum at pressure 20MPa. The second stage was performed at 1150-1200°C for 1-40h depending on size (15-90mm diameter) at pressure 20MPa in air or in O_2 rich atmosphere.

The measurements of temperature/frequency dependence of the permittivities of these ceramics under uniaxial pressure (0-1000 bar) applied parallel to the *ac* electric field have been carried out. It was found that uniaxial pressure significantly influences dielectric properties of these ceramics. With the increase of pressure the maximum intensity in ε(T) curve decreases, becomes more diffuse and shifts towards higher temperature, and dielectric dispersion decreases. It was concluded, that the applied uniaxial pressure or increasing Ti–ions content in PZT solid solutions induces similar effects. The results were discussed in term of an elastic changes in inter–onic distances in the crystal structure and domain switching under the action of electromechanical loading.

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DIELECTRIC NONLINEARITY IN $(NH_4)_3H(SeO_4)_2$ CRYSTAL

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The results of dielectric measurements on $(NH_4)_3H(SeO_4)_2$ crystal near the ferroelectric phase transition are presented. The experimental data are described according to the Landau theory, and expansion coefficients of the Landau potential A_0, B, C are estimated. The measurement at constant temperatures revealed dependence of the effective nonlinearity coefficient B on temperature in a range of $T_c + 10$ K.

INFLUENCE OF Sr-SUBSTITUTION ON DIFFUSENESS OF $(Ba_{1-x}Sr_x)TiO_3$ PHASE TRANSITIONS

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The results of X-ray diffraction (XRD), scanning electron microscopy (SEM) and dielectric measurements performed for samples of the polycrystalline solid solutions of $(Ba_{1-x}Sr_x)TiO_3$, was presented. An increase of diffuseness of ferroelectric phase transition and a decrease of the temperature T_m (the temperature of the maximum value of the real part of the electric permittivity; □') with increase of Sr concentration was observed. Values of the phase angle (approximate to -90^0) measured in broad range of temperatures suggest the occurrence of polar regions (clusters) in the temperature range of paraelectric (PE)-ferroelectric (FE) phase transition.

DILATOMETRIC AND DIELECTRIC INVESTIGATIONS OF PHASE TRANSITIONS IN $[(CH_3)_2CNH_3]_4Cd_3Cl_{10}$ CRYSTAL

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Thermal expansions and dielectric properties of $[(CH_3)_2CNH_3]_4Cd_3Cl_{10}$ crystal were performed along three crystallographic directions in the temperature range 265 – 375 K. Anomalies of elongation $\Delta l/l$ and thermal expansion coefficient α were observed at 353 K and 294 K. The character of anomalies gives evidence that phase transition at 353 K is of the first order and at 294 K is continuous one. The obtained from measurement volume expansion $\Delta V/V$ shows abrupt decrease of volume at 353 K and characteristic for the first order phase transition and continuous decrease below 293 K. Corresponding volume expansion coefficient α_v shows anomalies at phase transitions temperatures. Additional dielectric studies were performed to confirm the character of phase transitions.

EPR SPECTROSCOPY AND FERROELASTIC DOMAIN STRUCTURE IN THE $LiRbSO_4$-$LiCsSO_4$ SYSTEM

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Electron paramagnetic resonance (EPR) spectra of vanadyl ions doped in mixed crystals of $LiCs_{1-x}Rb_xSO_4$ (x=0.05) have been studied at room temperature in the X-band frequency. The EPR spectra have been found to be very complicated with a large number of lines. For this reason only the intense EPR lines have been taken into account. They are ascribed to two kinds of vanadyl complexes, VO(I) and VO(II) located in different chemical environments. The spin-Hamiltonian parameters are calculated for them at room temperature.

The ferroelastic domain studies were carried out using a polarizing microscope. The seeding of domains was observed at 208 K. Domain patterns and domain wall orientations are characteristic of transition from the rhombic-to-monoclinic system.

TECHNOLOGY AND MAIN PROPERTIES OF $(Pb_{0.75}Ba_{0.25})(Zr_{0.65}Ti_{0.35})_{1-z}Sn_z)O_3$ CERAMICS

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The results of obtaining and investigations of $(Pb_{0.75}Ba_{0.25})(Zr_{0.65}Ti_{0.35})_{1-z}Sn_z)O_3$ (PBZST) ceramics within the range $0.02<z< 0.1$. Samples have been obtained from powders PbO, ZrO_2, TiO_2, SnO_2 and $BaCO_3$. Powders were milled using FRITSCH ball-mill by 15h. Next step of technology was calcination at T_{calc}=850°C/t_{calc}=3h. The final step was pressureless sintering (free sintering FS) at 1250°C/4h.

Results of main microstructural investigations, XRD investigations, dielectric, d.c. conductivity and electromechanical investigations are presented. SEM images of samples with z=0.02 and z=0.04 are presented in Fig.1. It is seen that the microstructures are different what lead to differences in other properties of investigated ceramics.

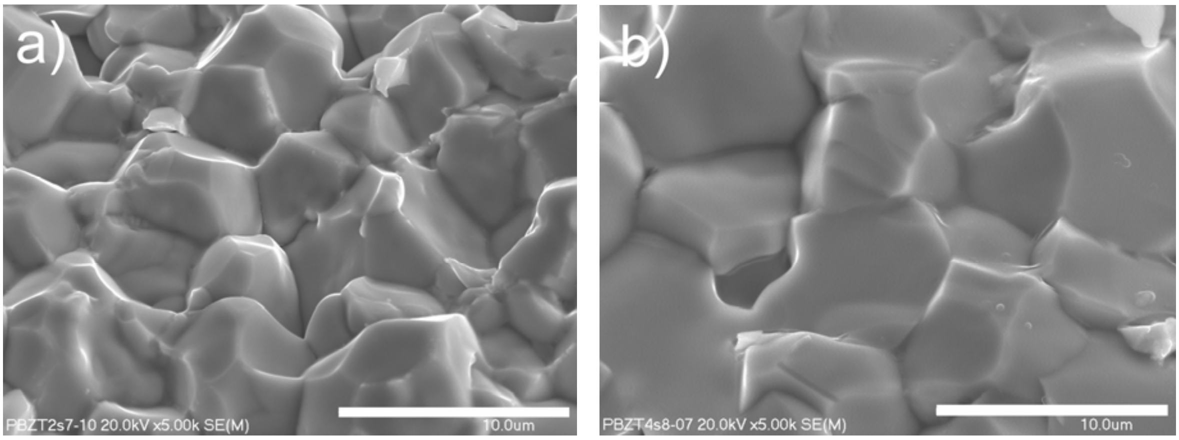

Fig. 1: SEM images of ceramic samples:

(a)- $(Pb_{0.75}Ba_{0.25})(Zr_{0.65}Ti_{0.35})_{0.98}Sn_{0.02})O_3$

(b)- $(Pb_{0.75}Ba_{0.25})(Zr_{0.65}Ti_{0.35})_{0.96}Sn_{0.04})O_3$

FERROELECTRIC PROPERTIES OF PBZTN CERAMICS

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We present the results of investigations of Nb^{5+} doped PBZT ferroelectric ceramics with composition $(Pb_{0.84}Ba_{0.16})_{1.01}(Zr_{0.54}Ti_{0.46})_{0.97}Nb_{0.02}O_3$ (PBZTN). Ceramic samples have been obtained with final pressureless sintering (free sintering FS). XRD investigations, SEM investigations of microstructure, TEM investigations of domain structure have been done for obtained samples as well as investigations of dielectric properties and hysteresis loops. It has been stated that the introduction of Nb into PBZT leads to improve of electromechanical properties and decreases the ferroelectric hardness. The properties of such ceramics suggest that it can be applied in bimorph and ignitron transducers. High values of dielectric permittivity allow to use the material in multilayered ceramics capacitors.

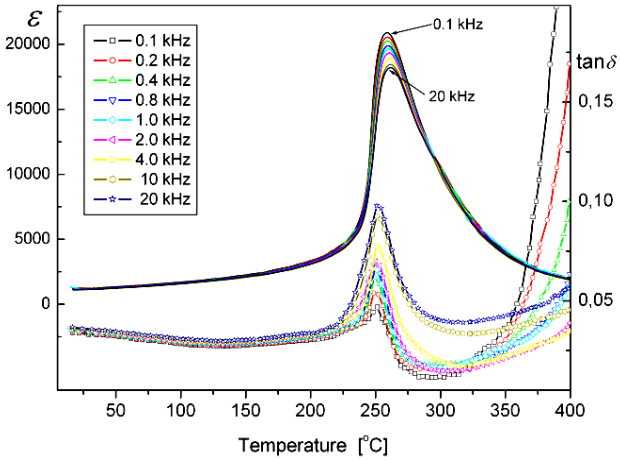

Fig.1. Temperature dependences of the electric permittivity (ε) and the tangent of the angle of dielectric losses (tanδ) for investigated PBZTN ceramics.

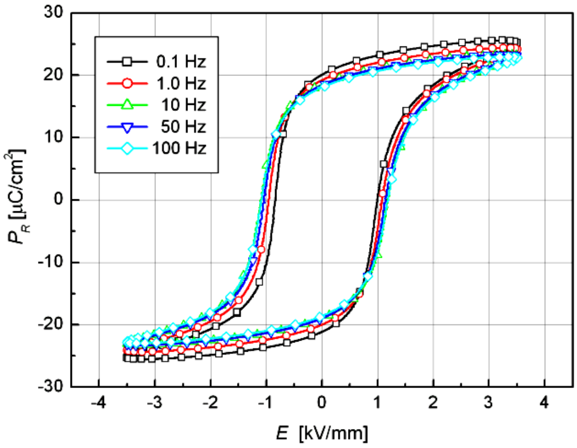

Fig.2. Hysteresis loops at room temperature for investigated PBZTN ceramics.

EFFECTS OF VTE TREATMENT ON COMPOSITION OF LITHIUM TANTALATE SINGLE CRYSTALS

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The whole set of physical properties of lithium tantalate ($LiTaO_3$) crystals is strongly affected by the change of the Li\Ta ratio. However, the stoichiometric lithium tantalate crystals grown from melt are not homogeneous and of good enough quality for optical applications. In the case of specimens of relatively small size (up to 3 mm thick), a beneficial method for making the Li/Ta ratio up to 1 is treatment by Vapour Transport Equilibration (VTE). The method consists of prolonged annealing of the samples of lithium tantalate single crystal the composition of which is different from stoichiometric, in a lithium-saturated atmosphere at high temperatures.

Lithium tantalate plates of two different crystallographic orientations (Z-section and Y\128^o section) were treated by VTE to study possible ways of producing materials with predetermined properties. Samples of both types were annealed at 1050 and 1200 ^{0}C in a ceramic crucible covered with lid prepared by original techniques from a mixture containing 50 % $LiTaO_3$ + 50% Li_3TaO_4.

Results of DTA and of exact measurements of the hexagonal lattice parameters of specimens after VTE treatment allowed inferring that:

1. Diffusion under VTE conditions proceeds by several mechanisms: Li^+ fills the available lithium vacancies in the crystalline lattice. Simultaneous diffusion between lattice points, along dislocations and surface is likely possible.
2. Density of dislocations in the direction perpendicular to plane (1014) is higher ($\sigma \sim 10^5$ cm^{-2}) and $\sigma \sim 5 \cdot 10^4$ cm^{-2} for (0001) suggesting diffusion along dislocations and between lattice points is higher compared with diffusion along Z-axis.
3. Extending the time of VTE at constant temperature does not increase the Li/Ta ratio since the VTE treatment is a dynamic process.

Further studies have revealed the presence of layers of different phase composition in lithium tantalate specimens after VTE treatment. There is a 15-30 μ thick layer containing lithium orthotantalate near the surface of plane-parallel specimens. This layer being polished off a 400-600 μ thick layer of composition close to stoichiometric is revealed. In layers further down the Li\Ta ratio monotonously decreases with depth. Polarisation curves P(E) of the different lithium tantalate layers were measured in the range from room temperature up to ~360 ^{0}C. At 360 ^{0}C distinct Barkhausen hops are observed, the hysteresis loops remaining unsaturated and unsymmetrical, which is also a characteristic of partial switching. This confirms that VTE treatment under the conditions above promotes formation of thin layers of composition close to stoichiometric, with smaller coercive field values compared with bulk where switching is absent. As a result, the filed intensity in the surface layer is much lower than in the bulk, and switching is only partial.

The thin layers of a close to stoichiometric composition can be integrated in optical devices of a regular domain structure.

The study has been supported by grants 09-03-00141, 09-03-00189, 09-03-90401, 09-03-12004 of the RFBR, grant of the President of the RF SS – 2762.2008.3, and grant of Carl Zeiss.

DIELECTRIC RESPONSE IN LEAD FERROTANTALATE WITH RESPECT TO BIAS FIELD EFFECT

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The study is focused on dielectric properties of $PbFe_{1/2}Ta_{1/2}O_3$ ceramics at low and infra-low frequencies under bias field in a wide range of temperature.

Behaviour of dielectric permittivity ε′(T) and dielectric loss ε"(T) at frequencies of 1Hz and 1 kHz at different bias field intensities is illustrated in Figure 1. The observed shift of the ε′(T) maximum to higher temperature is linear of the order of 1.9 degrees per KV/cm at 1 Hz and 1.6 degrees per kV/cm at 1 kHz. The other revealed anomalies of the dielectric response at $T < T_m$ are explained by structural transformations within the region of the broad phase transition while those at $T << T_m$ – by magnetic-electric interaction in $PbFe_{1/2}Ta_{1/2}O_3$.

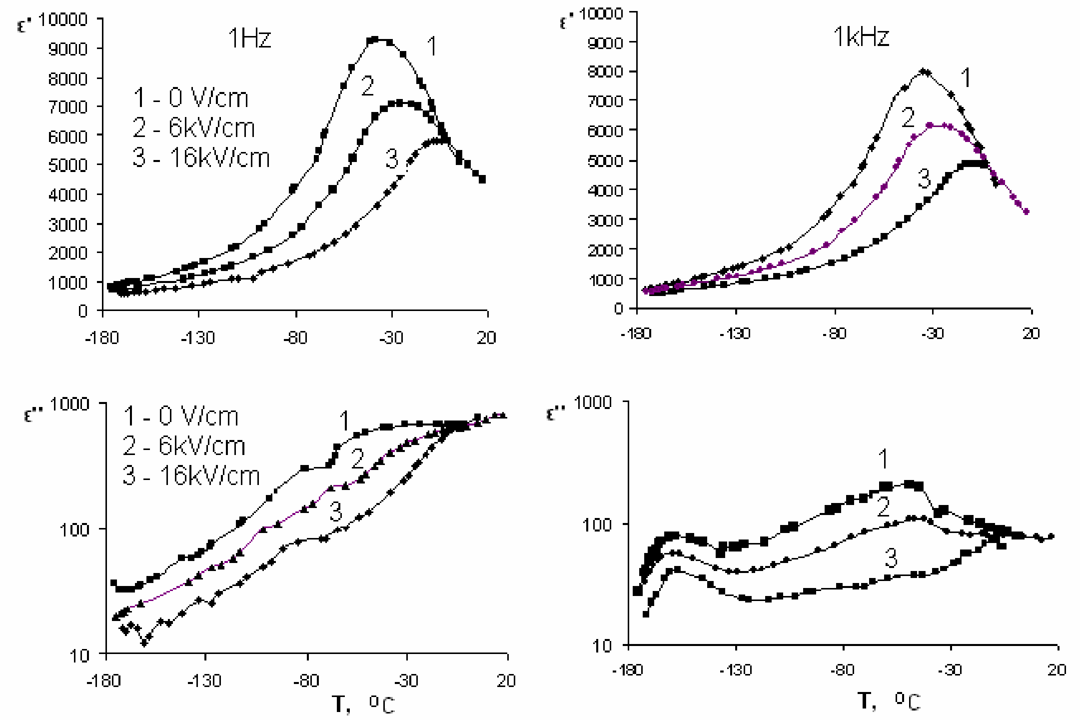

Fig. 1. Behaviour of ε′(T) and ε"(T) curves in $PbFe_{1/2}Ta_{1/2}O_3$ ceramics at frequencies of 1 Hz and 1 kHz at different bias field intensities.

