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1. Main Research Results

- 1) Development of Lead Free Piezoelectric Material
Novel lead-free system with large piezoelectricity comparable to lead-based materials with high Curie temperature was developed.
- 2) Preparation of Perovskite Oxide at Low temperature
Low temperature preparation of lead free piezoelectric materials with good properties comparable to that prepared above 1000°C was succeed.
- 3) Growth of Epitaxial Nan rod allays
Allays of Nana rods with several 10 nm in width and over 1 μ m in length were successfully obtained.

2. The list of publications

Original articles

- 1) “Experimental evidence for orientation property of $\text{Pb}(\text{Zr}_{0.35}\text{Ti}_{0.65})\text{O}_3$ by manipulating polar axis angle using CaF_2 substrate”, Satoru Utsugi, Takashi Fujisawa, Yoshitaka Ehara, Tomoaki Yamada, Masaaki Matsushima, Hitoshi Morioka, and Hiroshi Funakubo, *Appl. Phys. Lett.*, 96 (2010)1029051-3.
- 2) “Determination Factors of Strain-relaxed Complex Domain Structure Observed in Thick Epitaxial $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$ Films”, Hiroshi Nakaki, Satoru Utsugi, Takashi Fujisawa, Mitsumasa Nakajima, Yoshitaka Ehara, Tomoaki Yamada, Hitoshi Morioka, Toshihiro Ifuku, and Hiroshi Funakubo, *Mater. Res. Soc. Symp. Proc.*, 1199 (2010) 1199-F08-08.
- 3) “Effect of deposition time on film thickness and their properties for hydrothermally-grown epitaxial KNbO_3 thick films”, Mutsuo Ishikawa, Hiro Einishi, Mitsumasa Nakajima, Tomohito Hasegawa, Takeshi Morita, Yoshifumi Saijo, Minoru Kurosawa, and Hiroshi Funakubo, *Jpn. J. Appl. Phys.*, 49 (2010) 07HF01-1-4.
- 4) “Orientation control of (001) and (101) in epitaxial tetragonal $\text{Pb}(\text{Zr},\text{Ti})\text{O}_3$ films with (100)/(001) and (110)/(101) mixture orientations”, Satoru Utsugi, Takashi Fujisawa, Yoshitaka Ehara, Tomoaki Yamada, Shintaro Yasui, Mohamed-Tahar Chentir, Hitoshi Morioka, Takashi Iijima, and Hiroshi Funakubo, *J. Ceram. Soc. Jpn.*, 118 [8] (2010)627-630.
- 5) “Composition dependence of crystal structure and electrical properties for epitaxial

films of $\text{Bi}(\text{Zn}_{1/2}\text{Ti}_{1/2})\text{O}_3$ - BiFeO_3 solid solution system”, Keisuke Yazawa, Shintaro Yasui, Hitoshi Morioka, Tomoaki Yamada, Hiroshi Uchida, Alexei Gruverman, and Hiroshi Funakubo, *J. Ceram. Soc. Jpn.*, 118[8] (2010) 659 – 663.