STRUCTURAL AND ELECTRIC PROPERTIES OF SODIUM LITHIUM NIOBATE CERAMIC SOLID SOLUTION $Li_{0.08}Na_{0.92}NbO_3$

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Sodium niobate $Na^{+1}Nb^{+5}O_3$ is a part of family of compounds ABO_3, which are considered to be promising materials for wide technological applications. One of the most interesting and extensively studied compound is a solid solution based on $NaNbO_3$: sodium nobate-lithium niobate system ($NaNbO_3$-$LiNbO_3$).

The $Li_{0.08}Na_{0.92}NbO_3$ ceramic sample was prepared by a two-stage hot-pressing technology. A powder X-ray diffraction pattern shows that the $Li_{0.08}Na_{0.92}NbO_3$ polycrystalline ceramics at room temperature belongs to the orthorhombic phase with calculate lattice constants a=0.5491 Ĺ, b=1.5505 Ĺ and c=0.5552 Ĺ. From the obtained results it werestated that the values of lattice parameters in $Li_{0.08}Na_{0.92}NbO_3$ decrease in comparison with the pure $NaNbO_3$. The purity and chemical composition of the sample was investigated by an EDS analysis. No significant impurities were detected in an EDS survey spectrum. The morphology of $Li_{0.08}Na_{0.92}NbO_3$ was performed by means of scanning microscope (Hitachi S4700). The SEM images show that the sample is perfectly sintered, dense and possess a compact structure. The grains are clearly visible, they are regular in shape and their average size is about 16 □m. Electrical conductivity investigations were performed depending on temperature and frequency of electric measuring field in heating and cooling processes. Local minima appearing in the temperature range from 303 to 353 K are associated with a polaronic transport mechanism of electrical conductivity. The minimum values of this electrical conductivity are a results of the change in the nature of the conduction mechanism: from the tunnel into the hopping mechanism, that is the short-range into the long-range mechanism. At low temperatures (303-353K), with the increase in temperature we observe a strong decrease in the value of the electrical conductivity, which is followed by its increase. In the phase transition region the electric conductivity reaches a local minimum for each value of frequency of the measuring electric field. At high temepartures at all requencies the value of a.c. conducitivity differs little from the value of d.c. conductivity.

The analysis of the experimental data shows that that the a.c. conductivity changes with frequency according to the formula: $\sigma(\omega) = \omega^n$. The calculated values of the coefficient *n* are slightly smaller than unity. This indicates that a current arises as a result of the dipoles reorientation in the alternating electric field.

ELECTRICAL TRANSPORT PROPERTIES OF LEAD–FREE $(Na_{0.5}Bi_{0.5})_{(1-x)}Ba_xZr_{0.04}Ti_{0.96}O_3$ CERAMICS (x=0, 0.06, 0.085, 0.1)

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The lead-based materials such as PZT and PZT-based multicomponent ceramics are widely used for various applications, however these ceramics cause crucial environmental pollution. Therefore it is necessary and urgent to develop lead–free materials with excellent properties to replace the lead–based ceramics. Some lead–free materials, including $Na_{0.5}Bi_{0.5}TiO_3$ (NBT) and NBT-based solid solutions, with promising electromechanical properties could be good candidates for it.

The lead-free ceramics $(Na_{0.5}Bi_{0.5})_{(1-x)}Ba_xZr_{0.04}Ti_{0.96}O_3$ were prepared by a solid phase hot pressing sintering process. The powders of $(Na_{0.5}Bi_{0.5})_{(1-x)}Ba_xZr_{0.04}Ti_{0.96}O_3$ were obtained by a solid phase synthesis from high purity grade oxides and carbonate: BaO, Na_2CO_3, Bi_2O_3, ZrO_2, TiO_2. The mixture of starting raw materials was homogenized and milled in an agate ball mill in ethanol for 20 hours, dried and then calcined. The calcined powder was reground, cold pressed and depending on BaO concentrations, sintered by hot pressing for 2 hours at temperature ranging from 1160 to 1200°C and pressure of 25 MPa. The details connected with the samples preparation are given in Ref. [1]. The values of density of obtained samples are higher than 95 per cent of the theoretical ones. The samples with x=0.06, 0.085 and 0.1 were investigated in the present work. For these samples an *ac* and *dc* electric conductivity were studied. A low frequency (100Hz-100kHz) *ac* conductivity obeys the power law characteristic for disordered materials. The *dc* conductivity has a thermally activated character. The barrier hopping model is found to explain the mechanism of charge transport in these materials. The NBT-BTZ system is expected to be a new and promising candidate for the lead–free electronic ceramics.

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STRUCTURAL, PYROELECTRIC AND ELECTRIC PROPERTIES OF $Na_{1-X}Li_XNbO_3$ CERAMICS

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Ceramics materials showing ferroelectric behavior are being used in many applications in electronics and optics. The ferroelectric with perovskit structure especially are very interesting materials because their strong anomalies in their physical properties, such as dielectric, electric-optical, piezoelectric, pyroelectric properties. These anomalies can be easily modified by apply of an admixture. Solid solution based on sodium niobate has ferroelectric and piezoelectric properties. In this paper XRD analysis, measurements of temperature changes of dielectric permittivity, pyroelectric current and remanent polarization for $Na_{1-x}Li_xNbO_3$ for x=0 to 0,06 are presented. The lattice parameters and ferroelectric properties of this solid solution depend of Li ions concentration. Thermal hysteresis are existing at heating-cooling maximal $\square\varepsilon \approx 860$ for x=0.04 maximal $\square T \approx 80K$ for x=0.01. The pyroelectric measurements were performed in quasistatic method on heating. The remanent polarization depends on the concentration of Li-, it increases for x=0 – 0,06, and it decreases for x>0,02 to maximal value $Pr = 15{,}2\ \square C/cm^{-2}$**.** The investigation of the pyroeffect reveals the existence of polar microregions in the temperature range above T_m.

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MAGNETOVIBRONIC THEORY OF $BiFeO_3$-TYPE MULTIFERROICS

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Multiferroics are multifunctional materials, in which two or more order parameters (ferroelectric, ferro(antiferro)magnetic and ferroelastic) coexist in a single compound [1-3]. In [4-6] it is shown that the electron-phonon (vibronic) covalent hybridization in the ABO_3 -type oxide-perovskites between empty d^0 states of the B ions and occupied oxygen 2p electronic states leads to the occurrence of the ferroelectric order. The theory of ferroelectric displacive phase transitions with an order-disorder component in the ABO_3 -type perovskites is developed in [6]. In [7] an approach is proposed for explaining of ferroelectric properties of the Bi-based multiferroic-perovskite-oxides ($BiFeO_3$, $BiMnO_3$, etc.). It is shown that the electron-lattice covalent hybridization between the Bi $6s^2$ lone electron pair and the empty oxygen 2p states causes instability of R_{25} and R_{15} polar vibrations in the reference cubic phase. The ferroelectric instability holds in the $R\overline{3}c$ phase and the observed ferroelectric transition $R\overline{3}c \rightarrow R3c$ is connected with the softening of the A_{2u} mode. In this report we take into account magnetic and vibronic couplings in the Bi-based multiferroics. Magnetism and ferroelectrcity are involved with local spins (Fe,Mn) and off-center structural distortions (Bi), respectively. We obtained the free energy of the Bi-based multiferroics in external electric and magnetic fields. On the basis of this free energy we investigated the magnetoelectric effects in the Bi-based multiferroics.

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INVESTIGATIONS OF LOW TEMPERATURE PHASE TRANSITIONS IN $BiFeO_3$ CERAMIC BY INFRARED SPECTROSCOPY

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Bismuth-ferrite $BiFeO_3$ (BFO) ceramic belongs to class of magnetoelectric multiferroic materials in which ferroelectricity and magnetism coexist and are coupled. This kind of material attracts attention due to its possible applications in the electronic industry.

BFO samples have been obtained by a conventional ceramic processing method.

The performed SEM and EDS studies revealed that the samples are perfectly sintered, chemically homogeneous and the grains are well shaped. The X-ray diffraction analysis showed that the BFO is rhombohedral at room temperature and contains Bi_2O_3 less than 3 %. BFO compound exhibits four phase transitions in the temperature range from 300 to 8 K (T_1 = 230 K, T_2 = 200 K, T_3 = 142 K and T_4 = 50 K). We have performed the infrared spectroscopy analysis using Bruker VERTEX 70v vacuum Fourier Transform spectrometer in the temperature range from 297 to 8 K (wavenumber range from 4000 to 400 cm^{-1}) in order to investigate low temperature phase transitions.

BFO was cooled from 297 K with the constant rate of 3 K/min to desired temperature and then stabilized for 3 minutes before the spectrum was measured. At higher wavenumbers (above 900 cm^{-1}) quite strong background was observed probably due to scattering caused by the sample. Some interesting changes in the MIR spectra can be observed during cooling. At high temperature two bands between 900 and 750 cm^{-1} are clearly visible: one more intense at 814 cm^{-1} and second weak, appearing as a shoulder at 832 cm^{-1}. During cooling the band at 814 cm^{-1} shifts continuously towards higher wavenumbers and the second band starts to overlap. Below 200 K only one symmetric band is visible. The full width at half maximum (FWHM) of this band decreases exponentially on cooling. The shape of the band at 545 cm^{-1} related to A1 (TO) mode also changes between 60 – 40 K which corresponds to the phase transition at 50 K. With the decreasing of the temperature the broad of the band reveals its components and theirs full width at half maximum decrease.

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AFM AND OPTICAL MICROSCOPY STUDIES OF $KFe(MoO_4)_2$

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$KFe(MoO_4)_2$ belongs to a family of trigonal double molybdates and tungstates in which structural transitions to ferroelastic phases have been found [1]. $KFe(MoO_4)_2$ crystallizes in a trigonal system (space group P-3m1) and below T_1=311K a phase transition of the second order to the monoclinic system occurs [2]; another phase transition of the first order is observed at temperature T_2=139 K [3].

This contribution gives a new insight into nature of the transition in $KFe(MoO_4)_2$ at T_2 through optical microscopy studies of ferroelastic domain structure. We also report the first AFM observation of the ferroelastically twinned $KFe(MoO_4)_2$ surface features and characteristics.

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THE PRESSURE INFLUENCE ON THE PROPERTIES OF FERROELECTRIC $(C_3N_5H_5)_5Bi_2Cl_{11}$

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$(C_3N_2H_5)_5Bi_2Cl_{11}$ (abbreviated as ICB) belongs to the large family of halogenobismuthates (III) described by the general formula $R_aM_bX_{(3b+a)}$ where R denotes organic cations, M denotes Bi(III) atoms and X stands for Cl, Br and I.

The crystal attracts special attention since it possesses interesting biferroic (ferroelectric/ferroelastic) properties with the following sequence of phase transitions at ambient pressure [1]: $P\bar{4}n2 \xrightarrow{360K} P2_1/n$ (I→II), $P2_1/n \xrightarrow{166K} P2_1$ (II→III).

The high temperature phase transition leads from the paraelastic phase (tetragonal symmetry) to the ferroelastic/paraelectric one (monoclinic). The lower temperature phase transition at 166 K leads to the ferroelectric order [1]. The phase transition mechanism at 166 K (an order – disorder type) is postulated to be governed by the dynamics of imidazolium cations. This model of the ferroelectric phase transition mechanism was confirmed by observation of the critical slowing down [2] as well as an ac - calorimetry studies [3].

In this contribution, the effect of hydrostatic pressure on the dynamical dielectric characteristics of $(C_3N_2H_5)_5Bi_2Cl_{11}$ (ICB) single crystals in the vicinity of the ferroelectric phase transition temperature is presented and discussed. The aim of the study was to find a relation between the ferroelectric phase transition and internal dynamics of the compound.

The samples of $(C_3N_2H_5)_5Bi_2Cl_{11}$ (ICB) single crystals were obtained from Prof. R. Jakubas, Department of Chemistry, Wroclaw University The studies have been performed in the frequency range 100Hz-1MHz between 77 K to 180K at selected values of the hydrostatic pressure within the range of 0.1 MPa to 300 MPa.

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NANODOMAIN FORMATION IN THE HETEROSTRUCTURE "SPM TIP ELECTRODE – THIN FERROELECTRIC FILM – SEMICONDUCTOR SUBSTRATE"

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Using Landau-Ginzburg-Devonshire theory we studied the process of cylindrical nanodomains formation in the heterostructure "tip electrode – thin ferroelectric film – semiconductor substrate" with the help of the probe Scanning Probe Microscopy (SPM). The inhomogeneous electric field induced by SPM tip is modeled by the distribution of electric potential on the ferroelectric surface.

During the local polarization reversal the thickness and properties of the charged layer in the semiconductor drastically changes, in particular the carriers depletion is transformed into accumulation and vise versa depending on the polarization direction. Such transformations are accompanied by the peaks of electric current, which shape and amplitude depend on the domain shape and sizes. The electric current could be registered *in-situ* by current SPM. Also it was shown that space charge effects introduce strong size-effect on spontaneous polarization in 20-40 nm epitaxial films of ferroelectric $BiFeO_3$ on half-metal $(La,Sr)MnO_3$ substrate

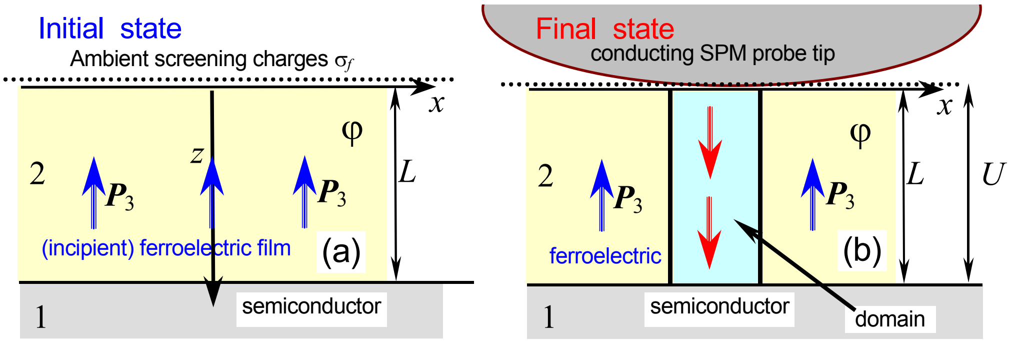

Fig. 1. (a) The initial state of the considered heterostructure: semiconductor/(incipient or proper) ferroelectric-dielectric film of thickness L. P_3 is the ferroelectric polarization. (b) The final state of the ferroelectric film is the local polarization reversal caused by the biased SPM probe.

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TUESDAY, SEPTEMBER 14, 2010

Oral presentations

LOW DENSITY PLASMA-ASSISTED ELECTRON AND ION EMISSION AT THE PHASE TRANSITION OF LATGS SINGLE CRYSTAL

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Electron and ion emission accompanying non-thermal plasma processes, initiated at the surface of LATGS single crystals by a drive ac electric field exceeding 5 x 10^3 V/cm, have been investigated. These plasma-assisted electron and ion emission processes were examined by means of time and energy distribution measurements. The experimental set-up and procedure are described in previous papers given by Biedrzycki *et all*[1,2]. Time spectra of charges (electrons and ions) were obtained at temperatures below as well as above the Curie temperature. Time dependent energy spectra for electrons and ions involve charges with energies up to a few hundreds of eV. The examples of the obtained three-dimensional time and energy distribution of the emitted electrons are illustrated in the Figure 1 and 2, for 321 K and 333 K respectively. The amplitude of sinusoidal (500 Hz) drive ac electric field was 7.75 x 10^3 V/cm.

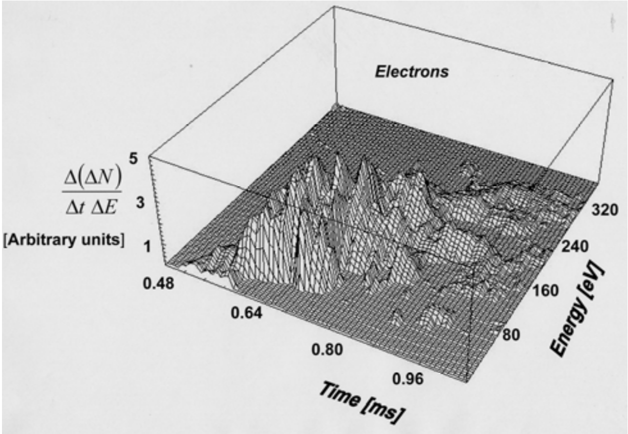

Fig. 1.

Time and energy distribution of the emitted electrons corresponding to the time region 0.48 – 1.2 ms of the 2 ms time window, for 321 K (Fig. 1) and 333 K (Fig. 2).

Time window 2ms is divided into 250 time channels.

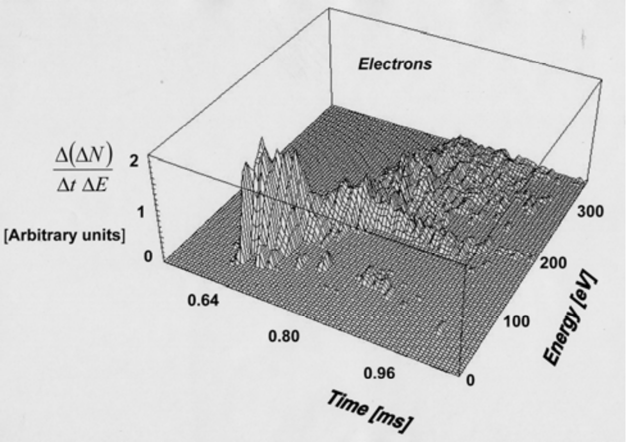

Fig. 2.

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PLZT-BASED LIGHT DRIVEN PIEZOELECTRIC TRANSFORMER

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Lanthanum-modifed lead zirconate titanate (PLZT) ceramic materials have gained considerable attention due to their photostriction, which is the superposition of photovoltaic and piezoelectric effects. This idea was implemented in construction of Piezoelectric Transformer (PT) and used for direct converting photonic energy to electrical one by implementing photostrictive actuators with piezoelectric generator in one complex structure of Piezoelectric Transformer.

Possible application in electronic industry needs thoroughly light correlated electrical characterisation of this new structure constructed from the PLZT material. We revealed by our measurements impedance anisotropy of this structure under light illumination. Additionally application of this material into the Piezoelectric Transformer structure revealed, that the efficiency and gain characteristics of PT device is addition result of starting material parameters and light intensity.

DIELECTRIC SPECTROSCOPY OF THE MIXED $CuIn_xCr_{1-x}P_2S_6$ CRYSTALS

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$CuCrP_2S_6$ and $CuInP_2S_6$ crystals are layered two-dimensional structures. They are formed by layers built up by cores of sulfur atoms where metal cations and P–P pairs fill octahedral voids, within a layer, the Cu, M, and P-P form triangular patterns. According to calorimetric, dielectric and x-ray data [1], $CuCrP_2S_6$ crystals undergo two phase transitions, at $T_{c1} \approx 190$ K and $T_{c2} \approx 150$ K, and separate into three phases: paraelectric [1] $(T > T_{c1})$, antiferroelectric [2] $(T < T_{c2})$ and intermediate quasi-antipolar in the temperature range $T_{c2} < T < T_{c1}$ [2]. X-ray studies have shown the paraelectric phase to have *C2/c* symmetry and the antiferroelectric phase *Pc* symmetry [1]. Detailed X-ray studies of $CuInP_2S_6$ crystals showed that at T_c=315 K the ferroelectric first-order, order-disorder phase transition occurs, which is related to the hoping motion of the Cu ions [3].

In this contribution we presented results of broadband (1 mHz -3GHz) dielectric studies of mixed $CuIn_xCr_{1-x}P_2S_6$ crystals. At higher temperatures and low frequencies dielectric dispersion is mainly caused by high Cu^+ ions conductivity, this was already observed in both $CuCrP_2S_6$ and $CuInP_2S_6$ crystals [4]. When x decreases from 1 to 0.7, then $CuIn_xCr_{1-x}P_2S_6$ crystals transition temperature T_C decrease from 315 K to 155 K. With decrease of x, dipolar glass phase occupy the main dielectric dispersion region. This dispersion was described with empirical Cole-Cole formula. For better description of dipolar glasses we calculated distribution of relaxation times of $CuIn_xCr_{1-x}P_2S_6$ crystals. The distribution of relaxation times has been calculated directly from the dielectric spectra. The parameters of the double well potentials of the Cu bonds, the local polarization distribution function and the average (macroscopic) polarization have been extracted from the dielectric measurements data.

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FERROELECTRIC AND SEMICONDUCTING PROPERTIES OF $Sn_2P_2S_6$ CRYSTALS WITH INTRINSIC VACANCIES

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Ferroelectrics-semiconductors $Sn_2P_2S_6$ (SPS) at room temperature have p-type electric conductivity that strongly depends on the samples stoichiometry. On thermoelectric data n-type conductivity appears at heating till 450 – 500 K. This material has energy gap near 2.3 eV, but at deviation from stoichiometry the color of SPS samples could be changed from yellow to yellow-red. Also several “brawn” SPS samples were grown. For by vapor transport grown SPS crystals strong relaxational dielectric dispersion is observed at high temperatures in paraelectric phase, what have been related to its activation conductivity. Grown by Bridgeman method SPS crystals usually have lower electric conductivity. The first-principles calculations in LDA approach were performed for the SPS paraelectric and ferroelectric phases. The evolution of electron energetic spectra and charge density distribution clearly illustrate the hybridization of cation and anion orbitals that determines stereoactivity of tin atoms lone pair electrons and its leading role in the PT nature. Energetic levels and charge density redistribution together with structure relaxation were calculated for tin vacancies acceptors states and for sulfur vacancies donor states in the paraelectric phase of SPS crystals. Taking account of named donors and acceptors and with consideration of small electron polarons at Sn^+ cations and small hole polarons at S^- anions, next phenomena were explained: the dielectric relaxation in paraelectric phase for some SPS samples; the optic absorption and its change by illumination; the dark electroconductivity and long time persistence electric conductivity after band gap lighting; the white light sensitization of photorefractive recording at red and near infrared spectral range; the light induced EPR signals and its intensity behavior at monochromatic illumination for temperatures below 100 K. It was established the direct connection of thermally induced lattice multistability with light induced optical bistability for $Sn_2P_2S_6$ ferroelectrics – semiconductors.

STRUCTURE AND PHYSICAL PROPERTIES OF $Na_{1/2}Bi_{1/2}TiO_3$-$CdTiO_3$ SOLID SOLUTIONS

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Crystallographic structure, dielectric and electro-mechanical properties, as well as thermal expansion of $Na_{1/2}Bi_{1/2}TiO_3$-$CdTiO_3$ solid solutions were studied in concentration range till 80% of $CdTiO_3$. The structure of solid solutions changes from rhombohedral to pseudocubic, tetragonal and orthorombic as concentration of $CdTiO_3$ increases. The relaxor behaviour in concentration region of pseudocubic symmetry transfers to normal 2nd order ferroelectric phase transition with high value of dielectric permittivity at morphotropic phase boundary to tetragonal symmetry. The concentration dependence of electromechanical properties in relation to morphotropic phase boundaries is discussed.

THE POLARIZATION DISTRIBUTION IN GRADED BTS CERAMIC

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Ferroelectric ceramic materials have been widely used in radioelectronics, hydroacoustics, ultrasonic engineering, and quantum electronics. In particular, in recent years, functional ceramic materials with a composition gradient have been brought in production for a number of practical applications [1-3].

Lead-free $BaTi_{1-x}Sn_xO_3$ piezoceramics (BTS) finding its use in actuators and microsensors was studied with the aid of the thermal square wave method (TSWM) [4]. Functionally graded BTS with tin concentration of $0.075 \leq x \leq 0.15$ varying over the sample thickness were investigated. The samples were fabricated with smooth [3] and step gradient of concentration.

The polarization profiles were studied in a broad temperature range (from –10 to +75 °C) including the Curie temperatures of BTS15 and BTS7.5 composition. The spatial polarization distribution was examined for samples polarized at different temperatures: $T = 25^{\circ}C$, when the phase components having different tin concentrations exist in different (ferroelectric and paraelectric) phases and $T = -7^{\circ}C$, for which the ferroelectric state is realized in all layers of the sample.

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EFFECT OF Zr^{+4} DOPING ON THE ELECTRICAL PROPERTIES OF $BaTiO_3$ CERAMICS

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Barium titanate is a well known lead free ferroelectric material with wide potential application in the electronic industry. The electrical properties of perovskite $BaTiO_3$ (BT) can be significantly revised by the doping of respective cations. It is expected that these properties of BT will be improved by the substitution a small amount of Ti^{+4} by Zr^{+4} because Zr^{+4} is chemically more stable than Ti^{+4}.

The $BaZr_xTi_{1-x}O_3$ (for x=10, 15, 20) ferroelectric ceramics were prepared by a conventional solid state reaction method. These samples were determined by an X-ray diffraction (XRD), scanning electron microscopy (SEM) and atomic forse microscopy (AFM) for crystallographic, surface morphological and compositional studies. A single phase with perovskite structure was identified in the samples at room temperature. No significant impurities were detected in an EDS spectrum and the samples are in good stoichiometric ratio. The densities of the sintered samples were measured by an Archimedes method (in water) using a density balance. The ceramics are well densified and the value of density of the investigated samples are approximately 96% of the theoretical values.

The temperature dependence of electric conductivity were evaluated in the temperature range from 120K to 520K for selected frequency by an Agilent HP4284 LCR meter. The obtained results make it possible to determine the type and mechanism of electrical conductivity as well as the order of magnitude of charge carriers mobility and their activation energy.

STRUCTURE AND ELECTRICAL PROPERTIES OF $Li_{3-x}Sc_{2-x}Zr_x(PO_4)_3$ (x = 0; 0.1; 0.2) CERAMICS

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The results of X-ray diffraction shown that at room temperature the $Li_{3-x}Sc_{2-x}Zr_x(PO_4)_3$ (x = 0; 0.1) compounds belong to monoclinic symmetry (space group $P2_1/n$) and $Li_{2.8}Sc_{1.8}Zr_{0.2}(PO_4)_3$ compound belong to the orthorhombic symmetry (space group Pbcn). In the unit cells of the compounds there are four formula units. The electrical properties of the ceramic samples were investigated by complex impedance spectroscopy in the frequency range from 10 to 3×10^9 Hz and in temperature range from 300 to 600 K. Three relaxation dispersion regions of electrical properties were found. The processes are thermally activated. High frequency dispersion region may be attributed to the relaxation processes in bulk, while at lower frequency regions the dispersions are related to the processes in grain boundaries and blocking electrodes. Anomalies of bulk conductivities of $Li_3Sc_2(PO_4)_3$ ceramics can be seen in the temperature regions from 450 K to 470 K and from 470 K to 530 K. According to the results of the structural analysis [1] the anomalies in the temperature ranges (450 – 470) K and (470 - 530) K can be caused by $\alpha \leftrightarrow \beta$ and $\beta \leftrightarrow \gamma$ phase transitions from monoclinic to orthorhombic symmetry in the compounds. In the temperature range from 420 to 510 K exothermic anomalies of enthalpy, a decrease of the activation energy of the bulk conductivity of the ceramics, marginal anomalies of dielectric permittivity were found for $Li_{2.9}Sc_{1.9}Zr_{0.1}(PO_4)_3$. The phenomena are related to structural phase transition in the investigated material. At temperature 600 K and 900 K $Li_{2.9}Sc_{1.9}Zr_{0.1}(PO_4)_3$ compound belong to the orthorhombic symmetry (space group Pbcn) with four formula units in the unit cell. The thermal expansion coefficient of the lattice volume in orthorhombic phase of the $Li_{2.9}Sc_{1.9}Zr_{0.1}(PO_4)_3$ sample was found to be $1.77 \cdot 10^{-5}$ K^{-1}. The density $d_{X\text{-ray}}$ of the orthorhombic phase decrease with increase of the temperature.

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Cluster Theory Of $Rb_{1-x}(NH_4)_xH_2PO_4$ Type Mixed Crystals. Problems and Outlook

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We discuss a pseudospin model for proton glasses of the $Rb_{1-x}(NH_4)_xH_2PO_4$ ($Rb_{1-x}(ND_4)_xD_2PO_4$) type, which takes into account the energy levels of protons (deuterons) around a PO_4 group (within the cluster approximation), long-range interactions between the hydrogen bonds (within mean-field approximation) and a Gaussian deformational fields.