- 6) “Synchrotron X-ray Diffraction Study on a Single Nanowire of PX-Phase Lead Titanate”, Tomoaki Yamada, Jin Wang, Osami Sakata, Cosmin S. Sandu, Zhanbing He, Takafumi Kamo, Shintaro Yasui, Nava Setter, and Hiroshi Funakubo, *J. Euro. Ceram. Soc.*, 30 (2010) 3259–3262.
- 7) “Single crystal-like selection rules for unipolar-axis oriented tetragonal $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$ thick epitaxial films”, Mitsumasa Nakajima, Takashi Fujisawa, Yoshitaka Ehara, Tomoaki Yamada, Hiroshi Funakubo, Hiroshi Naganuma, Soichiro Okamura, Ken Nishida, Takashi Yamamoto, and Minoru Osada, *Appl. Phys. Lett.*, 97 (2010) 111901-1-3.
- 8) “Growth of polar axis -oriented tetragonal $\text{Pb}(\text{Zr},\text{Ti})\text{O}_3$ films on CaF_2 substrates with transparent $(\text{La}_{0.07}\text{Sr}_{0.93})\text{SnO}_3$ ”, Satoru Utsugi, Yoshitaka Ehara, Hidenori Tanaka, Tomoaki Yamada, Hiroshi Funakubo and Hiroshi Uchida, *J. Crystal Growth*, 312 (2010) 3127-3130.
- 9) “MOCVD growth and characterization of BiFeO_3 - $\text{Bi}(\text{Zn}_{1/2}\text{Ti}_{1/2})\text{O}_3$ ferroelectric films”, Keisuke Yazawa, Shintaro Yasui, Masaaki Matsushima, Hiroshi Uchida, and Hiroshi Funakubo, *Mater. Sci. Eng. B: Solid-State Mater. Adv. Tech.* 173(1-3) (2010) 14 – 17.
- 10) “Effect of film thickness and crystal orientation on the constituent phase in epitaxial BiFeO_3 - BiCoO_3 films grown on SrTiO_3 substrates”, Shintaro Yasui, Keisuke Yazawa, Tomoaki Yamada, Ken Nishida, Hiroshi Uchida, Masaki Azuma and Hiroshi Funakubo, *Jpn. J. Appl. Phys.*, 49(9) (2010) 09MB04-1-5.
- 11) “Structural Property and Electric Field Response of a Single Perovskite PbTiO_3 Nanowire Using Micro X-ray Beam”, Tomoaki Yamada, Jin Wang, Osami Sakata, Hidenori Tanaka, Yoshitaka Ehara, Shintaro Yasui, Nava Setter, and Hiroshi Funakubo, *Jpn. J. Appl. Phys.*, 49(9) (2010) 09MC09-1-4.
- 12) “In-situ Raman spectroscopy for characterization of the domain contributions to electrical and piezoelectric responses in $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$ films”, Mitsumasa Nakajima, Hiroshi Nakaki, Yoshitaka Ehara, Tomoaki Yamada, Ken Nishida, Takashi Yamamoto, Minoru Osada and Hiroshi Funakubo, *Appl. Phys. Lett.*, 97, (2010) 181907-1-3.
- 13) “Influence of Epitaxial Growth Orientation on Residual Strain and Dielectric Properties of $(\text{Ba}_{0.3}\text{Sr}_{0.7})\text{TiO}_3$ Films Grown on In-Plane Compressive Substrates”, T. Yamada,, T. Kamo, D. Su, T. Iijima, and H. Funakubo, *Ferroelectrics.*, 405

(2010)262–267.

- 14) “Spontaneous polarization estimation from the soft mode in strain-free epitaxial polar axis-oriented Pb(Zr, Ti)O₃ thick films with tetragonal symmetry”, Yoshitaka Ehara, Satoru Utsugi, Mitsumasa Nakajima, Tomoaki Yamada, Takashi Iijima, Hiroki Taniguchi, Mitsuru Itoh, and Hiroshi Funakubo, *Appl Phys Lett.*, 98 (2011) 141914 -1-3.