We explore the temperature dependences of heat capacity, local polarization of hydrogen bonds, Edwards-Anderson parameter, as well as of the real and imaginary parts of longitudinal and transverse dielectric permittivities in a wide range of sample compositions $x=[0,1]$ for the $Rb_{1-x}(ND_4)_xD_2PO_4$, $Rb_{1-x}(NH_4)_xH_2PO_4$, $K_{1-x}(NH_4)_xH_2PO_4$ and $Rb_{1-x}(NH_4)_xH_2AsO_4$ mixed systems. The theoretical phase diagrams obtained from the calculated dielectric permittivity are close to the experimental ones. Deviation of the theory from experiment takes place for all systems at compositions were the transition between "pure" states is observed. For the $Rb_{1-x}(ND_4)_xD_2PO_4$ mixture the theory qualitatively well describes the temperature behavior of the real and imaginary parts of longitudinal and transverse permittivities within the range of "pure" phases at $x \approx 1; 0.5; 0$.

We also discuss possible ways of developing the theory in order to describe the mixed states and to improve the description of the dynamic characteristics.

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RELAXATION PHENOMENA IN $Li_2Ge_7O_{15}$ CRYSTAL

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The abstract is devoted to investigations of relaxation processes occuring in AC and DC electric field in pure and doped with 3d- ions (Cr, Mn) lithium heptagermanate $Li_2Ge_7O_{15}$ crystals. Measurements of temperature- frequency dependencies of dielectric permittivity ε and conductivity σ are completed by EPR spectroscopy of impurity ions. EPR spectra are analyzed in order to clear the microscopic structure of the Cr, Mn doping centers in $Li_2Ge_7O_{15}$ structure.

The performed researches have allowed to propose the microscopic mechanisms of the polarization and charge transfer phenomena in $Li_2Ge_7O_{15}$. It has been shown, that relaxation phenomena in nominally pure and doped $Li_2Ge_7O_{15}$ crystals are determined by the processes of various nature: - thermally activated re- orientations of the dipole moments of Cr^{3+}-Li^+ pair centers; - migration mobility of interstitial Li^+ ions through the channels of crystal lattice; - accumulation of a charge carriers near to blocking Pt electrodes. It has been demonstrated, that lithium germanate crystal matrix is appropriate for design of new materials with high ionic conductivity. A strong influence of doping ions on lithium heptagermanate electrical properties is registered. It has been shown, that introducing of heterovalent impurities allows to control $Li_2Ge_7O_{15}$ electric conductivity in a wide range.

SOFT MODE INVESTIGATIONS IN FERROELECTRICS AND RELAXORS BY TIME DOMAIN THZ SPECTROSCOPY

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Relaxor ferroelectrics have attracted considerable attention in recent years due to their unusual physical behavior and excellent dielectric and electromechanical properties. The broadband dielectric spectroscopy of relaxors showed that the dielectric relaxations appears below Burns temperature in the THz frequency range, anomalously slows down and splits into two part close to freezing temperature. Dielectric dispersion in order-disorder ferroelectrics slows down close to phase transition temperature. Therefore very interest problem is to investigate dielectric dispersion in various disordered solids, which demonstrate ferroelectrics, relaxors and mixed properties. In this work the dielectric properties of relaxors disordered PMN-PSN ($Pb(Mg_{1/3}Nb_{2/3})O_3$-$Pb(Sc_{1/2}Nb_{1/2})O_3$), ALNBKT ($0.85Ag_{0.9}Li_{0.1}NbO_3$–$0.15Bi_{0.5}K_{0.5}TiO_3$), NKN ($Na_{1/2}K_{1/2}NbO_3$) ceramics, quantum ferroelectric $La_{1/3}NbO_3$ and ferroelectric ordered PMN-PSN ceramics were investigated by time domain THz spectroscopy in frequency 100 GHz – 3 THz and temperatures 300 K -10 K ranges. Measurements at THz frequencies from 200 GHz to 3 THz were performed in the transmission mode using a time-domain THz spectrometer based on a femtosecond laser system. The samples with apertures of 5 mm and 40 µm thick were investigated. A cryostat with thin teflon windows was used for measurements down to 10 K. The dielectric spectra were calculated from transmission spectra according to Fresnel formula. Below room temperature dielectric dispersion in all investigated relaxors is very broad and it is originating from polar nanoregions volume fluctuations. Only at very low temperatures evidence for resonant soft mode existence in investigated relaxors was presented. The resonant mode was described by

$$\varepsilon^* = \varepsilon_\infty \prod_j \frac{\omega_{LOj}^2 - \omega^2 + i\omega\gamma_{LOj}}{\omega_{TOj}^2 - \omega^2 + i\omega\gamma_{TOj}} \qquad (1)$$

where ω_{TOj} and γ_{Toj} denote the transverse optical (TO) and ω_{LOj} and γ_{LOj} the longitudinal optic (LO) j-th polar phonon mode frequency and damping, respectively; ε_∞ is the high-frequency permittivity originating from the electronic polarization and from the polar phonons above 3 THz. The critical temperature of this mode is close to Burns temperature [1, 2]. The resonant soft mode exist also in ferroelectric PMN-PSN ceramics. However, the critical temperature of these ceramics is close to ferroelectric phase transition temperature. Also in these ceramics relaxational soft mode was observed. Therefore, the nature of phase transition in these ceramics is mixed between displacive and order-disorder type. The polar clusters exist already in ordered PSN ceramics and their dynamics provides the origin of broad dielectric dispersion at low frequencies. The size of polar clusters decreases by PMN doping and, at higher PMN concentrations (for $x \le 0.6$) the dielectric dispersion is dominated by the dynamics of polar nanoregions. Nevertheless, the microscopic origin of ferroelectric phase transition in PMN-PSN ferroelectrics and microscopic origin of appearance of polar nanoregions in PSN-PMN ceramics is the same: softening of rigid BO_6 octahedra vibrations against Pb atoms. In quantum ferroelectric $La_{1/3}NbO_3$ was observed hardening of soft mode on cooling down to lowest temperatures.

FERROELASTIC PHASE TRANSITIONS IN MIXED KSc(MoO_4)$_2$ BASED TRIGONAL DOUBLE MOLYBDATES

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The influence of ionic substitution on the phase transitions in mixed $K_{1-x}Rb_xSc(MoO_4)_2$ double trigonal molybdates (x<or=0.2) is studied by EPR of Cr^{3+} probes and optical microscopy. The EPR results indicate that there is a distribution of environment for Cr^{3+} in the mixed crystals which results from the substitution of K^+ by Rb^+ ions. From the temperature dependence of the EPR spectra the critical temperature of the trigonal to monoclinic structural phase transition that appears in these crystals has been determined as a function of x. It has been found that this transition point shifts down with increasing x and the temperature range over which the trigonal phase exists tends to become greater. A critical exponent β has been found in the x = 0.03 and x = 0.1 samples. The ferroelastic domain structure of $K_{1-x}Rb_xSc(MoO_4)_2$ crystals has been observed and investigated in detail by means of a polarizing microscope.

ORDER-DISORDER PHENOMENA IN THE LOW-TEMPERATURE PHASE OF $BaTiO_3$

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Magnetic resonance is known to be a powerful tool for studying local ordering processes in materials showing ferroelectric, antiferroelectric and dipolar glass (proton glass) order or commensurably and incommensurably modulated phases [1 - 7]. Nuclear magnetic resonance (NMR) and electron paramagnetic resonance (EPR) can sensibly complement each other because of their different time windows of investigation. NMR is the more generally applicable technique for studying such materials because most of them embody nuclei with a non-zero nuclear spin. The application of EPR is much more restricted because dielectric crystals are usually not paramagnetic. However, very often the crystals can be doped with appropriate paramagnetic ions like e.g. the transition metal ions Cr^{3+}, Cr^{5+}, Mn^{2+}, Mn^{4+}, Fe^{3+}, Gd^{3+} or can be irradiated by X- or γ -rays in order to create paramagnetic probes. Sometimes, ENDOR (electron nuclear double resonance) gives still more detailed information. Dopants can of course always influence the dielectric properties under study. But as EPR is a very sensitive technique, the low concentration of paramagnetic centers ($< 10^{-2}$ at.%) needed for investigations is in many cases only slightly above the natural impurity concentration. The power of EPR spectroscopy for studying complex ordering processes has been demonstrated e.g. with the ^{Cr} EPR studies of Cr^{3+} doped dimethylammonium gallium sulfate hexa-hydrate (DMAGaS) $(CH_3)_2NH_2Ga(SO_4)_2 \cdot 6H_2O$ [3-6] and the isomorphous dimethyl-ammonium aluminum sulfate hexahydrate (DMAAS) $(CH_3)_2NH_2Al(SO_4)_2 \cdot 6H_2O$ and the ENDOR investigations of the proton glass betaine phosphite/phosphate [2, 6]. For these both examples NMR fails because the spectrum changes related to the relatively fast dipolar reorientations are averaged out for the NMR but not for the EPR experiment or because NMR spectra were not selective enough.

Here the problem of the coexistence of displacive and order-disorder phenomena at the phase transitions of $BaTiO_3$ shall be discussed which met growing interest in recent time [8-11]. X- and Q-band EPR measurements are reported on Mn^{4+} doped $BaTiO_3$ single crystals in the rhombohedral low-temperature phase [7]. The Mn^{4+} probe ion is statistically substituting the isovalent Ti^{4+} ions. The angular dependent Mn^{4+} EPR spectra were recorded for selected crystal planes in order to find appropriate directions for the temperature dependent measurements. At selected directions of the static magnetic field B_0, the temperature dependence of the Mn^{4+} EPR spectra was measured. A critical line broadening was detected in the rhombohedral phase when approaching the transition to the orthorhombic phase. This line broadening is interpreted as being the result of an order-disorder process within the off-center Ti subsystem. The observations indicate a model where microscopic clusters of the orthorhombic Ti off-center displacements exist already in the rhombohedral phase that transform into orthorhombic domains at the actual phase transition temperature. This behavior is convincingly confirmed by the agreement of the very characteristic experimental angular dependence of the line broadening with the simulation based on the model. From the data it is concluded that $BaTiO_3$ shows a special type of phase transition where displacive and order-disorder character do coexist not only at the cubic-tetragonal transition as shown by NMR [10] and terahertz dielectric response [11] but also at the orthorhombic-rhombohedral transition at low temperatures.

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ULTRASONIC INVESTIGATION OF PIEZOELECTRIC SENSITIVITY IN $CuInP_2S_6$ FAMILY LAYERED CRYSTALS

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The layered crystals of $CuInP_2S_6$ family are the promising materials for the applications in functional electronics, because they exhibit ferroelectric and semiconductor properties as well as high ionic conductivity. These crystals crystallize in a layered two-dimensional structure of the $Cu^IM^{III}P_2S_6$ (M=In, Cr,Bi) type. Recently the new crystals and solid solutions were obtained after substitution Cu to Ag, In to Cr ,Bi or S to Se. In this contribution we present the ultrasonic method for investigation of phase transitions and piezoelectric properties of these crystals and solid solutions. The ultrasonic pulse-echo method was applied for piezoelectric sensitivity detection. Piezoelectric response was measured when receiving ultrasonic transducer in conventional pulse-echo method was replaced by thin plate of layered crystal under investigation [1]. In this case the piezoelectric sensitivity anomalies corresponding to elastic anomalies were observed in phase transition region. This ultrasonic method allows also detecting piezoelectric sensitivity in nonpolar paraelectric phase in bias DC field due to electrostriction [2]. In pure $CuInP_2S_6$ crystals piezoelectric sensitivity exists below the first order ferroelectric phase transition at $T_c = 312$ K. After substitution In to Cr and S to Se phase transition temperature decreased and intermediate phases, possibly incommensurate, appeared above ferroelectric phase transition point. Substitution Cu to Ag leads to the shift down of ferroelectric phase transition and the transition type changes to the second order. In polarised layered ferroelectric crystals the piezoelectric effect was large enough, what promises possible ultrasonic transducer applications. In pure $AgInP_2S_6$ and $AgInP_2Se_6$ crystals we did not observe phase transitions and ferroelectricity in the temperature range of 100 - 300 K. However, in these crystals piezoelectric sensitivity was observed in bias electric field applied along c-axis. The influence of high ionic conductivity of these layered crystals on ultrasonic attenuation also is discussed.

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TUESDAY, SEPTEMBER 14, 2010

Poster presentations

THE TEMPERATURE DEPENDENCES OF THE FERROELECTRIC AND DIELECTRIC PROPERTIES OF PURE AND NICKEL DOPED SBN SINGLE CRYSTALS

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Relaxor ferroelectrics strontium barium niobates, SBN ($Sr_xBa_{1-x}Nb_2O_6$, $0.25 \leq x \leq 0.75$), are the important functional materials. They have interesting properties suitable for appliance in optoelectronics, namely their electro-optic and non-linear optical coefficients. They are also interesting from a theoretical point of view as relaxor ferroelectrics. Relaxors still remain a theoretical challenge since they reveal difficulties in finding suitable models to explain their properties.

The purpose of the paper is to provide additional evidence that the broad phase transition and frequency dispersion, that are exhibited by relaxor ferroelectrics such as SBN, have a strong link to the configurations of the ferroelectric microdomains.

Temperature dependencies of dielectric constants and switching properties of the SBN:61 pure and doped with Ni ions, were investigated from room temperature up to the phase transition temperature.

Our results show that broad transition has direct reflection in hysteresis loop behavior. Additionally, the observation of domain pattern evolution during polarization reversal in dc electric field, at room temperature, has been presented by means of the nematic liquid decoration technique.

HOPPING CONDUCTIVITY IN FERROELECTRIC $[Pb(Fe_{1/3}Sb_{2/3})Ti_yZr_z]O_3$

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Studies on direct current (DC) and altering current (AC) electrical conductivity in ferroelectric ceramics $[Pb(Fe_{1/3}Sb_{2/3})Ti_yZr_z]O_3$ with $x + y + z = 1$, $x = 1$ and $y = 0.44$ and 0.47 have been performed. The measurements were carried out in the range of temperatures from 77 K to 750 K and frequencies from 50 Hz to 100 kHz. AC conductivity $\sigma(\omega)$ fulfills the relation $\sigma(\omega) = A\omega^S$, where the exponent s depends on temperature T and frequency ω. The exponent s increases with decreasing temperature and in the vicinity of 77 K equals to 0.988 and 0.991 for sample with $y = 0.44$ and 0.47, respectively. At low temperatures, the temperature dependence of DC conduction $\sigma(0)$ is described by Mott's law $\sigma(0) \propto \exp[(T_0/T)^{1/4}]$. Parameter T_0 calculated from this relation equals to 4.51×10^6 K and 1.55×10^6 K for samples with $y = 0.47$ and 0.44, respectively. Knowing T_0 and estimating α^{-1}, the spatial extend of the localized wave function, as $\alpha^{-1} = 1.61$Å, the density of states at the Fermi level $N(E_F)$ was calculated as 1.51×10^{22} cm^{-3} eV^{-1} and 3.37×10^{22} cm^{-3} eV^{-1} for samples with $y = 0.44$ and 0.47, respectively. The most probable Mott hopping distance R and the hopping energy W were also determined; at 298 K, $R = 1.11$ nm and $W = 0.058$ eV for sample with $y = 0.47$, and $R = 1.35$ nm and $W = 0.071$eV for sample with $y = 0.44$. The density of states at the Fermi level calculated from DC and AC conductivities are of the same order and great values of s suggest that (i) hopping of carriers takes place over the potential barrier W and (ii) conductivity $\sigma(\omega)$ is due to the dipole polarization currents.

PRESSURE BEHAVIOUR OF THE BIREFRINGENCE OF IN $TlInS_2$ LAYERED CRYSTALS WITH INCOMMENSURATE STRUCTURE

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Layered ferroelectric are interesting for investigation since their characteristics and properties are determined by quasi-two-dimensionality and strong structural anisotropy. This class of materials includes $A^{III}B^{III}C^{VI}$ chalcogenides (A = Tl, B = In, Ga, C = S, Se), among which ferroelectric crystal $TlInS_2$ is especially interesting since it possesses an incommensurate phase in the interval 201–216 K. The studies of the (p, T)-diagram of this crystal give the evidence for the existence of a polycritical phenomena in the pressure range $p > 0.58$ GPa [1] . In the paper under consideration the results of the investigation of birefringence in $TlInS_2$ crystals along the p,T-diagram are given.

$TlInS_2$ single crystals were grown by Bridgman technique. The studies of birefringence $TlInS_2$ single crystal were carried out in the temperature interval 170–300 K and in the hydrostatic pressure range up to 0.58 GPa. The birefringence measurements were performed by Senarmont technique at the laser wavelengths of 650 nm and 532 nm. The measurements were carried out in a high-pressure optical cell.

At atmospheric pressure, in the temperature dependences of birefringence in the $TlInS_2$ crystal anomalies at T_i= 216 K and T_c= 193 K are observed corresponding to phase transitions into incommensurate and ferroelectric phases, respectively. With the pressure increase to 0.58 GPa the anomalies of the birefringence are observed to shift towards higher temperatures respectively to the phase diagram. The positive sign of the pressure-related shift of the temperatures of the anomalies means that the corresponding phase transformations can be treated as phase transitions of order/disorder type. The relevant pressure coefficients are determined.

The features of the observed birefringence anomalies and the pressure behaviour of birefringence in the incommensurate and commensurate phase are being discussed.

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MANIFESTATION OF STRUCTURAL DISORDER IN THE POLARIZATION KINETICS OF NON-UNIFORM FERROELECTRICS AND RELAXOR CRYSTALS

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The purpose of this report was to perform a comparative experimental analysis of the kinetics polarization in non-uniform $Te(OH)_6 2NH_4H_2PO_4(NH_4)_2HPO_4$ (TAAP) ferroelectrics and relaxor crystals $Sr_{0.61}Ba_{0.39}Nb_2O_6$ (SBN). Micro-scale studies of switching process in conjunction with electrical measurements allowed us to establish a relationship between local properties of domain dynamics and macroscopic response; polarization hysteresis loop and switching currents. We investigated the microscopic features of 180 degree domain walls dynamics and relate its dynamics to the defect structure in both type of the crystals. The domain structure was studied by means of the nematic liquid crystal decoration method.

The switching process has two stages in both type of the crystals: a fast nucleation stage and a slow sidewise motion of domain walls. The fast stage has a threshold nucleation field and corresponds to the forward growth of domains. The relaxation of polarization during the switching process are qualitatively similar in both type of crystals, and can be described by a stretched exponential function ęxp(-t/t0)^n where 0<n<1 , widely used in the dielectric relaxation studies[1] .The exponent n is associated with a distribution of activation energy of nucleation and domain growth . However, the microscopic features of domain wall dynamics, related to the defect structure differs considerably in both type of the crystals. The stabilization mechanism of the domain structure appears to have a correlation to a defect induced internal bias field ,Eb, in non-uniform TAAP crystals . These crystals are highly inhomogeneous; regions exist that exhibit switchable polarization and there are parts with more fixed polarization. In SBN relaxor crystals very strong pinning forces , due to quenched random fields are active , which give rise to decelerated dynamics of domain walls, forming a maze type domain pattern.

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INFLUENCE OF VANADIUM DOPANT ON DIELECTRIC PROPERTIES OF $BaBi_2Nb_2O_9$ CERAMICS

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Ferroelectric materials with Bi-layered structure such as $SrBi_2Ta_2O_9$ and $SrBi_2Nb_2O_9$ are now intensively investigated in view of their application in non-volatile computer memories and high temperature piezoelectric transducers. The most useful aspect of these materials are their high fatigue resistance against polarization switching. When Sr is submitted with Ba ions (BBN), structural disorder is induced and material exhibits significant broadening of the phase transition. The causes of this diffuse phase transition were widely discussed in the following papers [1-3]. A wide range of applications required materials with not only good dielectric properties, but also high density and good grain structure. Based on literature information [4,5] the very promising seems to be vanadium dopant. In case of SBN ceramics the dopant remarkably improved not only the quality of ceramics, but also the dielectric properties. In presented work we described influence of V dopant not only on dielectric properties, but also on lattice parameter and grain structure.

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IMPEDANCE SPECTROSCOPY IN $BaBi_2Nb_2O_9$ CERAMICS

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The classical ceramics based on lead titanate zirconate (PZT) have been extensively used in the electronic industry for its high dielectric constant, including the fabrication of multilayer ceramic capacitors, piezoelectric transducers and ferroelectric memories. However the applications are limited by ageing and retention of polarization. It was main reason, why the other, led free ceramics have generated much interest recently. Very promising materials due to their multi-functionality seem to be so called Bi-layered perovskites-firstly sinthesized by Aurivillius. The best known representatives of this family are $SrBi_2Ta_2O_9$,(SBT), $SrBi_2Nb_2O_9$, (SBN) and $Ba_{1-x}Sr_xBi_2Nb_2O_9$, (BSBN).

Investigations presented in the work concerning the other member of the Aurivillius family, the $BaBi_2Nb_2O_9$ (BBN), the compound which seems to be very interesting from the point of view of the potential applications. The dielectric properties of those materials and influence of sintering condition on grain structure and properties of those materials were very widely described in our previous paper. Now we would like to present the hitherto unraported electric properties of $BaBi_2Nb_2O_9$ ceramics, investigated by using the impedance spectroscopy (IS). IS is nondestructive method, which allows us to separate grain and grain – boundary effects due to the different frequencies of the relaxation process in grain and grain boundaries. This enables us to determine the temperature dependences of grain (R_g) and grain-boundary (R_{gb}) resistance and evaluate their respective activation energies (E_a). Additionally the attempts to compare macroscopic results obtained by Impedance Spectroscopy and direct local LC-AFM conductivity measurements were taken.

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CERAMICS WITH TETRAGONAL TUNGSTEN BRONZE TYPE STRUCTURE FOR TEXTURED CERAMICS-POLYMER COMPOSITES

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For piezoelectric sensors maximization of piezoelectric properties is highly desirable. It is difficult to achieve high piezoelectric response in randomly oriented polycrystalline materials. For this reason textured piezoelectric materials: ceramics and ceramics-polymer composites are of special interest. To develop textured ceramics it is important to choose materials which grow anisotropically. Ceramic materials with asymmetric unit cells often grow anisotropically. The tetragonal tungsten bronze type structure (TTB) family is an example of system in which fiber texture can be achieved. TTB type ferroelectrics are useful for electro-optic, piezoelectric, pyroelectric, millimeter wave, and photorefractive applications.

In this work textured $Sr_{0.7}Ba_{0.3}Nb_2O_6$-PVDF (SBN70-PVDF) composites were fabricated. Strontium barium niobate ($Sr_xBa_{1-x}Nb_2O_6$ - SBN) in the range $0.25<x<0.75$ is a ferroelectric material with tetragonal tungsten bronze type structure. Powders of strontium barium niobate $Sr_{0.7}Ba_{0.3}Nb_2O_6$ (SBN70) were prepared by wet-chemical technique. The SBN70-PVDF textured composites samples were fabricated by hot-pressing method. The composite surface images were obtained by AFM tapping mode (NT-MDT Solver P47).

QUALITY OF TGS SAMPLES SURFACE INVESTIGATED BY AFM

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Dielectric measurements of ferroelectric materials require plane-parallel samples, whose opposite surfaces are covered with electrodes, which creates a capacitor. Method of samples preparation, especially of the surface which are to be covered with electrodes, depends on the kind of material tested (monocrystal, ceramics).

The measurements carried out concern samples of TGS monocrystal. In this case we apply one of the basic methods of processing:

- crystal cleavage along the cleavage plane of the monocrystal,
- mechanical cutting the sample out of a big crystal and polishing the surface which is to be covered with electrodes.

Because the samples are prepared of crystals growing from water solutions, they should be rejuvenated before measurements. This process can be carried out before, or after the electrodes are fitted.

Each of the described methods of samples preparation is a cause of their surfaces aimed to be covered with electrodes different qualities.

For the investigations of TGS samples surfaces, atomic force microscope (P47 NT-MDT) was used.

PECULIARITY OF THE PHASE TRANSITIONS OF $Na_{0.5}Bi_{0.5}TiO_3$ CRYSTALS

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$Na_{0.5}Bi_{0.5}TiO_3$ (NBT) is relaxor ferroelectric with $A'_{1-x}A''_xBO_3$ -type structure. The cations distribution and ordering in the corresponding sublattices influence on the physical properties of this material.

It is well known that NBT has cubic structure (m3m) above 520°C. Below this temperature NBT has tetragonal nonpolar structure (4/mmm). And at last below 200°C up to room temperature the rhombohedral structure (3m) is realized. There are too many opinions about the nature and physical properties behavior in the temperature region 200°C – 320°C.

On the base of our dielectric and optical measurements and the analysis of literature data we want to propose the succession of phase transitions in NBT:

m3m – (520 °C, second order) – 4/mmm – (320 °C, second order) – 4mm – (240°C at cooling and 280-290 °C at heating, first order) – 3m.

What is the base of this approach? The phase transition 4/mmm – 4mm within the same syngony do not contradict the X-ray data and may explain a)the existing of very strong peak of dielectric permittivity and b) the disappearance of piezoelectric properties and second harmonic generation near 320°C [1]. As for transition 4mm – 3m the main argument is the presence of “isotropization” point at 240°C (280 -290°C) and the existing domain walls [100] type below this temperature only.

The possibility of existing orthorhombic phase that is typical for the perovskite structure is discussed.

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THE FERROELECTRIC PHASE TRANSITIONS IN THE POLYTYPES OF $TlGaSe_2$ CRYSTALS

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Many researchers investigated structural phase transitions (PT) in ternary-layered crystal $TlGaSe_2$ by using a great number of experimental methods. The results of comparative X-rays researches and measurements of dielectric permittivity of C-$TlGaSe_2$ and 2C-$TlGaSe_2$ polytypes in the temperature range T=100-300 K are presented now. Much attention was devoted to changes with the temperature of angular position, shape and number of (*00l*)-type reflections, because the PT at 245 K can be caused to the appearance of incommensurate modulated structure with the large period of modulation along C-axis. By the method of X-rays dilatometry it was shown, that temperature dependence of the unit cell parameter *c*(T) for C-$TlGaSe_2$ polytype in the temperature range T=140-300 K does not reveal hysteresis and is linear. For this polytype the form of (00l)-type reflections does not change with temperature. These results confirm absence of PT for C-$TlGaSe_2$ polytype at the mentioned temperatures. At the same time for 2C-$TlGaSe_2$ polytype at the temperatures T< 210 K dependence *c*(T) is nonlinear and the angle of dependence slope is increased with temperature drop. The changes of the (00.28) reflex shape of such polytype for the temperatures range below 240 K were revealed. The computer analysis of the indicated form reflex changes with the temperature allowed to suppose that the reason of form distortions can be the presence of modulated satellities. Angular position of such over structural reflexes give evidence about existence in crystals of 2C-$TlGaSe_2$ polytype below temperature 240 K incommensurate modulated structure. Modulation wave-length of that incommensurate structure along C-axis considerably exceeds unit cell parameter *c*. The kink, discovered at 245 K on temperature dependence of dielectric permittivity of 2C-$TlGaSe_2$ polytype, confirms X-rays structure information about an appearance of incommensurate phase at this temperature. If take into account the features of ferroelectric PT in the two different polytypes of $TlGaSe_2$ crystals, that was reported by us earlier, the our experimental results give evidence the crystals of C-$TlGaSe_2$ polytype belong to the ferroelectrics with an incommensurate phase of type I, and the crystals of 2C-$TlGaSe_2$ polytype belong to another type – to the type II.

EFFECT OF TANTALUM CONTENT ON SYNTHESIS OF LEAD-FREE PIEZOELECTRIC KNN CERAMICS

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Ceramics with a series of compositions $(K_{0.5}Na_{0.5})(Nb_{1-x}Ta_x)O_3$+0.5mol%$MnO_2$ (KNNTx) (x=0÷8) were produced by conventional ceramic processing method. XRD analysis revealed pure perovskite structure; no evidence of secondary phase was detected. It has been found that sintering densification for KNN ceramics occurs within a narrow temperature range. MnO_2 functions as a sintering aid and effectively improves the densification process at the same time homogenizing the microstructure of ceramics sample. From dielectric measurements it can be seen that Ta^{5+} substitution for Nb^{5+} decreased the phase transition temperatures (T_c for KNNT5 is 390°C). The T_c variations caused by Ta^{5+} ions substitution for Nb^{5+} may be associated with the occupation of B-site of the perovskite structure. This may be the reason also for the broadening of peaks which is observed when substitution level is increased.

STRUCTURAL, DIELECTRIC AND XPS STUDIES OF $BiFeO_3$ CERAMICS OBTAINED FROM MECHANOCEMICALLY SYNTHESIZED NANOPOWDER

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Bismuth ferrite $BiFeO_3$ has recently gained considerable interest due to the existence of both ferroelectric and antiferromagnetic ordering at room temperature (T_N~640K, T_C~1100K). Though the coupling between magnetization and dielectric polarization is not high the material is still considered for potential applications in electrically controlled magnetic data storage and spintronic devices [1].

$BiFeO_3$ nanopowder was synthesized from commercially available oxides Bi_2O_3 and Fe_2O_3 (Aldrich, 99% purity) weighted in 2**:**1 ratio. The starting oxides were milled for 7 h in Fritsch Pulverisette 6 planetary ball mill under normal pressure. The calculated parameters [2] of the high energy milling were the following: the kinetic energy per hit amounted to 300 mJ/hit and the shock frequency to 100 Hz. The obtained nanopowder was hot-pressed at 800^0C under pressure of 200 MPa during 2 h. The synthesis of $BiFeO_3$ was control by X-ray powder diffraction studies (X'Pert-PANalytical diffractometer) [Fig. 1]. The structure was found to be trigonal ($R3c$). The mean grain size calculated according to the Scherer formula was 26 nm after milling and 64 nm after hot – pressing. Dielectric response (1 Hz–1 MHz) of the ceramics was measured using Alpha-A High Performance Frequency Analyzer (Novocontrol GmbH) in the temperature range 120-575 K. Dielectric relaxation behaviour of $BiFeO_3$ ceramics was very similar to that observed in the giant dielectric constant materials such as $Ba(Fe_{1/2}Nb_{1/2})O_3$, $Sr(Fe_{1/2}Nb_{1/2})O_3$ and $Cu_3CaTi_4O_{12}$ [3]. Two relaxation processes observed in the temperature ranges 220 – 370 K and 370 – 470 K can be attributed to the carrier hopping process between the mixed valences Fe^{2+} and Fe^{3+} (fig. 2) and to the grain boundary effect, respectively. The XPS spectra (VG Scienta System) of the samples revealed the presence of Fe^{2+} in addition to the Fe^{3+} valence.