Collaborator

- 1) “Self-assembled ferroelectric-dielectric nanocomposite films for tunable applications”, T Yamada, C S Sandu, M Gureev, A K Tagantsev, P Muralt, H Funakubo, and N Setter, *Mater. Sci. Eng.*, 8 (2010)012010-1-7.
- 2) “Large constriction of lattice constant in epitaxial magnesium oxide thin film: Effect of point defects on lattice constant”, Satoru Kaneko, Takatoshi Nagano, Kensuke Akiyama, Takeshi Ito, Manabu Yasui, Yasuo Hirabayashi, Hiroshi Funakubo, and Mamoru Yoshimoto, *J. Appl.Phys.*, 107 (2010) 073523-1-3.
- 3) “Dielectric Properties of Highly (001)-plane Oriented SrBi₄Ti₄O₁₅ Thin Films”, Yuki Mizutani, Hiroshi Uchida, Hiroshi Funakubo, and Seiichiro Koda, *Key Eng. Mater.*, 445 (2010) 131-134.
- 4) “Comparison of BST film microwave tunable devices based on (100) and (111) MgO substrates”, Minoru Noda, Tomoaki Yamada, Kousuke Seki, Takashi Kamo, Kaoru Yamashita, Hiroshi Funakubo, and Masanori Okuyama, *IEEE Transactions on Ultrasonics, Ferroelectrics, and Frequency Control*, 55(10) (2010) 2221-2227.
- 5) “Geometric Phase Analysis of Nano-Scale Strain Fields Around 90° Domains in PbTiO₃/SrTiO₃ Epitaxial Thin Film”, Takanori Kiguchi, Kenta Aoyagi, Toyohiko J. Konno, Satoru Utsugi, Tomoaki Yamada, and Hiroshi Funakubo, *Mater. Res. Soc. Symp. Proc.*, 1199(2010) 1199-F09-08.
- 6) “Process-dependent coercive fields in undoped and Mn-doped BiFeO₃ films formed on SrRuO₃/Pt(111) electrodes by rf sputtering”, Jeong Hwan Kim, Hiroshi Funakubo, Yoshihiro Sugiyama, and Hiroshi Ishiwara, *Mater. Res. Soc. Symp. Proc.*, 1199 (2010) 1199-F06-33.
- 7) “In-situ lattice-strain analysis of a ferroelectric thin film under an applied pulse electric field”, O. Sakata, S. Yasui, T. Yamada, M. Yabashi, S. Kimura, and H. Funakubo, *AIP Conf. Proc.* , 1234(2010)151155.
- 8) “One-axis Oriented CaBi₄Ti₄O₁₅ and SrBi₄Ti₄O₁₅ Films Prepared on Silicon Wafer by Chemical Solution Deposition Technique”, Yuki Mizutani, Hiroshi