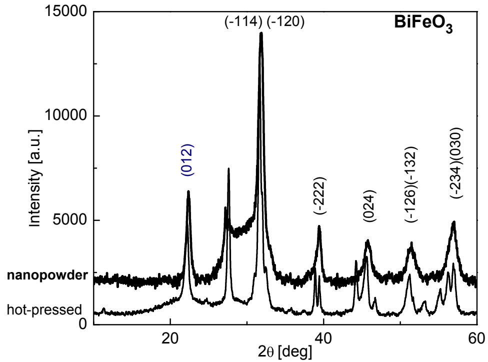

Fig. 1. XRD pattern of $BiFeO_3$ nanopowder and ceramics

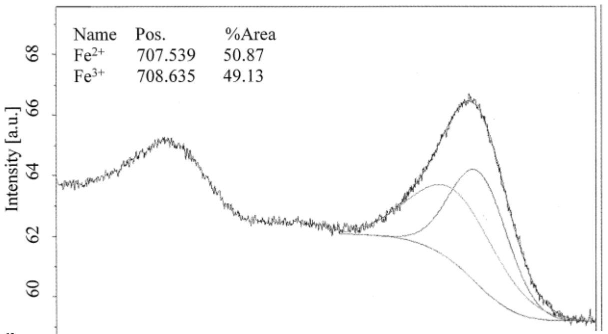

Fig. 2. XPS spectrum in Fe2p region.

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RESTRICTED DOMAIN GROWTH AND SWITCHING KINETICS IN STRONTIUM BARIUM NIOBATE CRYSTALS

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In this report we investigate the domain structure and switching process in pure and Ni doped $Sr_{0.61}Ba_{0.39}Nb_2O_6$ relaxor ferroelectric crystals. We present real-time studies of the nucleation and growth processes by nematic liquid crystal (NLC) decoration technique. NLC method involves averaging over macroscopic scale. This allows us to correlate the domain structure dynamics of relaxor crystals with macroscopic characterization techniques such as switching current and hysteresis loop registration. The observed complexity of the domain structure could be understood in terms of the slow inhomogeneous domain growth in the presence of random pinning fields in relaxor crystals. In such crystals fast growing domains coexist with slower ones, corresponding to different local barrier heights, giving rise to suppression of the switching process. The experimental investigation were undertaken to explain why classical nucleation and growth model (Kolmogorov-Avrami- Ishibashi model) is incapable in the case of SBN relaxor crystals, especially at low electric field range.

AFM OBSERVATIONS OF THE DOMAIN STRUCTURE IN THIN FILMS OF $BiFeO_3$

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Bilayered epitaxial $BiFeO_3/YBa_2Cu_3O_7$ films were fabricated on (100) $(LaAlO_3)_{0.3}(Sr_2TaAlO_6)_{0.7}$ – (LSAT) substrates by sputtering method, [1].

For structural comparison the bilayered $BiFeO_3/La_{0.67}Sr_{0.33}MnO_3$ – (LSMO) films were also deposited on (100) $SrTiO_3$ substrates. The perovskite $BiFeO_3$ is ferroelectric (T_C = 1103 K) and antiferromagnetic (T_N = 643 K). In single crystals, the spontaneous polarization (P_S) is 3.5 $\mu C/cm^2$ along the (001) direction, achieving a value of 6.1 $\mu C/cm^2$ along the (111) direction at 77 K. For thin films the remanent polarization value is an order of magnitude higher than the highest reported value for the bulk crystal. The heterostructures prepared at the Institute of Physics P.A.S. were studied by various AFM techniques to reveal ferroic properties. Structure of layers was similar to presented in the Fig.1. Domain structure observations and its transformation have been performed by piezoresponse technique. We have also recorded features indicating presence of the magnetic domain structure by magnetic force method. Domain widths were estimated and compared with a model of Chen et al. [2].

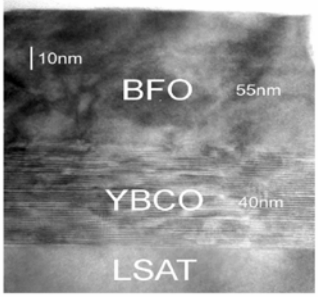

Fig.1. TEM crosssection of the BFO/YBCO bilayer deposited on LSAT substrate, [1].

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THE STRUCTURE AND EVOLUTION OF NANODOMAINS IN (1-x)$PbMg_{1/3}Nb_{2/3}O_3$-(x)$PbTiO_3$ CERAMICS VISUALIZED BY PIEZORESPONSE FORCE MICROSCOPY

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Lead magnesium niobate – lead titanate (PMN-PT) solid solutions nowadays are intensively studied by different techniques mainly due to their excellent electromechanical properties [1]. These properties of PMN-PT are known to be mainly influenced by small nanometer-size polarized areas, called polar nanoregions (PNRs). Due to small size, PNRs cannot be detected by conventional optical microscopy. However, the development of piezoresponse force microscopy (PFM) enables the direct observation of PNRs with sizes up to several nanometers. Up to now only few of such studies have been performed for (1-x)$PbMg_{1/3}Nb_{2/3}O_3$-(x)$PbTiO_3$ single crystals, and a complex pattern of nanodomain structure (existing even above the structural transition temperature) has been found for 0.9PMN-0.1PT single crystal [2]. A preferable orientation of PNRs along (110) crystallographic direction with the mean PNRs size of about 70 nm was determined by autocorrelation technique of PFM. This work is the extension of previous studies of PMN-PT single crystals by PFM studies of their ceramics.

The PFM investigations of (1-x)PMN-(x)PT (x=0.08, 0.1, 0.12) ceramics have been performed using commercial atomic force microscope (Multimode, Veeco). The out-of- plane piezoresponse images revealed a labyrinth-like structure of polarized areas with the mean PNRs size decreasing with increase of x. The increase of temperature up to 373 K did not destroy the PNRs structure, but lowered the piezoresponse contrast. The studies of induced polarization decay revealed non-exponential character of obtained curves. The fits by two-exponent function demonstrated that the relaxation rate increases with increase of x.

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INFRARED AND TERAHERTZ STUDIES OF MULTIFERROIC $Bi_{0.85}Nd_{0.15}FeO_3$ CERAMICS

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Sarah Karimi[3] and Ian M. Reaney[3]

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Bismuth ferrite $BiFeO_3$ (BFO) belongs to a scarce class of multiferroic magnetoelectric materials, exhibiting ferroelectric and ferromagnetic ordering simultaneously. At room temperature, pure bismuth ferrite is of rhombohedral *R3c* [1] symmetry. Levin *et al*. have carried out a very thorough structure investigation of neodymium-doped BFO [2][3]. They conclude that for the Nd-doping > ~14 mol% the system undergoes an orthorhombic Pbam (antiferroelectric) ↔ orthorhombic Pbmn (paraelectric) phase transition, instead of rhombohedral R3c (ferroelectric) ↔ orthorhombic Pbmn (paraelectric) one in pure or only slightly doped BFO.

We will present far-infrared and terahertz spectra, revealing the lattice dynamics and intrinsic permittivity. A strong central peak emerges close to the T_C. It is also clear that the modes of neodymium-doped BFO are damped more heavily compared to the ones of pure bismuth ferrite ceramics [4], especially at low temperatures. We were able to resolve eighteen modes at 10 K, although many of them are not expressed very well. As the damping increases on heating, only about ten modes can be clearly distinguished at temperatures slightly below the phase transition at ~600 K [5]. At higher temperatures, the number of modes reduces further to six, and this points out that the structure of the paraelectric phase is not cubic, as in such case only three polar modes would be permitted by symmetry. It is still an open question if the neodymium-doped bismuth ferrite undergoes another phase transition to the pure cubic phase.

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NOISE INVESTIGATIONS OF THE MULTIFEROIC COMPSITE $BaTiO_3$ WITH $CoFe_2O_4$

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Ferroelectromagnets, which display simultaneous magnetic and electric ordering, have recently stimulated much scientific and technological interest. The coexistence of magnetic and electric subsystems engenders the material with the "product" property (i.e., the composite exhibits responses that are not available in the individual component phases), thus allowing an additional degree of freedom in the design of actuators, transducers, and storage devices. However, the choice of single-phase materials exhibiting coexistence of strong ferro/ferrimagnetism and ferroelectricity is limited. Van Suchtelen [1] proposed that composites of piezoelectric and magnetostrictive phases can be electromagnetically coupled via stress mediation. Subsequent theoretical and experimental work has focused on bulk ceramics. In present research, composite multiferroic materials having a combination of two or more materials with ferroelectric (e.g., $BaTiO_3$ and $PbTiO_3$), ferromagnetic (e.g., $CoFe_2O_4$ and $NiFe_2O_4$) and other properties have drawn tremendous attention. It is well know that $BaTiO_3$ is an excellent ferroelectric material. It has been used in capacitors for half a century due to its remarkable ferroelectric and piezoelectric properties. Among ferromagnetic materials, $CoFe_2O_4$ is also a well-known hard magnetic material owning to its magnetic properties such as high coercivity and moderate magnetization. Therefore, $CoFe_2O_4$-$BaTiO_3$ composite system shows good ferromagnetic, ferroelectric properties, magnetoelectric response and has been studied widely both in theory and experiment.

It is well known that low frequency noise characteristic investigation enables clearing up physical processes in various materials. The aim of the present work was to investigate noise characteristics in the phase transition region of the composites.

The voltage noise spectral density was measured under continuous wave operation in a frequency range from 20 Hz to 20 kHz. The measured noise signal was processed by low-noise amplifier, filter system and analog digital converter (National Instrument^{TM} PCI 6115 board). The noise spectrum density, S_V, was calculated using the standard Cooley–Tukey fast Fourier transform algorithm and evaluated comparing with the thermal noise of standard resistor R_e:

$$S_V = \frac{\overline{V^2} - \overline{V_s^2}}{\overline{V_e^2} - \overline{V_s^2}} 4kT_0R_e, \ [V^2s];$$

where $\overline{V^2}$, $\overline{V_s^2}$ and $\overline{V_e^2}$ are respectively the sample, the measuring system, and the standard resistor thermal noise variances in the narrow frequency band Δf; T_0 is the absolute temperature of the standard resistor.

The obtained results are discussed in terms of the phase transitions of $BaTiO_3$.

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Poster presentations
TUESDAY, SEPTEMBER 14

DIELECTRIC AND DILATOMETRIC INVESTIGATIONS OF THE SIZE EFFECT IN $NH_4H_2PO_4$ – POROUS GLASS COMPOSITIES

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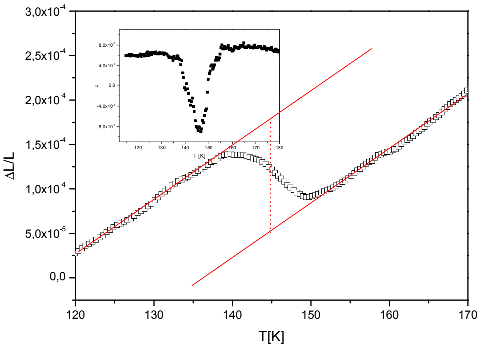
The results of the dielectric and dilatometric investigations of the ADP-porous glasses nanocomposites are presented. On the basis of dielectric and dilatometric measurements it was shown that in the composites ADP-porous glasses with the mean values of pores radius 160nm and 23 nm the phase transition similar to that observed in bulk ADP crystals occurs. It was found that decreasing of average pores size does not change the order but causes smearing of the phase transition.

It was found that the phase transition in ADP crystals embedded in porous glass decreases with decreasing of mean value of pores size. Experimentally observed shift of phase transition temperature is the superposition of the size effect and pressure effect caused by negative pressure created by the difference of thermal expansion coefficients of ADP nanoparticles and glass matrix and spontaneous deformations of nanocrystals at the phase transition temperature.

Dielectric measurements performed in wide frequency range showed low frequency dispersion in phase transition region. It's worth to notice that this kind of dispersion has not observed neither in bulk ADP nor in porous glasses. The low frequency dispersion is probably connected with ionic conductivity caused by proton motion.



Temperature dependence of the relative thermal deformation *ΔL/L*, and thermal expansion coefficient *α* (in the insert) of ADP-PG160. The approximated spontaneous deformation of the sample is marked by the straight line.

BROADBAND DIELECTRIC INVESTIGATION OF SODIUM POTASSIUM NIOBATE CERAMICS WITH 8% ANTIMONY SUBSTITUTION

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Andris Sternberg[2], Karlis Bormanis[2]

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Lead containing ceramics like $Pb(Zr_xTi_{1-x})O_3$ (PZT) or $Pb(Mg_{1/3}Nb_{2/3})O_3$ (PMN) have excellent piezoelectric properties and are widely used in sensors, transducers and other micro and macro electromechanical devices. However it is desirable to use lead-free materials for environmental protection, lead-containing materials should be replaced with lead-free materials[1]. One of such ceramic potentially could be sodium potassium niobate $K_xNa_{1-x}NbO_3$ (KNN) near morphotropic phase boundary (MPB) which is at about 50% K and 50% Na for KNN. The main problem with KNN is sintering: it is difficult to obtain well-sintered KNN ceramic using an ordinary sintering process because of the high volatility of alkaline elements at high temperatures[2].

In this work we present dielectric permittivity results of KNN ceramics doped antimony $(K_{0.5}Na_{0.5})(Nb_{0.92}Sb_{0.08})O_3$ with sintering aid MnO_2 (0.5 mol. % MnO_2). Its dielectric properties was investigated in broad frequency (from 20 Hz to 35 GHz) and temperature (from 125 K to 500 K) range.

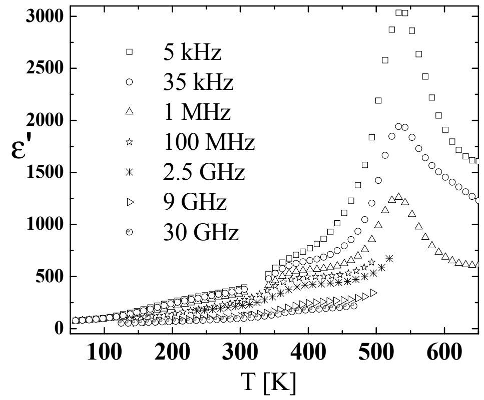

Figure 1 Temperature dependence of dielectric permittivity of $(K_{0.5}Na_{0.5})(Nb_{0.92}Sb_{0.02})O_3$ ceramic at fixed frequencies.