- Uchida, Hiroshi Funakubo, and Seiichiro Koda, *Mater. Res. Soc. Symp. Proc.*, 1199 (2010) 1199-F03-14.
- 9) “In-situ Observation of a MEMS-based $\text{Pb}(\text{Zr,Ti})\text{O}_3$ Micro Cantilever Using Micro-Raman Spectroscopy”, Masamichi Nishide, Maresuke Kuzuhara, Takeshi Tai, Takashi Katoda, Hitoshi Morioka, Hiroshi Funakubo, Ken Nishida, and Takashi Yamamoto, *J. Ceram. Soc. Jpn.*, 118[8] (2010) 644 – 647.
 - 10) “Piezoelectric Anomalies at the Ferroelastic Phase Transitions of Lead-Free Tungsten Bronze Ferroelectrics”, Takayuki Watanabe, Jumpei Hayashi, Takanori Matsuda, Toshihiro Ifuku, Bong-Yeon Lee, Takashi Iijima, Hiroshi Funakubo, Houzhona Yu and Nobuhiro Kumada, *J. Ceram. Soc. Jpn.*, 118 [8] (2010) 717-721.
 - 11) “Large Lattice Misfit on Epitaxial Thin Film: Coincidence Site Lattice Expanded on Polar Coordinate System”, Satoru Kaneko, Kensuke Akiyama, Takeshi Ito, Manabu Yasui, Masayasu Soga, Yasuo Hirabayashi, Hiroshi Funakubo, and Mamoru Yoshimoto, *Jpn. J. Appl. Phys.* 49 (2010) 08JE02-1-3.
 - 12) “A-Site-Modified Perovskite Nanosheets and Their Integration into High- K Dielectric Thin Films with a Clean Interface”, Bao-Wen Li, Minoru Osada, Tadashi C. Ozawa, Kosho Akatsuka, Yasuo Ebina, Renzi Ma, Kanta Ono, Hiroshi Funakubo, and Takayoshi Sasaki, *Jpn. J. Appl. Phys.*, 49(9) (2010) 09MA01-1-5.
 - 13) “Crystal Structure and Dielectric Property of Bismuth Layer-Structured Dielectric Films with c -axis Preferential Crystal Orientation”, Yuki Mizutani, Takanori Kiguchi, Toyohiko J. Konno, Hiroshi Funakubo and Hiroshi Uchida, *Jpn. J. Appl. Phys.*, 49(9) (2010) 09MA02-1-6.
 - 14) “Effect of Grain Size on Mechanical Properties of Full-Dense $\text{Pb}(\text{Zr,Ti})\text{O}_3$ Ceramics”, Tetsu Miyoshi and Hiroshi Funakubo, *Jpn. J. Appl. Phys.*, 49(9) (2010) 09MD13-1-6.
 - 15) “Electronic, Structural, and Piezoelectric Properties of $\text{BiFe}_{1-x}\text{Co}_x\text{O}_3$ ”, Kaoru Miura, Makoto Kubota, Masaki Azuma and Hiroshi Funakubo, *Jpn. J. Appl. Phys.*, 49(9) (2010) 09ME07-1-4.
 - 16) “Robust High- κ Response in Molecularly-Thin Perovskite Nanosheets”, Minoru Osada, Kosho Akatsuka, Yasuo Ebina, Hiroshi Funakubo, Kanta Ono, Kazunori Takada, and Takayoshi Sasaki, *ACS Nano*. 4(9) (2010) 5225–5232.
 - 17) “Electrical properties and x-ray photoelectron spectroscopy studies of $\text{Bi}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ doped $\text{Pb}(\text{Zr}_{0.4}\text{Ti}_{0.6})\text{O}_3$ thin films”, M. H. Tang, J. Zhang, X. L. Xu, H. Funakubo, Y. Sugiyama, H. Ishiwara, and J. Li, *J. Appl. Phys.*, 108, 084101-1-5 (2010).

- 18) “Antiferrodistortive Structural Phase Transition in Compressively-strained Epitaxial SrTiO₃ Film Grown on (La, Sr)(Al, Ta)O₃ Substrate”, T.Yamada, T.Kiguchi, A.K.Tagantsev, H.Morioka, T.Iijima, H.Ohsumi, S.Kimura, M.Osada, N.Setter, and H.Funakubo, *Integ. Ferroele.*, 115, 57 (2010).
- 19) “Engineered Interfaces of Artificial Perovskite Oxide Superlattices via Nanosheet Deposition Process”, Bao-Wen Li, Minoru Osada, Tadashi C. Ozawa, Yasuo Ebina, Kosho Akatsuka, Renzhi Ma, Hiroshi Funakubo, and Takayoshi Sasaki. *ACS Nano*, 4 (11)(2010) 6673–6680.
- 20) “Epitaxial Growth of (100)-oriented □-FeSi₂ Film on 3C-SiC(100) plane”, Kensuke Akiyama, Teiko Kadowaki, Yasuo Hirabayashi, Mamoru Yoshimoto, Hiroshi Funakubo and Satoru Kaneko, *J. Crystal Growth* 316 (2011) 10-14.
- 21) “Identification of crystal symmetry of Bi₄Ti₃O₁₂ epitaxial thin films with (100)/(010) orientations by a modified method of synchrotron-based reciprocal-space mapping”, Osami Sakata, Takayuki Watanabe and Hiroshi Funakubo, *J. Appl. Cryst.* 44 (2011) 385–391.
- 22) “Nano-strip grating lines self-organized by a high speed scanning CW laser”, Satoru Kaneko, Takeshi Ito, Kensuke Akiyama, Manabu Yasui, Chihiro Kato, Satomi Tanaka, Yasuo Hirabayashi, Akira Mastuno, Takashi Nire, Hiroshi Funakubo, and Mamoru Yoshimoto, *Nanotechnology* 22 (2011) 175307-1-6.
- 23) “Incubation Time Free CVD-TiO₂ Film Preparation Using Novel Precursor of Ti-DOT”, Hirokazu Chiba, Ken-ichi Tada, Toshiki Yamamoto, Kohei Iwanaga, Atsushi Maniwa, Tadahiro Yotsuya, Noriaki Oshima, and Hiroshi Funakubo, *Mater. Res. Soc. Symp. Proc.* 1288 (2011) mrsf10-1288-g06-38.
- 24) “Enhancement of magnetization at morphotropic phase boundary in epitaxial BiCoO₃-BiFeO₃ solid solution films grown on SrTiO₃ (100) substrates”, Hiroshi Naganuma, Shintaro Yasui, Ken Nishida, Takashi Iijima, Hiroshi Funakubo, and Soichiro Okamura, *J. Appl. Phys.*, 109, (2011) 07D917-1-4.

Review

- 1) “Chemical vapor deposition of ferroelectric thin films: a critical review”, Hiroshi Funakubo, Shintaro Yasui, Mutsuo Ishikawa, and Tomoaki Yamada, “Ferroelectric Thin Films at Microwave Frequencies”. edited by TK Jackson, PM Suherman and P bao, *Research Signpost, India*(2010)172-182.
- 2) “Electronic and Structural Properties of ABO₃ : Role of the B-O Coulomb Repulsions for Ferroelectricity”, Kaoru Miura, Masaki Azuma and Hiroshi Funakubo, *Materials*, 4 (2011) 260-273.

4. Invited presentation in International/domestic conference

(4-1) International conference

- 1) H. Funakubo, M. Nakajima, H. Nakaki, T. Yamada and H. Miroka, "Enhancement of piezoelectric response in tetragonal Pb(Zr, Ti)O₃ thick films by 90°-domain wall density control", 2010 U.S. Navy Workshop on Acoustic Transduction Materials and Devices, The Penn Stater Conference Hotel, State College, PA, May 12, 2010, V.1. [Invited only]
- 2) H. Funakubo, K. Yazawa, S. Yasui, M. Matsushima, H. Uchida, T. Iijima, J. Wang, Y. Lu, H. Morioka, T. Yamamoto, and Y. Ikuhara, "Growth of Tetragonal Ferroelectric Films with More Than Three Times Larger Tetragonal Distortion Than PbTiO₃", 7th Asian Meeting on Ferroelectricity and 7th Asian Meeting on Electro Ceramics, June 28th (Monday) ~ July 1st (Thursday), 2010, Ramada Plaza Hotel, Jeju island, Korea, 1a-1-11 [Invited]
- 3) Hiroshi Funakubo, Shintaro Yasui, Kesiuke Yazawa, Masaki Matsushima and Tomoaki Yamada, Hitoshi Morioka, Osami Sakata, Hiroshi Uchida, Takashi Iijima, Junling Wang and You Lu, Takashi Yamamoto, and Yuichi Ikuhara, "Growth of Bi-based tetragonal ferroelectric thin films and their electrical properties:", Materials Science & Technology 2010 Conference & Exhibition, (MS&Y 10), Oct. 18th, 2010, Huston, USA, p.100 [Invited]
- 4) Mutsuo Ishikawa, Hiro Einishi, Tomohito Hasegawa, Takeshi Morita, and Minoru Kurosawa, and Hiroshi Funakubo, Low temperature synthesis of epitaxial KNbO₃ thick films grown by hydrothermal Method PZT-BZT materials, 3rd International Congress on Ceramics (ICC3), Nov. 14-18, 2010, Osaka, Japan, S6-045 [Invited]

5. Others

(5-1) Patent

Total 5 (Japan, 2010)

(5-2) Award

Total 2 including

- 1) Shintaro Yasui, Yoshitaka Ehara, Hiroshi Funakubo, (technical Photo Awards of Ceramics society of Japan 2010)
- 2) Shintaro Yasui, Best Presentation Awards of Doctor Thesis.