Above room temperature obtained results shows two phase transitions at T= 538 K and T=385 K which could be attributed to phase transitions from paraelectric cubic into ferroelectric tetragonal state and from tetragonal into orthorhombic state because in undoped $K_{0.5}Na_{0.5}O_3$ ceramics it was observed the same sequence of transitions with Tc=677 K and $T_{T\text{-}O}$=464 K respectively[3]. Partial substitution of Nb^{+5} ions with Sb^{+5} lowers both of these phase transitions temperatures compared to undoped KNN ceramics. Below room temperature investigation revealed one more dielectric permittivity dispersion, which could be attributed to diffused phase transition corresponding to phase transition in pure KNN from orthorhombic into rhombohedral ferroelectric state at $T_{O\text{-}R}$=165 K[3]. Below 165 K there is a dielectric dispersion which is similar to the relaxor ferroelectric from the real part of dielectric permittivity frequency dependence, what is confirmed by linear dependence of dielectric losses.

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DIELECTRIC AND CONDUCTIVE PROPERTIES OF HYDROTALCITE

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Hydrotalcite (HT) type materials are made of positively charged two-dimensional sheets of mixed hydroxides with water and exchangeable charge-compensating anions. The general formula for the HT materials is $[M^{II}_{1-x} M^{III}_{x} (OH)_2]^{x+}(A^{m-})_{x/m}\cdot nH_2O$, where M are bivalent or trivalent cations with similar radii, and A is an interlayer anion, usually CO_3^{2-}. The crystal structure of hydrotalcite consists of octahedral brucite ($Mg(OH)_2$) like layers. The interlayer space between each brucite sheets is filled with water and anions that compensate the positively charged layers. This region is highly disordered and the amount of water is a function of the temperature, of the water vapour pressure and of the nature of the anions present [1]. The nature of the layer cations can be changed using a wide range of main group (e.g., Mg, Al) or transition metal (e.g., V, Cr, Mn, Fe, Co, Ni) cations [2].

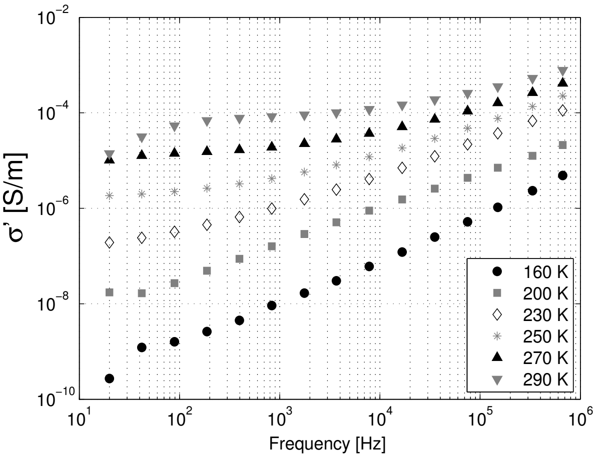

Fig. 1: Conductivity frequency dependence

The aim of the present work was to study classic type hydrotalcite with M^{II}=Mg, M^{III}=Al, x=0.25 and A^{m-} =CO_3^{2-} by means of dielectric spectroscopy. Dielectric properties of hydrotalcite dependent heavily on amount of water, thus two types of samples were studied – dry samples in vacuum and samples, which spent 16 hours in distilled water.

It can be clearly seen from figure 1, that there is quite high conductivity, which can be a result of water-introduced proton hopping as well as water molecule migration. Moreover, a DC component of conductivity is observed, which is different from [3].

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DIELECTRIC INVESTIGATION OF $PbFe_{0.5}Ta_{0.5}O_3$ CERAMICS

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$PbFe_{0.5}Ta_{0.5}O_3$ (PFT) ceramics are compounds belonging to the lead based complex perovskites with general formula $Pb(B'_{0.5}B''_{0.5})O_3$. Lead iron tantalite was first synthesized by Smolenskii *et al* in 1959 [1]. These relaxor ferroelectrics are receiving much attention due to their excellent dielectric properties [2-4]. PFT has a disordered perovskite- type structure and exhibits the diffuse paralelectric- ferroelectric transition at about 243 K [5].

Dielectric and conductivity measurements of $PbFe_{0.5}Ta_{0.5}O_3$ ceramics in the temperature range of 300 – 450 K have been reported. Ceramics exhibit strong low-frequency dispersion of dielectric permittivity above room temperature (Fig. 1.) which is caused by dipole relaxational and overlapping charge carrier migrational components. The activation energies of 0.78eV and 0.96eV were obtained for relaxation and conducting mechanism, respectively.

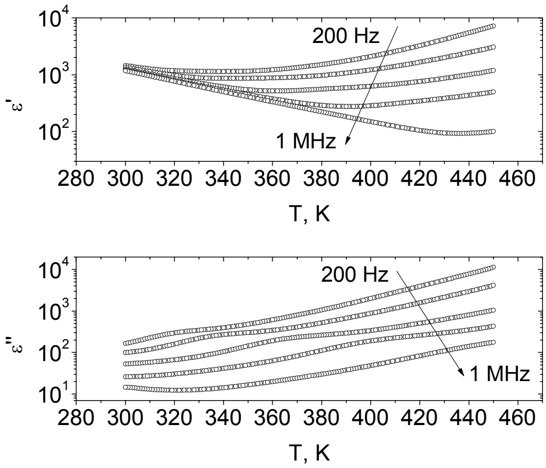

Fig. 1. Temperature dependencies of real and imaginary parts of dielectric permittivity of $PbFe_{0.5}Ta_{0.5}O_3$ ceramics

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DIELECTRIC AND IMPEDANCE SPECTROSCOPY OF xNBT–(1-x)LMT CERAMICS

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Lead-free ceramics based on $(Na_{0.5}Bi_{0.5})TiO_3$ (NBT) are of an increasing interest due to high dielectric permittivity and high Curie temperature value (up to 790 K [1]), making these materials promising in a wide spectrum of applications including those in microwave range. Furthermore, there are evidences of good piezoelectric properties of the NBT-based ceramics. Permittivity peak is diffuse and frequency-dependent, which means possibility of adjusting peak temperature when substituting NBT with another compositin. However, pure NBT ceramics exhibit high dielectric losses due to high conductivity.

$La(Mg_{0.5}Ti_{0.5})O_3$ (LMT) is a low-loss dielectric ceramic, which becomes a very promising substituent, because it reduces losses of NBT–LMT solid solution dramatically [2].

In this work, ceramics with x equal to 0.80, 0.85, 0.90 and 0.95 were studied in 300 – 800 K temperature range and 100 Hz – 1 GHz frequency band.

With addition of LMT ferroelectric phase transition shifts to lower temperatures. Furthermore, addition of LMT decreases activation energy of DC conductivity, which correlates well with [3] and could be related to increase of concentration of La.

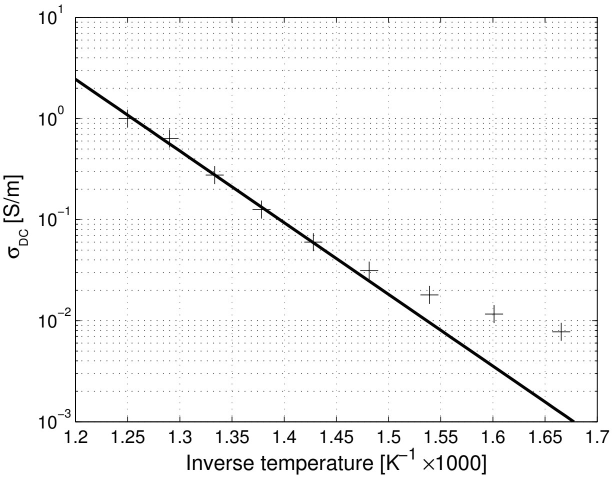

Fig. 1: Inverse temperature dependence of DC conductivity component when x=0.95

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MICROWAVE DIELECTRIC INVESTIGATIONS OF THE 0.6BaTiO$_3$–0.4(Ni,Zn)Fe$_2$O$_4$ MULTIFERROIC COMPOSITES

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The multiferroics are materials combining several properties in the same structure, such as electrical polarisation and magnetisation. The study of multiferroics is one of the most active field of the material science in the last years. Magnetoelectric composite materials have been made by sintering a mixture of a piezoelectric and magnetic phases.

Dielectric measurements of the 0,6BaTiO$_3$–0,4(Ni,Zn)Fe$_2$O$_4$ (0,6BT-0,4NZF) in the temperature range of 150 K to 420K show three phase transitions, which temperatures almost matches those of bulk barium titanate.

Dielectric measurements in the frequency range of 100 Hz to 40 GHz show pronounced volume conductivity at lower frequencies range (lower than 100 kHz.). At frequencies higher than 100 kHz, the conductivity is practically eliminated and the fundamental dielectric dispersion can be observed.

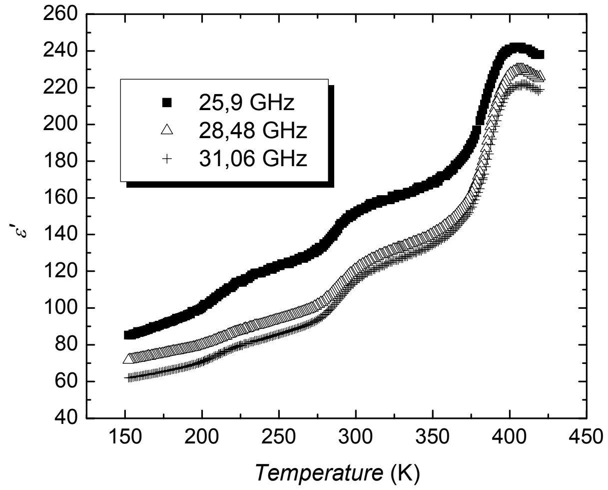

Fig.1 Temperature dependence of the real part of dielectric permittivity of 0.6BaTiO$_3$–0.4(Ni,Zn)Fe$_2$O$_4$ in microwaves

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XPS SPECTROSCOPY OF IMPURITIES INFLUENCE ON ELECTRONIC STRUCTURE OF $Sn_2P_2S_6$ FERROELECTRICS

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Tin hypothiodiphosphates ($Sn_2P_2S_6$) are known as ferroelectric–semiconductor crystals photosensitive in the red and near-infrared spectral ranges, characterized by a relatively narrow band gap (2.3 eV) and high electro-optic coefficients. Their fast photorefractive response together with high nonlinear optical properties makes them attractive as photorefractive media. The photorefractive properties of $Sn_2P_2S_6$ crystals can be changed by introducing of different dopants. In this report the results of experimental, by X-ray photoelectron spectroscopy (XPS), and theoretical investigations of electronic structure of ferroelectric $Sn_2P_2S_6$ crystals with In, Sb, Bi, Te, Pb and Ge impurities will be presented.

Doped $Sn_2P_2S_6$ crystals were obtained by the vapor transport technique and by the Bridgman method. The XPS of valence band and principal core levels were measured using a PHI 5700/660 Physical Electronics Spectrometer with monochromatic Al Kα radiation. The electron spectra were calculated in the framework of local density approximation (LDA) of Density Functional theory implemented in SIESTA software package. The calculations of doped crystals electron properties were performed using 1x1x2 supercell in order to reduce concentration of impurities.

It was determined influence of impurities on the electronic structure of the valence band and the shifts in the core level binding energies. The donor and acceptor levels for different impurities were found in the energy gap. These data are used for explanation of electric conduction, dielectric, optic and photorefractive properties of doped $Sn_2P_2S_6$ crystals.

HIGH-FREQUENCY DIELECTRIC PROPERTIES OF $Pb(Fe_{1/2}Nb_{1/2})O_3$ THIN FILMS GROWN BY PULSED LASER DEPOSITION

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Due to its structural simplicity and magnetoelectric coupling between the ferroelectric and antiferroelectric orders, lead iron niobate, $Pb(Fe_{1/2}Nb_{1/2})O_3$ or PFN, is considered a prototype multiferroic material. Increasing number of the dielectric studies on PFN bulk ceramics is performed, and some of which have showed that the nature of magnetoelectric coupling arises indirectly via ferroelestic contribution. We report results on investigation of the dielectric properties of the thin films of PFN in the wide frequency range.

The epitaxial quality thin films were grown on (001) $SrTiO_3$ substrate buffered by highly conductive $SrRuO_3$ layer. Structure and phase formation was confirmed using XRD. Gold film thermally evaporated on a top of the thin film were patterned by applying optical lithography processes to prepare an electrodes used for impedance measurements.

Measurements have been made on 240 nm thin films in the ferroelectric phase around room temperature at frequencies up to 10 GHz. Dielectric spectra have shown two-stage dispersion, below and above 100 MHz frequency. At that frequency, at room temperature, dielectric permittivity in the PFN thin film has value of 800. Temperature-dependent parameters of the dielectric relaxation were calculated. Also, voltage tunabilty data were obtained at room temperature for low and high frequency regions of the microwave spectrum investigated.

ULTRA LOW FREQUENCY DIELECTRIC RESPONCE MEASUREMENTS OF $PbMg_{1/3}Nb_{2/3}O_3$ RELAXOR FERROELECTRICS: SINGLE CRYSTAL AND CERAMICS

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Understanding the exceptional physical properties of relaxor ferroelectrics has been a real challenge since their discovery in the 1950s [1,2]. The recent growth of interest in materials exhibiting relaxor behaviour is to some extent due to findings pointing to the existence of a nonergodic structural glassy state below a certain temperature.

Relaxation processes in many electric, magnetic, mechanical and other systems are governed by Kohlrausch-Williams-Watts law, $P \propto \exp[-(t/\tau)^{\beta}]$ (where τ is the relaxation time and $0<\beta\leq 1$ is the parameter describing the width of the distribution of relaxation times) which describes the approach of dielectric polarization to the equilibrium with time t.

The relaxation in highly disordered systems such as glasses or relaxors is of particular interest. In many cases it does not follow the classical Arrhenius behavior. Instead, on cooling τ tends to infinity at a non-zero temperature T_{τ} which can be considered as the freezing temperature below which the system becomes nonergodic.

The complex dielectric permittivity ($\varepsilon^{*} = \varepsilon' - i\varepsilon''$) of relaxor ferroelectric $PbMg_{1/3}Nb_{2/3}O_3$ single crystal and ceramics was measured in the frequency range 10^{-3}-10^{5} Hz and temperature range 20-300 K.

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SURFACE-INDUCED PIEZOMAGNETIC, PIEZOELECTRIC AND LINEAR MAGNETOELECTRIC EFFECTS

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Using symmetry theory we calculate how symmetry breaking inevitably present in the vicinity of any surface gives rise to surface piezomagnetic, piezoelectric and magnetoelectric effects.

Actually, for any spatially confined system the inversion center disappears in the surface normal direction and only the symmetry axes and planes normal to the surface are conserved. Thus the magnetic and space symmetry group should be reduced to one of its subgroup, consisting of the transformation matrices A_{ij}^S, which satisfy the relations $n_i A_{ij}^S n_j = 1$ where n_j are the components of the surface normal. As a result the surface piezoeffect tensors $d_{lpn}^{(Sm)}$ and $d_{lmn}^{(Se)}$ (existing even in a cubic symmetry lattice near the surface) should obey other transformation laws $d_{ijk}^{(Sm)} \equiv (-1)^{tr} \det(\mathbf{A}^S) A_{il}^S A_{jp}^S A_{kn}^S d_{lpn}^{(Sm)}$ and $d_{ijk}^{(Se)} \equiv A_{il}^S A_{jm}^S A_{kn}^S d_{lmn}^{(Se)}$. The transformation laws for the linear surface ME axial tensor are $\gamma_{ij}^S = (-1)^{tr} \det(\mathbf{A}^S) A_{ik}^S A_{il}^S \gamma_{kl}^S$ for nonzero components, i.e. the laws differ from the ones existing in the bulk material: $\widetilde{\gamma}_{ij} = (-1)^{tr} \det(\mathbf{A}) A_{ik} A_{il} \gamma_{kl}$. The analysis, similar to the one we performed for the piezomagnetic tensor, shows that the ME effect exists in nanosystems of ninety magnetic classes.

As a result piezomagnetism and strong size-dependent linear magnetoelectric coupling are predicted in nanomaterials, which are nonpiezomagnetic and/or nonpiezoelectric in the bulk, but belong to the one of the existing 90 bulk magnetic classes. Thus piezomagnetics, piezoelectrics and linear magnetoelectrics should appear even among nanomaterials like simple binary oxides EuO, CoO and Er_2O_3.

Since our predictions are based on the symmetry theory, we should underline that although the surface-induced symmetry lowering (usually connected with the reduction of coordination number) proclaims the possibility of the magnetic moments appearance at the surface, the microscopic physical mechanisms, which lead to the magnetism origin and determine its peculiarities should be manifold and thus a separate first principles based study is required in every concrete case.

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WEDNESDAY, SEPTEMBER 15, 2010

Oral presentations

THE ESTIMATION OF THE Mn ATOMS CHEMICAL BONDING IN $(Na_{(1-X)}Bi_X)(Nb_{(1-Y)}Mn_Y)O_3$ CERAMICS AND CHANGEOVER IN THE ELECTRICAL PROPERTIES

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The $(Na_{1-x}Bi_x)(Mn_yNb_{1-y})O_3$ ceramics were sintered, they can be assumed as materials composed of multiferroics $NaNbO_3$ and $BiMnO_3$. The content of elements was checked by electron probe microanalysis (EPMA). To analyze the chemical bonding of Mn atoms in the studied materials the x-ray absorption spectra were measured taking advantage of their atomic selectivity. The X-ray Absorption Near Edge Structure (XANES) measurements were carried out on the Mn $L_{3,2}$ edges. In the case of $L_{3,2}$ edges the 3d empty states of Mn atoms were probed. This analysis allowed distinguishing three groups of Mn atoms bonding in the analyzed samples. Manganese ions replace the Nb^{5+} ions in these ceramics. In all compound the Mn^{CV} state – named covalent bonding - around 644.5 ± 0.3 eV was found. The Mn^{+2} state was found also in the compounds without Bi co-doping. In case of introduction of Bi to the Na sublattice, the Mn^{+3} ion was formed with different proportion between Mn^{CV} and Mn^{+3} state. The marked dispersion in the electric properties of the $(Bi_xNa_{1-x})(Mn_yNb_{1-y})O_3$ ceramics series was found in the real part of the effective electric permittivity $\varepsilon'(T,f)$. It markedly increased with temperature, reaching high value $\sim 10^5$ for the high Bi and Mn content, at $f = 100$ Hz. The smearing and disappearance of the electric order phase transitions were deduced. The thermally activated behaviour of the electric conductivity dominated within high temperature range (900-950 K) and the adiabatic nearest-neighbour small polaron mechanism of conduction was more adequate within lower temperature ranges (above room temperature). The activation energy values of the electric conductivity were estimated. The obtained results were discussed assuming that the difference in the valence of the ions created the electric dipoles whereas the variation of the ionic radii resulted in additional elastic interaction.

NONLINEAR DIELECTRIC RESPONSE OF ROCHELLE SALT

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Rochelle salt (RS, sodium potassium tartrate tetrahydrate, $NaKC_4H_4O_6\times4H_2O$) is a material in which ferroelectric properties had been discovered for the first time [1]. RS exhibits two Curie points. The ferroelectric state with monoclinic point group 2 is confined to the temperature region between $T_{C1} \approx 255\,\mathrm{K}$ and $T_{C2} \approx 297\,\mathrm{K}$. While the upper Curie point is similar to regular ferroelectric phase transition, the lower one – the point where the spontaneous polarization is lost when cooling, and the system becomes paraelectric (disordered) again - is not trivial, since the RS crystal structure above the higher Curie point and below the lower Curie point is the same (orthorhombic point group 222). The linear, and the second- and third-order nonlinear susceptibilities were measured using our specialized susceptometer [2] in order to verify conclusions stemming from the theory of the ferroelectric effect in Rochelle salt proposed by Mitsui [3] and concerning the behavior of the susceptibilities in the vicinity of the phase transition points. Particular attention is focused on the nontrivial "inverse melting" point. Our experimental results well comply with the theoretical predictions.

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FORMATION OF STRIPE STRUCTURES WITH AND WITHOUT LONGER-RANGE INTERACTIONS

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Self-organized stripe structures are usually observed in two dimensional systems with competing short- and long-range interactions of electrostatic, magnetic or elastic origin. Stripes were experimentally found in very different (some of them, nanoscale) materials: binary liquids, surface-adsorbate systems, ferroelectrics, ferromagnetic and Langmuir films. The phase transition model describing the competition of short- and long-range forces is called dipolar Ising model [1]. The thermodynamics of this model has been studied both for square [1,2] and hexagonal [3,4] lattices. The width of stripe domains obtained in this model depends on a ratio of short- and long-range interactions. Here we introduce dipolar Ising model and present the results of Monte Carlo simulation of phase transitions from stripe phase to isotropic stripe phase on a hexagonal lattice. We discuss the obtained phase diagram and demonstrate the histogram method to determine the order of the phase transitions to stripe phases.

We also discuss the model with two interaction constants (nearest neighbor pair repulsion v_1 and trio in a line attraction v_t) which shows stripe domains formation on hexagonal lattice *without* longer range interactions usually required for stripes occurence. Stripe domains of the simplest stripe structure, (2x1), are obtained at low concentration of particles and $|v_t|/v_1$=1.0-1.3. Phase diagrams of this model are obtained by Monte Carlo simulation, and mixed (2x1)+gas phase stability is analyzed. We demonstrate that the (2x1)+gas phase can be the ground state structure at very low concentrations of particles, as well as the metastable structure characterized by large and very stable domains.

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SIZE-EFFECT IN FERROELECTRIC TGS, TGSe CRYSTALS UNDER HIGH HYDROSTATIC PRESSURE

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The dielectric parameters ε, T_C, P_S and E_C were investigated in TGS and TGSe crystals under high hydrostatic pressure. It has been found that in TGS above a critical pressure $p_c \approx 1.8$ GPa the derivative $\frac{dT_C}{dp}$ start to be negative and ferroelectricity disappear at about 2.5 GPa. The analysis of the reciprocal permittivity ε^{-1} points on relaxor-like behavior of TGS after the avalanche-type destruction of crystal structure on the critically-sized polar regions.

Lowest pressure for ferroelectricity vanishing is sufficient for TGSe crystals. Thus, high hydrostatic pressure can induce the structural inhomogenity even without compositional disorder.

SOME LESS KNOWN ASPECTS OF THE PARA-FERROELECTRIC PHASE TRANSITION IN FERROELECTRICS WITH HYDROGEN BONDS

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The nature of phase transition and the influence of an electric field not parallel to the ferroelectric axis in uniaxial ferroelectrics with hydrogen bonds belong to issues which are still investigated. As recently turned out, the transverse electric field can drastically change the critical (or tricritical) properties of such materials.

Although in some cases the phase transition in triglycine sulphate (TGS) can be approximately described by means of the Landau theory of the critical point, the results of experimental investigations concerning analogous phenomena in the structurally isomorphous triglycine selenate (TGSe) still remain difficult to specify. During the presentation, some evidence is shown for considerable deviations from the classic theory of phase transitions in TGSe. A possibility of the tricritical behavior is also discussed. According to the recent neutron diffraction experiments of R.R. Choundhury and R.Chitra [J.Phys.:Condensed Matter 21 (2009) 335901] distinct nature of phase transition in TGS and in TGSe is presented as a consequence of a difference in shape and height of the double well potential experienced by the amino group of GI glycine.

The influence of an electric field not parallel to the ferroelectric axis on dielectric properties of TGS, TGSe and Rochelle salt has been investigated for ten years. The recent experimental results suggest that the description of the phase transition should be expanded by an addition of a new parameter responsible for the properties of the crystal appearing due to its former exposure to the transverse electric field. Such a new freezing parameter can be considered equivalent to the other ones, such as polarization and temperature.

THURSDAY, SEPTEMBER 16, 2010

Oral presentations

ELECTRONIC PROPERTIES OF KDP AND DKDP CRYSTALS: *AB-INITIO* CALCULATIONS AND SPECTRAL ELLIPSOMETRY EXPERIMENT

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The crystals KH_2PO_4 (KDP) and KD_2PO_4 (DKDP) are well known and comprehensively studied ferroelectrics. However, the band structure and related electronic properties of these crystals in the photon energy range of valence electrons excitations, $E = 8…30$ eV, have been studied not enough. For example, the complex dielectric function $\varepsilon(E)$ in this range, as one of the basic electronic characteristics of a crystal, was unknown before (only the corresponding reflectance spectra $R(E)$ were studied). Also, experimental studies of the temperature dependence of complex dielectric function $\varepsilon(T)$ for this photon energy range and in the temperature range of ferroelectric phase transition are unknown. However, such knowledge, when is known, could help to understand better the microscopic origin of the phase transition in KDP-type crystals.

Results of *ab-initio* calculations of the band structure and complex dielectric function $\varepsilon(E)$ of KDP using VASP code are presented and discussed for the ferroelectric (sg $Fdd2$) and paraelectric (sg $F\bar{4}d2$) phases.

Experimental complex dielectric function $\varepsilon(E)$ of KDP and DKDP crystals have been obtained using the spectroscopic ellipsometry method and synchrotron radiation (BESSY II) in the photon energy range 5 to 17 eV. Reasonable agreement of the corresponding theoretical and experimental dependences has been found. Differences of the dependences $\varepsilon(E)$ for KDP and DKDP crystals have been explained with taking into consideration the nonadiabatic model of electronic excitations.

Temperature dependences of the complex dielectric function $\varepsilon(T)$ and the intensity of reflected light $I_R(T)$ for the constant photon energy $E = 9.5$ eV of KDP and DKDP crystals have been measured and discussed.

PHYSICAL PROPERTIES AND PHASE TRANSITIONS IN ADP EMBEDDED INTO THE POROUS GLASSES

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The results of investigation of physical properties of ADP-porous glasses composites with the average pores radius 160nm (ADP-PG160) and 23 nm (ADP-PG23) are presented. On the basis of calorimetric, dilatometric and dielectric measurements the negative size effect it was found. The influence of internal stress caused by the difference of thermal expansion coefficients of ADP nanoparticles and glass matrix and spontaneous deformations of nanocrystals at the phase transition temperature on thermal expansion, spontaneous deformation and phase transition temperatures are discussed. The low frequency dispersion in phase transition temperature region in ADP-PG composites it was found. At higher temperatures (above room temperature) the ionic conductivity it was observed. The activation energy of this process, calculated from the results of low frequency dielectric measurements are equal to 1.0 eV.

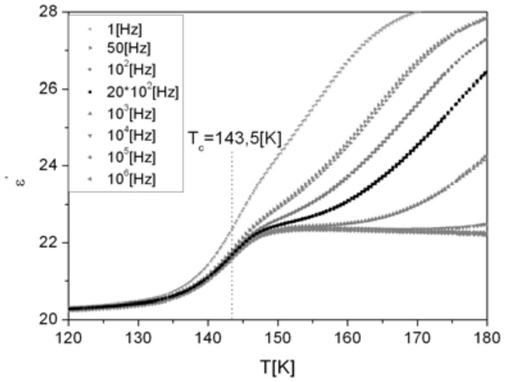

Fig. 1. Dielectric permittivity of ADP-PG160 composites as a function of temperature for several frequencies.

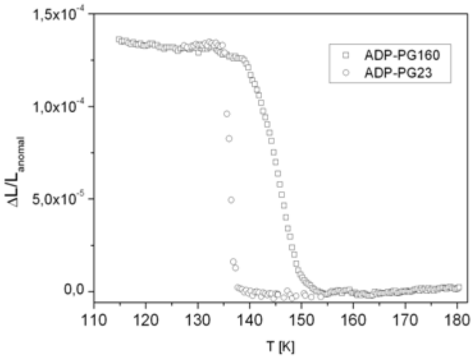

Fig. 2. Temperature dependence of the anomaly parts of relative thermal deformation *ΔL/L* of ADP-PG160 and ADP-PG23 as a function of temperature.

FERROELECTRIC AND FERROELASTIC CONTRIBUTIONS TO THE DIELECTRIC RESPONSE OF FERROELECTRICS

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Dielectric characterization is one of the most important tools to describe fundamental and practical properties of ferroelectrics. Displacive ferroelectrics are hybrid ferroics characterized by the presence of strongly coupled ferroelectric and ferroelastic domains and domain wall structures. Applications of ferroelectrics require necessarily the reorientation of the polarization through poling (for piezoelectrics, among others) or switching (for memories). The macroscopic polarization, dielectric and piezoelectric responses in poled polycrystalline ferroelectrics are strongly influenced by the relative orientation between neighbors domains, between domains and external electric field, and defects or impurities.

In this presentation it will be presented and discussed the influence of electric and mechanical (bidimensional mechanical stress) poling on the dielectric response of displacive ferroelectrics (PZT's). Experimental results are discussed considering the symmetry (rhombohedral -PZT 60/40 or tetragonal - PZT 40/60), measuring frequency (RF and MW) and temperature dependence of the dielectric permittivity. The influence of each poling is interpreted based on the piezoelectric contribution to the effective dielectric permittivity.

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STRUCTURAL PHASE TRANSITION IN PYRIDAZINE FLUOROBORATE

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Single crystals of pyridazine fluoroborate [$(C_4H_4N_2)HBF_4$] have been grown and characterized by X-ray diffraction, DSC, dielectric measurements and optical studies. At room temperature the crystal is monoclinic, space group $P2_1/n$. DSC measurement revealed the reversible phase transition at 333 and 327 K on heating and cooling, respectively. The temperature dependences of the electric permittivity exhibit anomalies characteristic for the first-order phase transitions at practically the same temperatures as those observed in DSC. Optical observations by means of polarizing microscope show that the crystal symmetry changes from the biaxial room temperature phase II to the optically uniaxial high temperature phase I. The orientation states domain arising after cooling from phase I reflects observed symmetry changes between monoclinic and hexagonal system at the phase transition. The crystal seems to be similar to $(C_4H_4N_2)HClO_4$.

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